Boron-Nitride/Carbon-Nanotube Hybrid Aerogels as Multifunctional Desulfurisation Agents

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Figure S1. Digital images of BN/rCNT aerogel adsorbents with 2.5 wt% and 10 wt% BN doping amounts.



Figure S2. TGA in air of the BN powder adsorbent and the BN/rCNT aerogel adsorbent.



Figure S3. Raman spectra of BN/CNT aerogel adsorbent before and after thermal reduction.



Figure S4. XPS full spectra of BN powder and BN/CNT aerogel adsorbents before and after DBT adsorption.



Figure S5. SEM image and EDX mappings of BN/rCNT aerogel.



Figure S6. TEM images of un-hybridised BN mesostructures: (a) low-resolution TEM image of a thin BN structure; (b) higherresolution TEM of a thin corrugated edge of one of the nanostructures. Red circles indicate pores with diameters < 5nm.



Figure S7. TEM images of BN/rCNT aerogel fragment. (a) Low-resolution TEM image. The red arrows indicate the BN mesostrcutures, the blue arrows indicate the rCNT network. (b) Higher resolution TEM image of the rCNT network.



Figure S8. DBT adsorption onto BN powder at different temperatures (DBT in n-octane, initial DBT concentration 500 ppm S).

Table S1. Density, electrical conductivity and DBT sorption capacity of BN/rCNT aerogel under 800 °C and 1000 °C treatments.

Sample name	Temp	Density	Electrical conductivity	I _G /I _D	DBT capacity	
	(°C)	(mg/cm ³)	(S/m)		(mg·S/g BN)	
BN/rCNT aerogel	800	25.43	1.47	0.78	13.4	
BN/rCNT aerogel	1000	15.85	5.92	0.86	32	



Figure S9. (a) Joule heating performance and (b) Raman spectra of BN/rCNT thermally treated at 800 °C and 1000 °C.

In terms of functional desulfurization performance, the lowering of the thermal conversion temperature during materials synthesis leads to a significant decrease in desulfursation capacity (as measured by DBT sorption from n-octane onto the BN/rCNT aerogels) data. This might be related to incomplete BN precursor conversion and/or changes in aerogel porosity. In terms of electrical conductivity, one of the key property investigated in our study, the lowering of the thermal conversion temperature leads to significant decrease in electrical conductivity (see Table above). As a consequence the Joule-heating regeneration process, investigated in the second part of our manuscript, becomes less efficient. Specifically, the BN/rCNT aerogels produced at 800 °C require larger electrical power inputs compared to the materials produced at 1000 °C to reach the same Joule heating temperature (see panel (a) in Figure below). Both reduced electrical conductivity and reduced Joule-heating efficiency are likely associated with the lower degree of CNT graphiticity of the sample produced at the lower temperature. This is confirmed by Raman spectroscopy which shows a clear reduction in IG/ID ratio for the material produced at 800 oC (see Figure S9).



Figure S10. DBT uptake capacities for rCNT aerogel, BN powder and BN/rCNT aerogel (DBT in n-octane, initial DBT concentration 500 ppm S).

The adsorption isotherms confirm that, for all initial DBT concentrations tested, organosulfur uptake onto the BN/rCNT aerogels was greater than onto pure, unsupported BN. Both isotherms fitted very well a conventional Langmuir model ($R^2 > 0.97$, see also table below).

$$q_e = \frac{q_m C_e K_L}{1 + C_e K_L}$$

Where q_e is the DBT concentration at equilibrium, q_m is the maximum DBT uptake capacity (mg·S/g), and K_L is related to the energy of adsorption (L/mg).

	Langmuir model				
Sample	$q_{ m m}$	KL	2م		
	(mg·S/g)	(L/mg)	Λ		
BN powder	29.6	5.7·10 ⁻³	0.974		
BN/rCNT aerogel	45.2	6.6·10 ⁻³	0.993		

Table S2. Isotherm fitting parameters for DBT sorption onto BN powder and BN/rCNT aerogel



Figure S11. IR spectra of BN and BN/rCNT aerogel adsorbents after DBT adsorption.



Figure S12. XPS spectra of BN powder before and after DBT adsorption. (a) C1s, (b) O1s and (c) N1s spectra of BN powder before and after DBT adsorption.



Figure S13. a) Chemical structures and abbreviation of inorganic sulfur compound and organic aromatic sulfur compounds. b) Adsorption desulfurization performance of various sulfur-containing onto BN/rCNT aerogels (sorption from n-octane; initial concentration for all compounds 500 ppm S).



Figure S14. Joule-heating temperature versus electrical conductivity of the produced BN/rCNT aerogel adsorbent.



Figure S15. Voltage and temperature control of BN/rCNT aerogel under Joule-heating. (a) Preconditioning of BN/rCNT aerogel at around 200 °C for 20 minutes to remove impurities (e.g. water and gases). (b) Step-wise Joule-heating.



Figure S16. Joule-heating of BN/rCNT aerogel in air atmosphere.



Figure S17. (a) Heating curves and (b) cooling curves of post-adsorption BN/rCNT aerogel.

	Physical parameters						
Aerogel	т	I	U	R	Р	Q	
	(°C)	(A)	(V)	(Ω)	(W)	(kW∙h)	
rBN/CNT [1st]	427	1.45	11.60	8.0	16.82	0.017	
rBN/CNT [2 ^{ed}]	434	1.55	12.53	8.1	19.43	0.019	
rBN/CNT [3rd]	410	1.30	13.52	10.4	17.58	0.018	
rBN/CNT [4 th]	435	1.47	10.96	7.5	16.11	0.016	
rBN/CNT [5 th]	481	1.80	11.23	6.2	20.21	0.020	
Average	438	1.52	11.97	8.1	18.03	0.018	

Table S3. Parameters of the Joule heated post-adsorption BN/rCNT aerogel, including temperature (T), electrical current (I),voltage (U), resistance (R), input electrical power (P), energy consumption (Q).