Supporting Information

Carbon Nanoarchitectures by Design:

Preorganizing Squaric Acid with Urea

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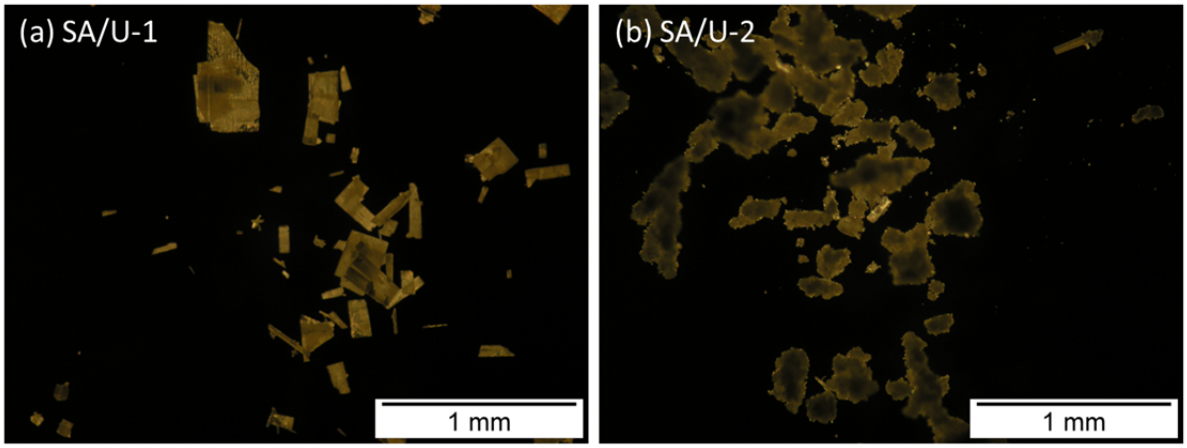

Figure SI-1: Optical microscope images of the primary supramolecular crystals formed from squaric acid and urea in the ratios (a) 1:1 (SA/U-1) and (b) 1:2 (SA/U-2).

Table SI-1: Assignement of the discussed vibrational peaks for the FT-IR-spectra of urea, squaric acid and the complexes SA/U-1 and SA/U-2.

Peak position				Assignement
Urea	**Squaric acid**	**SA/U-1**	**SA/U-2**	
	631 cm^{-1}			ring breathing modes[23]
	721 cm^{-1}			
	847 cm^{-1}			
716 cm^{-1}				$\delta(NH_2)$[22]
788 cm^{-1}				$\delta(OCNN)$[22]
1150 cm^{-1}		1144 cm^{-1}	1129 cm^{-1}	$\rho(NH_2)$[26]
1460 cm^{-1}		1465 cm^{-1}		$\nu(CN)$[26]
	1497 cm^{-1}			$\nu(C=C)$[23]
		1511 cm^{-1}, 1560 cm^{-1}	1560 cm^{-1}	$\nu(CC)$ + $\nu(CO)$[28,29]
1590 cm^{-1}		1627 cm^{-1}		$\nu(CO)$[26]
	1643 cm^{-1}	1634 cm^{-1}		$\nu(CO)$[23]
1674 cm^{-1}		1690 cm^{-1}		$\delta(NH_2)$[22]
	1804 cm^{-1}	1810 cm^{-1}		$\nu(CO)$[23]
		2780 cm^{-1}	2764 cm^{-1}	$\nu(NH_3^+)$[26]
		3156 cm^{-1}	3144 cm^{-1}	
3426 cm^{-1}		3464 cm^{-1}		$\nu(NH_2)$[22]

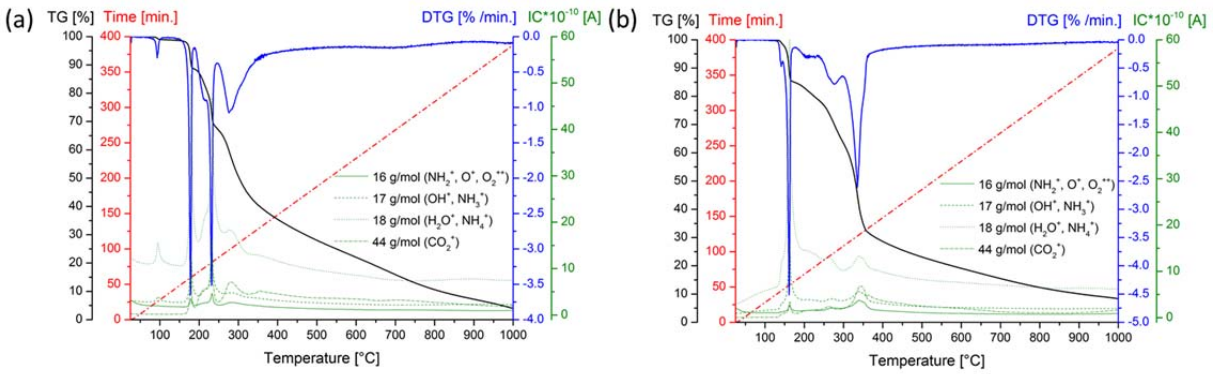

Figure SI-2: TGA-MS measurement of the samples (a) SA/U-1 and (b) SA/U-2.

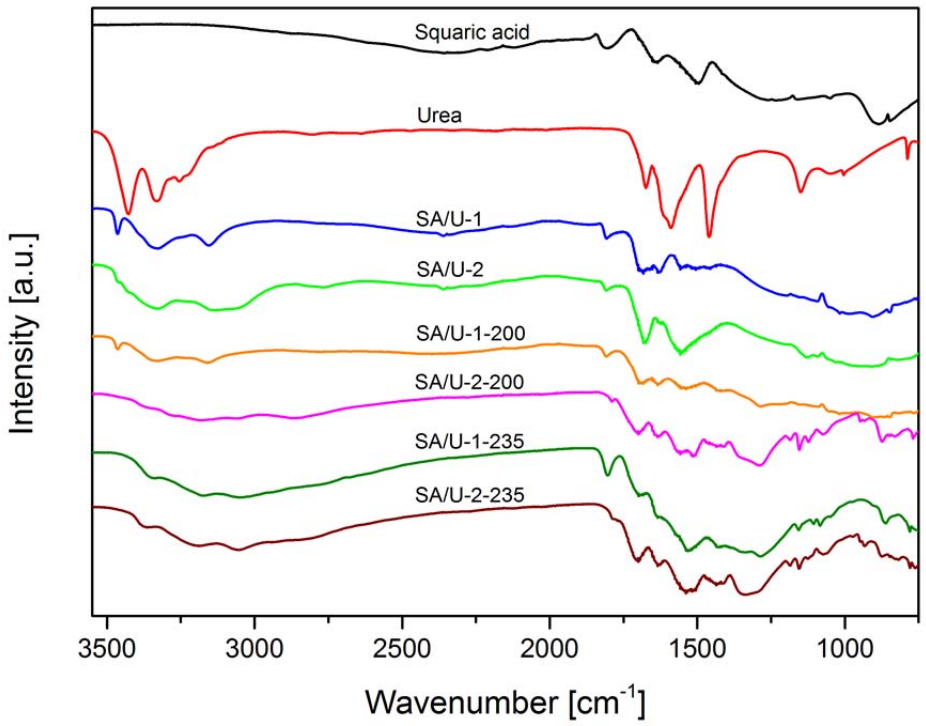

Figure SI-3:FT-IR spectra of the SA/U-complexes after thermal treatment at 200 °C (SA/U-1-200 and SA/U-2-200) and 235 °C (SA/U-1-235 and SA/U-2-235). For comparison, the spectra of squaric acid, urea and the complexes SA/U-1 and SA/U-2 are also shown.

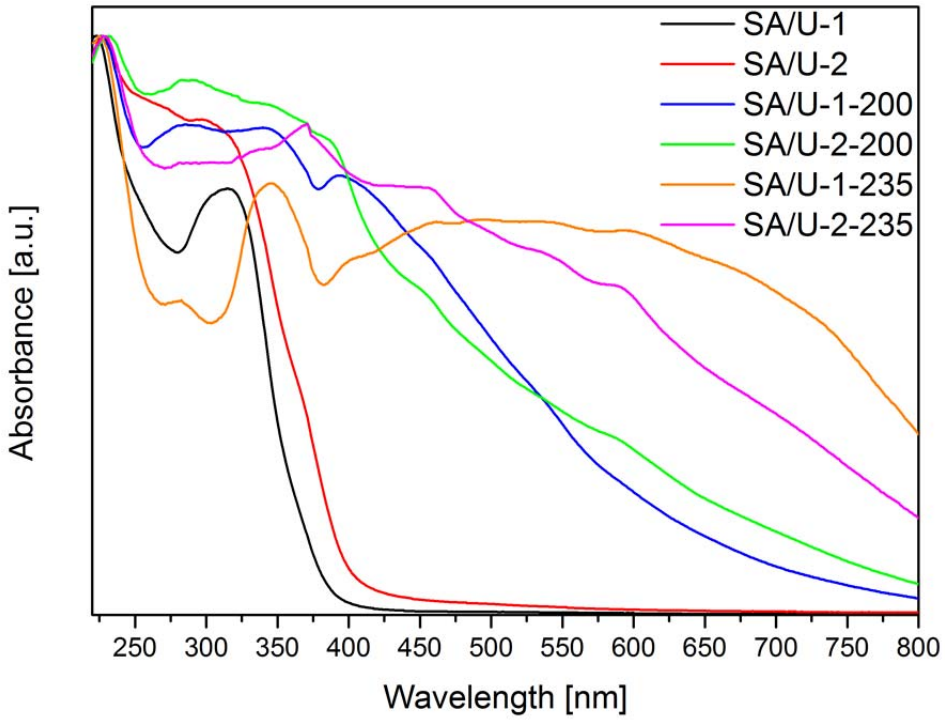

Figure SI-4: UV-Vis absorption spectra of the complex crystals (SA/U-1 and SA/U-2) and the complexes after thermal treatment at 200 °C (SA/U-1-200 and SA/U-2-200) and 235 °C (SA/U-1-235 and SA/U-2-235).

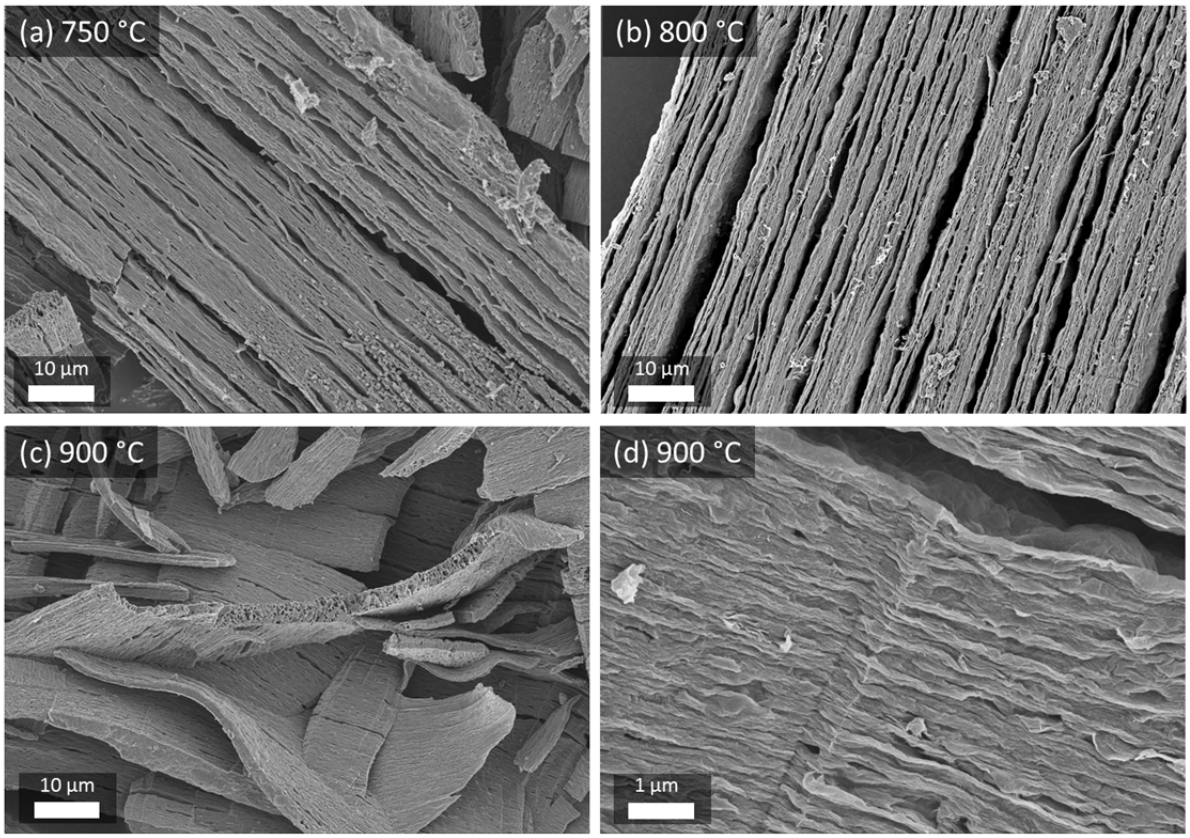

Figure SI-5: SEM-pictures of the 1:1 crystal after thermal condensation at (a) 750 °C (SA/U-1-750), (b) 800 °C (SA/U-1-800), (c) and (d) 900 °C (SA/U-1-900).

Table SI-2: Elemental composition of the samples SA/U-r-t and the yields for their corresponding thermal condensation processes. *The denoted yields for the samples SA/U-1 and SA/U-2 refer to the crystallization process.

Sample:	C [wt%]:	N [wt%]:	H [wt%]:	C/N-ratio (molar ratio):	Yield (thermal condensation):
SA/U-1	34	15	4	1.9	68 %*
SA/U-2	31	23	4	1.1	68 %*
SA/U-1-150	35	15	3	2.0	95 %
SA/U-2-150	33	25	4	1.1	89 %
SA/U-1-200	40	19	3	1.8	93 %
SA/U-2-200	35	26	4	1.2	83 %
SA/U-1-550	62	26	2	2.1	31 %
SA/U-2-550	61	27	2	2.0	24 %
SA/U-1-750	72	22	1	2.7	24 %
SA/U-2-750	72	22	1	2.7	18 %
SA/U-1-800	75	21	1	3.0	22 %
SA/U-2-800	75	21	1	3.0	16 %
SA/U-1-900	80	18	1	3.8	20 %
SA/U-2-900	81	17	1	4.1	15 %

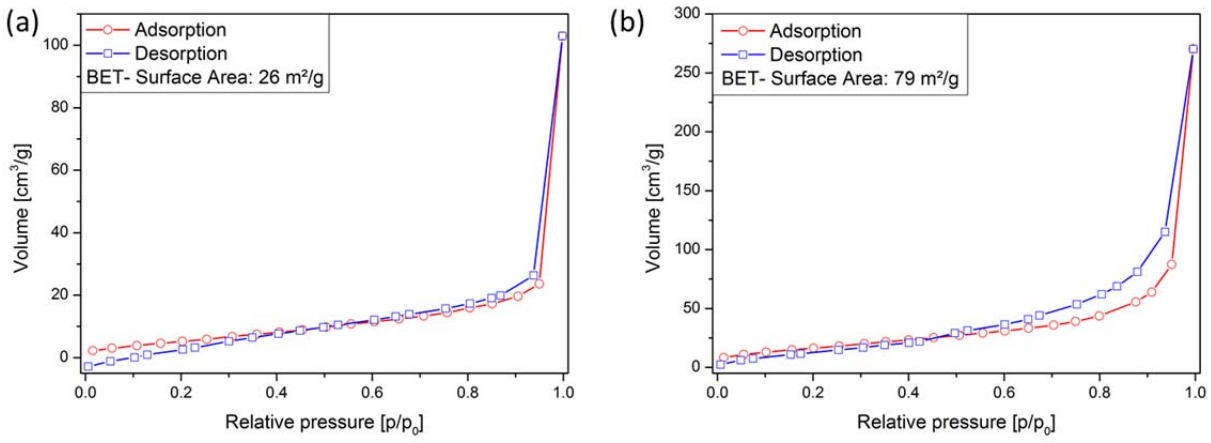

Figure SI-6: Nitrogen sorption isotherms and specific BET-surface area of the samples (a) SA/U-1-550 and (b) SA/U-2-550.

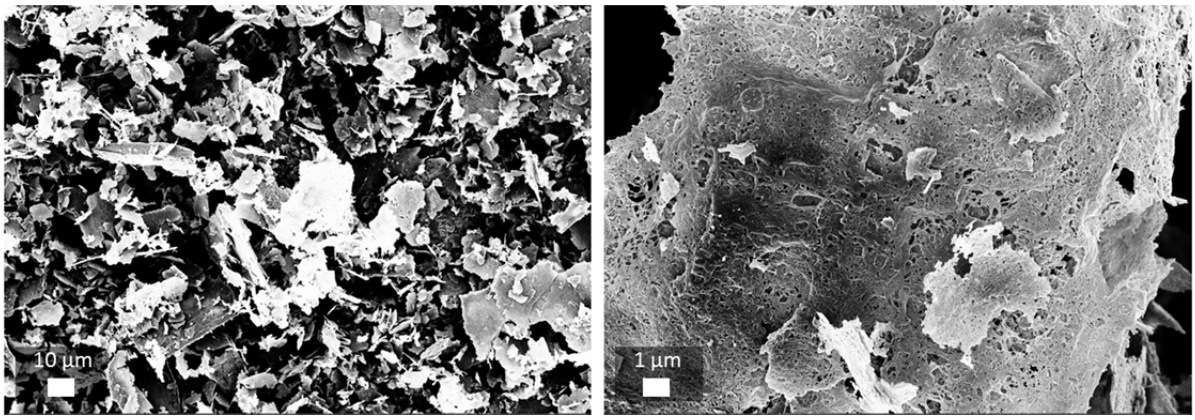

Figure SI-7: SEM-pictures of squaric acid, after thermal treatment at 550 °C for 1h (heating rate 2.5 K/min).