

## *Electronic Supplementary Information*

# **Evolution of Porosity in Carbide-Derived Carbon Aerogels for Efficient Carbon Dioxide Adsorption**

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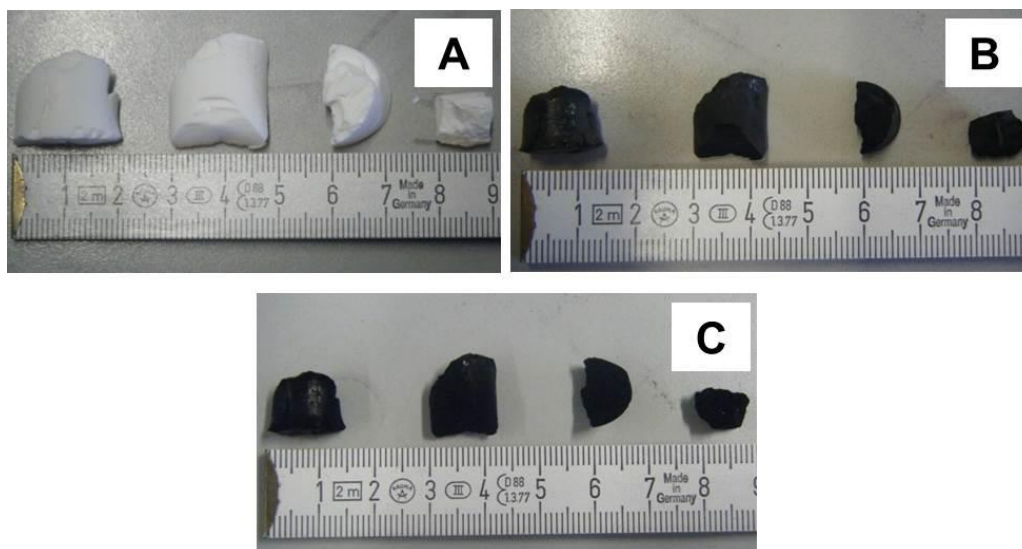
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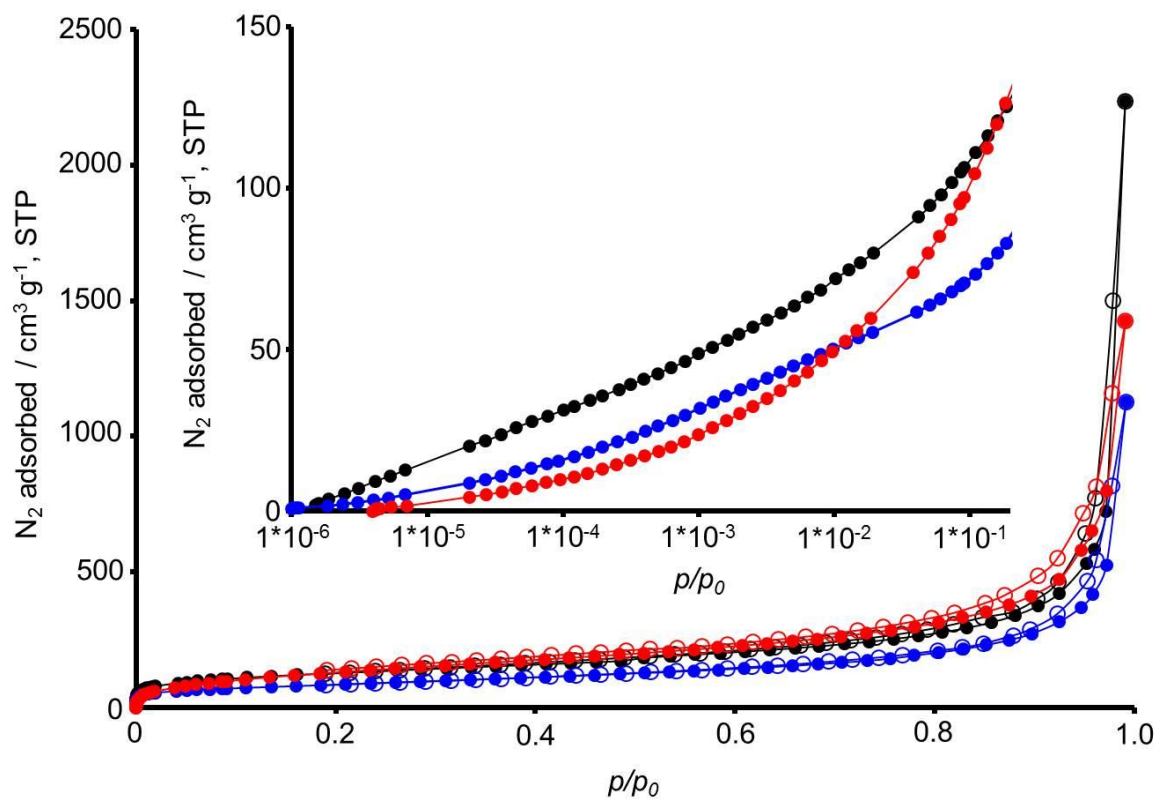
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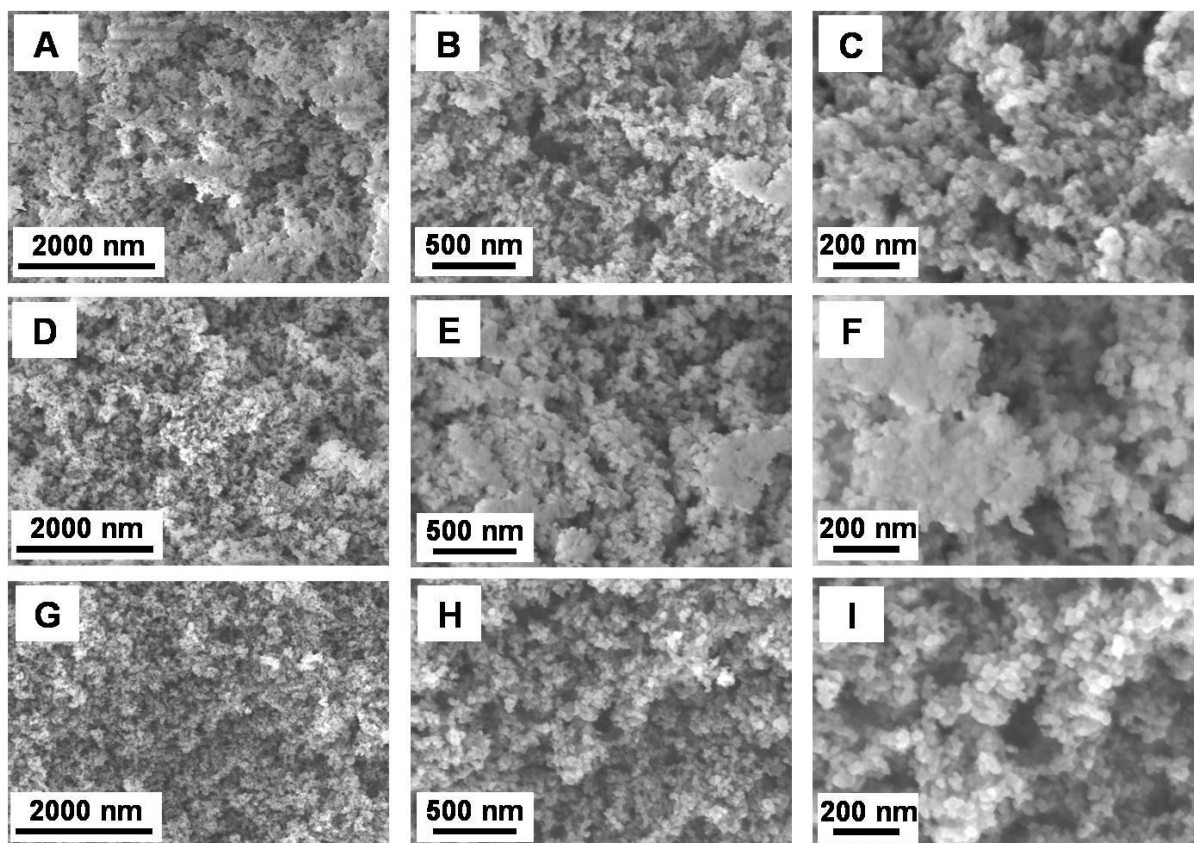
# Figures



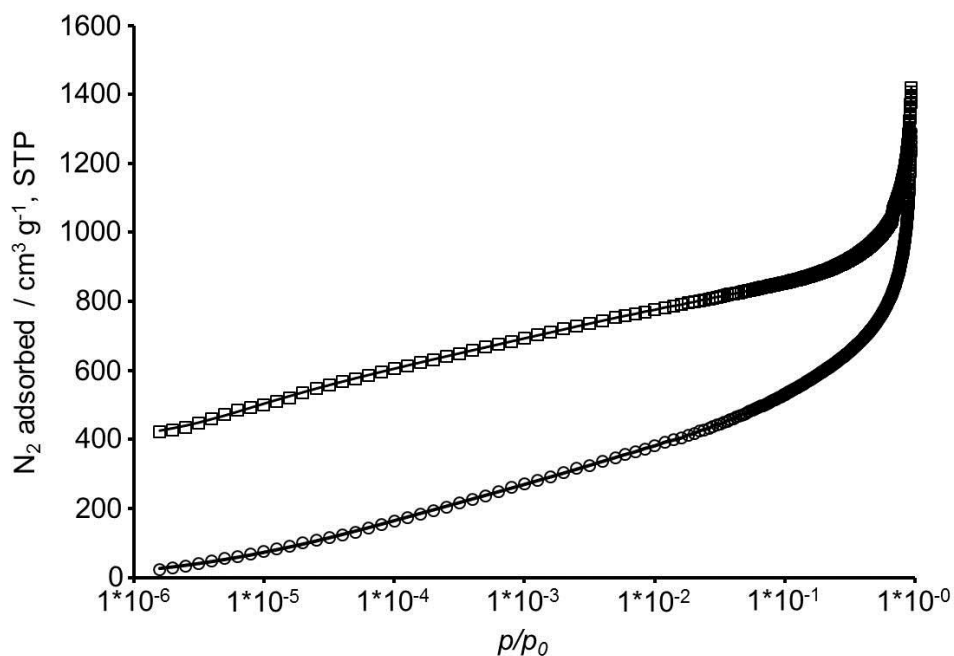
**Figure S1:** Photographs of (A) the PCS aerogel monoliths, (B) SiC-aerogel 700°C monoliths and (C) CDC aerogel 700°C monoliths in different synthesis stages.



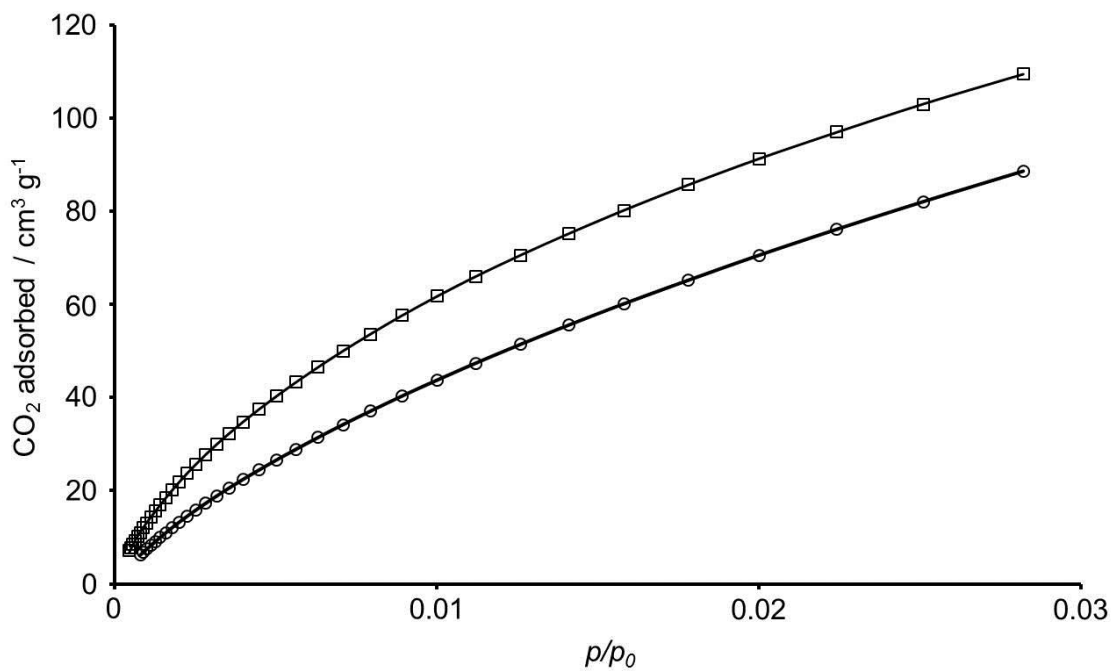
**Fig. S2:** Linear and semi-logarithmically (low pressure region; inset) plotted nitrogen adsorption/desorption (filled symbols/empty symbols) isotherms (-196°C) of the PCS aerogel (red) and the SiC aerogels prepared at 1000°C/700°C (blue/black).



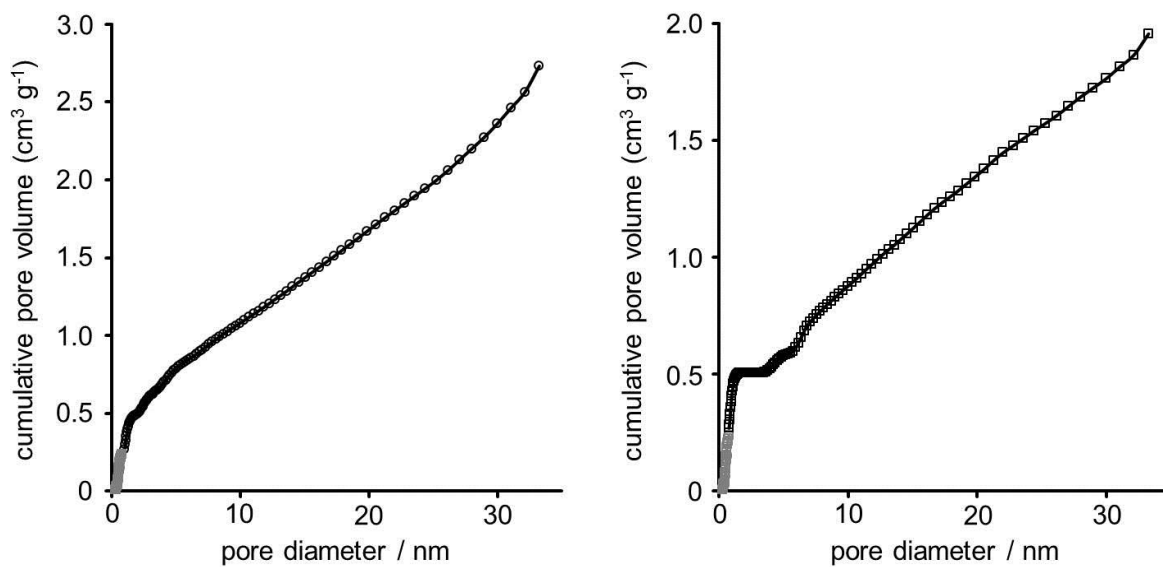
**Fig. S3:** Scanning electron microscopy images of (A-C) the PCS aerogels, (D-F) the SiC aerogels prepared at 700°C, and (G-I) the SiC aerogels prepared at 1000°C.



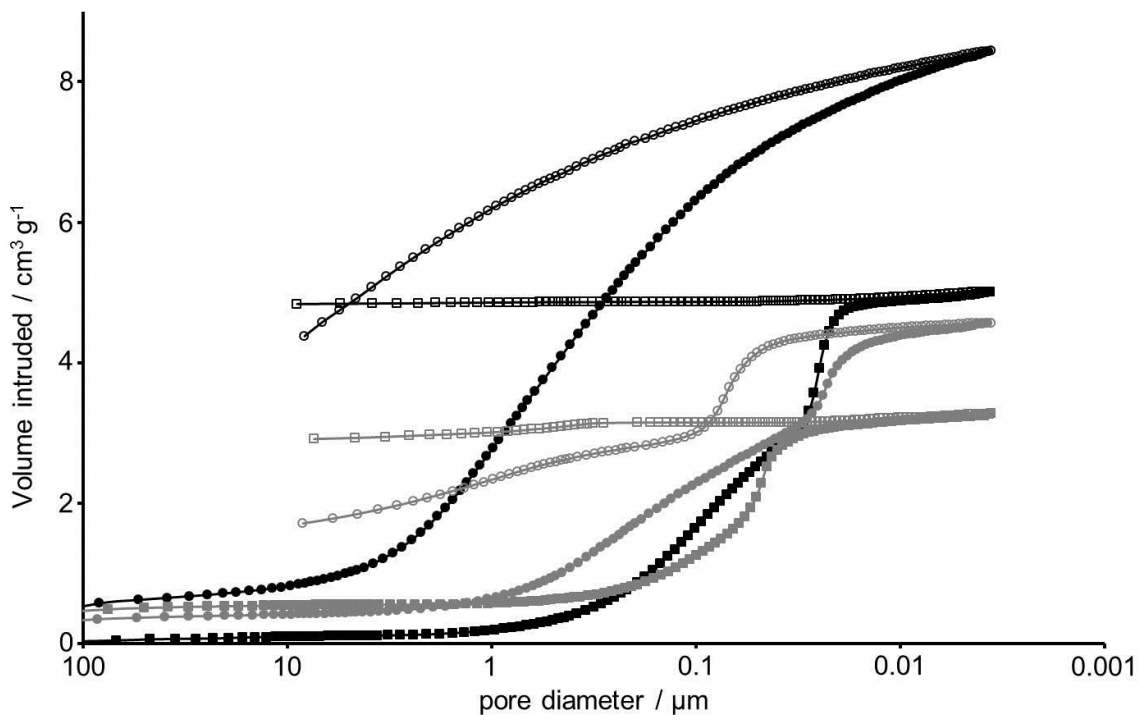
**Fig. S4:** QSDFT fits between the used nitrogen on carbon (slit/cylindrical pores) adsorption branch kernel (solid lines) and the experimental data (nitrogen physisorption at -196°C, empty symbols) for DUT-85 prepared at 1000°C/700°C (squares/circles). The data of DUT-85 1000°C is vertically offset by 400 cm<sup>3</sup> g<sup>-1</sup>.



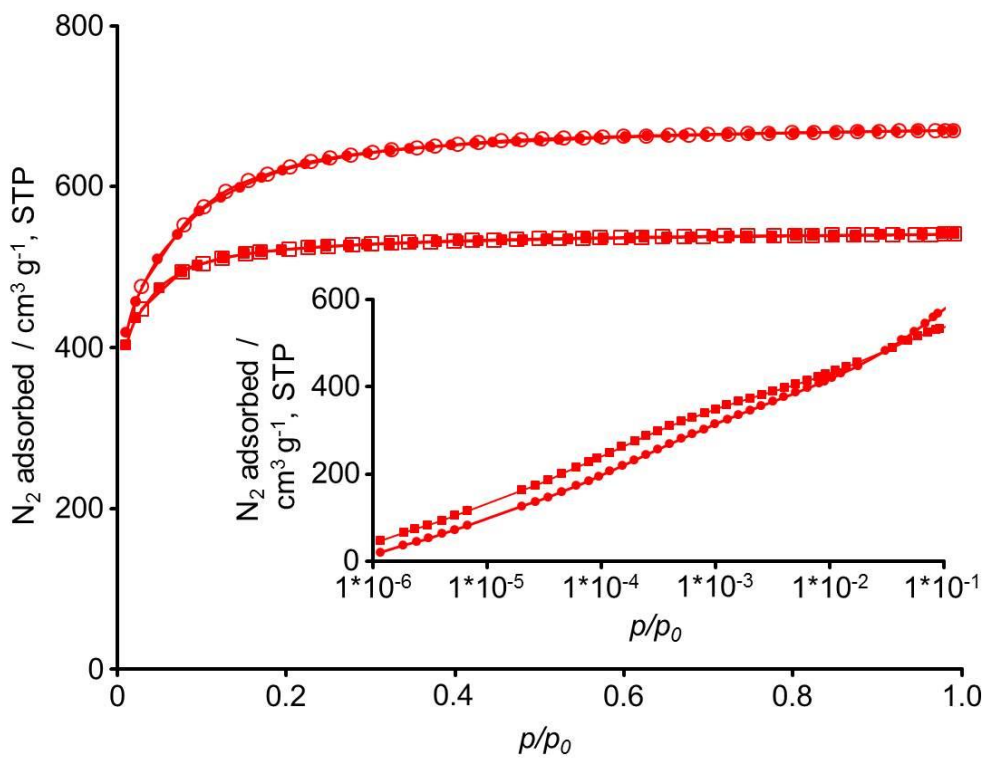
**Fig. S5:** NLDFT fits between the used carbon dioxide on carbon kernel (solid lines) and the experimental data (carbon dioxide physisorption at 0°C, empty symbols) for DUT-85 prepared at 1000°C/700°C (squares/circles).



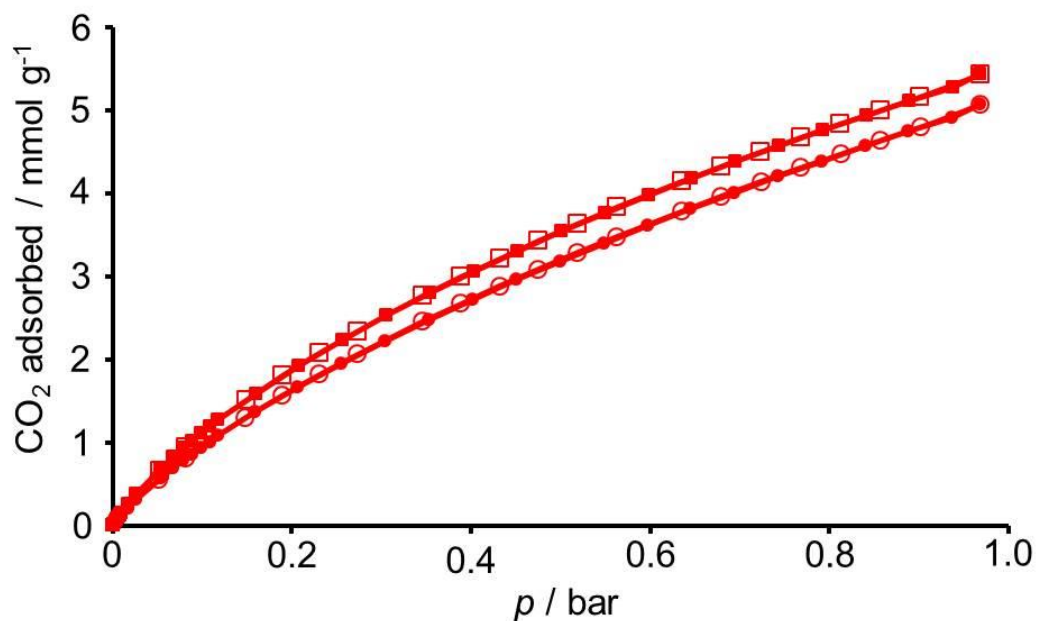
**Fig. S6:** Cumulative pore volumes of DUT-85 700°C (circles, left) and DUT-85 1000°C (squares, right) obtained from combined DFT analysis of carbon dioxide (grey) and nitrogen (black) physisorption isotherms.



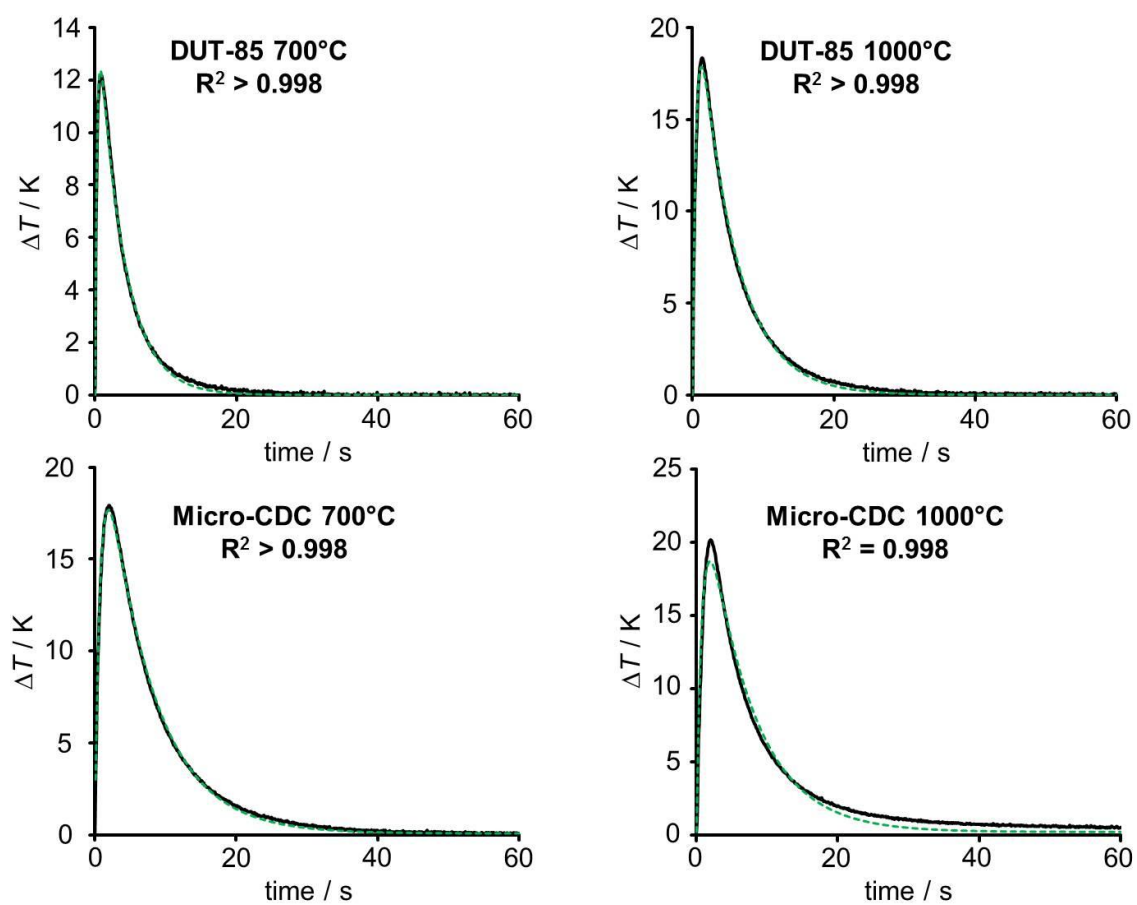
**Fig. S7:** Mercury intrusion/extrusion (filled symbols/empty symbols) curves of DUT-85 (black) and the SiC precursor aerogels (grey) prepared at 1000°C/700°C (squares/circles).



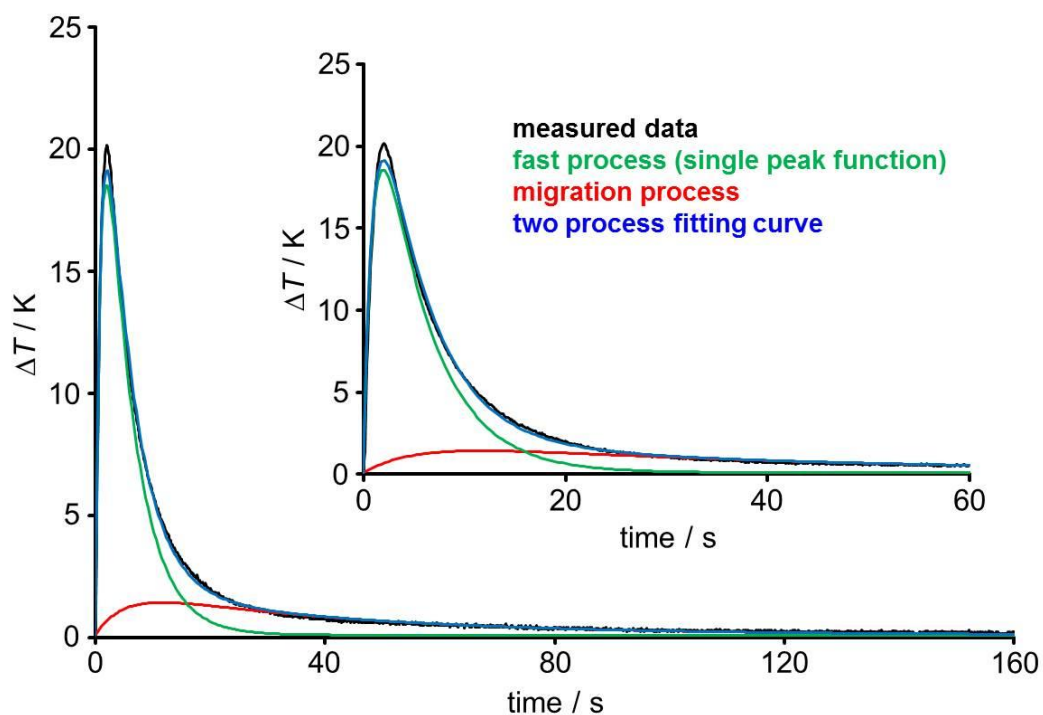
**Fig. S8:** Linear and semi-logarithmically (low pressure region; inset) plotted nitrogen adsorption/desorption (filled symbols/empty symbols) isotherms (-196°C) of the microporous CDC reference materials prepared at 1000°C/700°C (squares/circles).



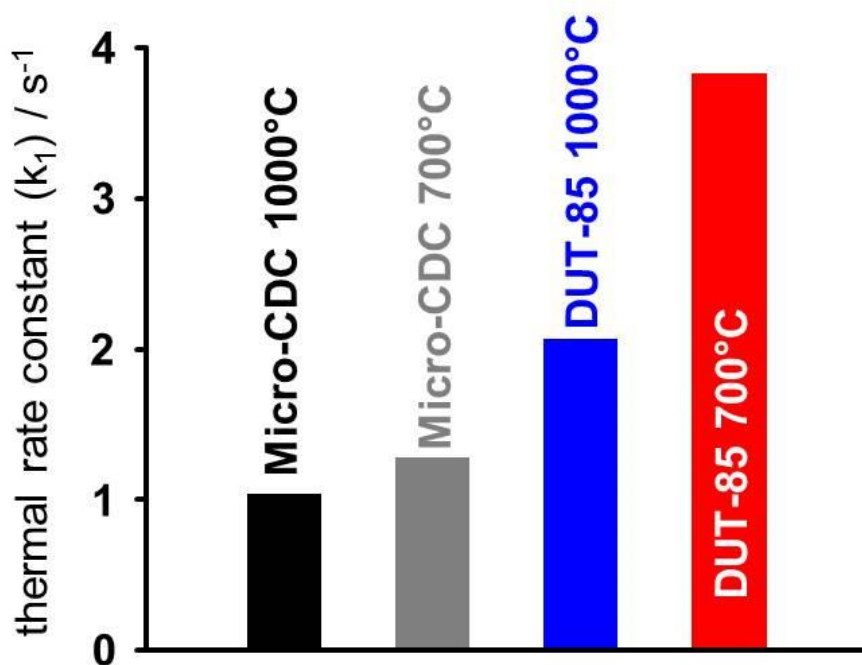
**Fig. S9:** Carbon dioxide (0°C) adsorption/desorption (filled symbols/empty symbols) isotherms of the microporous CDC reference materials prepared at 1000°C/700°C (squares/circles).



**Fig. S10:** Fitting curves (dotted green lines) of the single peak thermal response function (Equation 1) to the measured data (straight black lines).



**Fig. S11:** Fitting curve (blue) of the double peak (green and red line) thermal response function (Equation 2) to the measured data (straight black line) for the microporous CDC reference prepared at 1000°C.



**Fig. S12:** Thermal rate constants for adsorption process of 1<sup>st</sup> order ( $k_1$ ).

# Tables

**Tab. S1.** EDX elemental analyses of the SiC and CDC aerogels (DUT-85) at different synthesis temperatures.

<b>Material</b>	Carbon [wt.%]	Silicon [wt.%]	Oxygen [wt.%]	Chlorine [wt.%]	Platinum [wt.%]
<b>SiC Aerogel 700°C</b>	61.2	28.3	10.1	0.3	0.1
<b>SiC Aerogel 1000°C</b>	68.1	23.5	8.1	0.2	0.1
<b>CDC Aerogel 700°C</b>	99.3	<0.1	<0.1	0.5	0.2
<b>CDC Aerogel 1000°C</b>	99.7	<0.1	<0.1	<0.1	<0.1

**Tab. S2.** Porosity data Summary of the microporous CDC reference materials.

<b>Material</b>	SSA [m <sup>2</sup> g <sup>-1</sup> ] <sup>a)</sup>	V <sub>Pore, Total</sub> [cm <sup>3</sup> g <sup>-1</sup> ] <sup>b)</sup>	V <sub>Pore, &lt; 2 nm</sub> [cm <sup>3</sup> g <sup>-1</sup> ] <sup>c)</sup>	V <sub>Pore, &lt; 0.7 nm</sub> [cm <sup>3</sup> g <sup>-1</sup> ] <sup>d)</sup>
<b>Microporous CDC 700°C</b>	2298	1.04	0.92	0.22
<b>Microporous CDC 1000°C</b>	2017	0.84	0.78	0.23

<sup>a)</sup> Specific surface areas calculated from nitrogen physisorption using the BET equation (0.05 – 0.20  $p/p_0$ ); <sup>b)</sup> Total pore volumes from nitrogen physisorption calculated at  $p/p_0 = 0.99$ ; <sup>c)</sup> Micropore volume calculated from QSDFT cumulative pore volumes from nitrogen physisorption at a diameter of 2 nm.

<sup>d)</sup> NLDFT cumulative pore volumes from carbon dioxide physisorption at a diameter of 0.7 nm.