

## Next Generation Solid Amine Sorbents for Scalable Direct Air Capture of Carbon Dioxide

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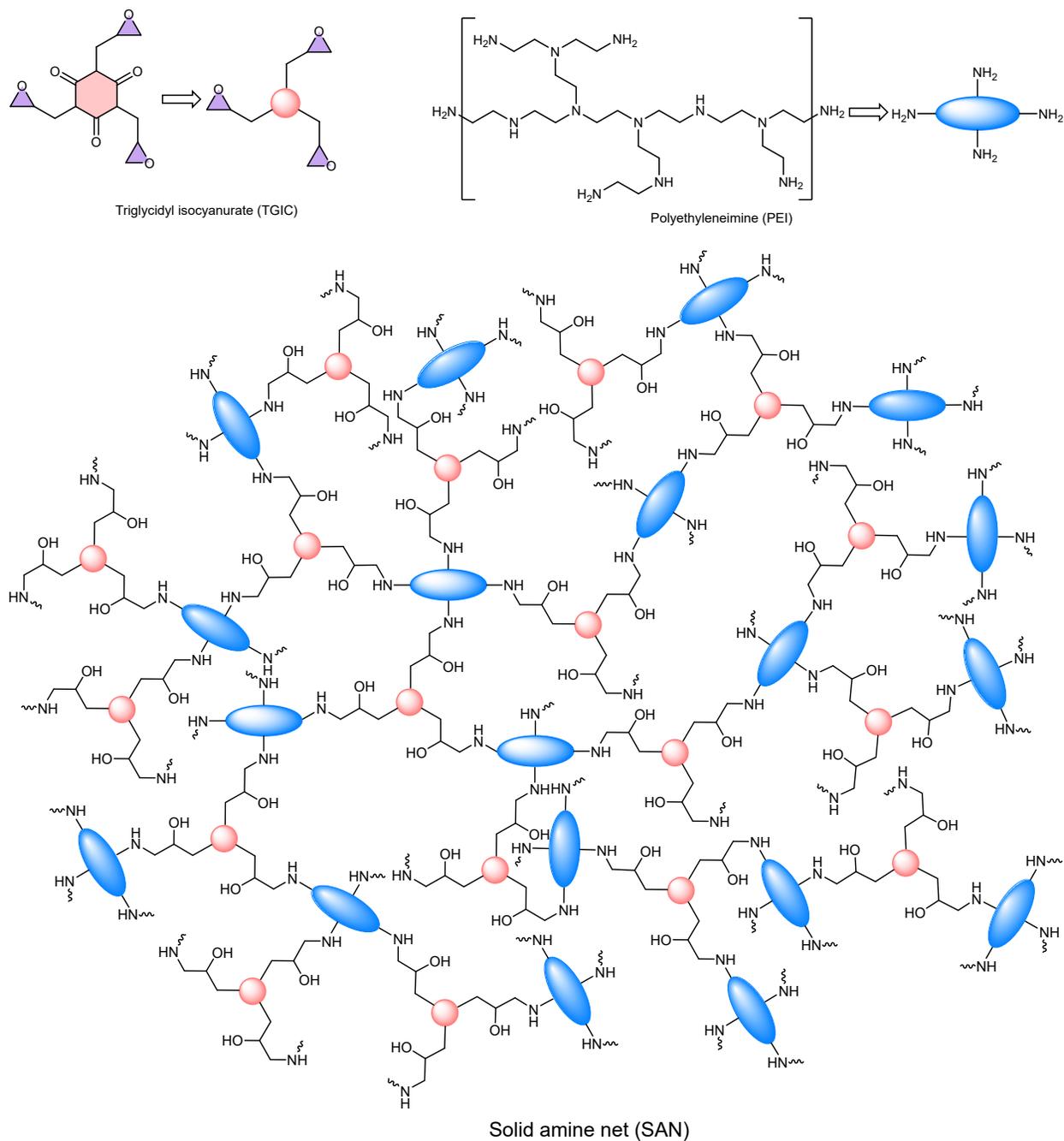
**Table S1** The alignment of synthesis for Class IV sorbents with the twelve principles of green chemistry

The 12 Principles of Green Chemistry	Alignment of Class IV Sorbents with Green Chemistry
1. Prevent waste	No waste produced in this reaction
2. Atom economy	All reactants added incorporate into the final product, making 100% atom efficiency
3. Less hazardous synthesis	All chemicals are easy to handle and widely used for other applications
4. Design benign chemicals	The polymer is inflammable, non-hazardous and non-volatile
5. Benign solvents & auxiliaries	The liquid used acts as a swelling agent and minimal to no solvents are used
6. Design for energy efficiency	The reaction is conducted under ambient conditions without additional supply of heat and pressure
7. Use of renewable feedstock	The reactants are cheap, mass-produced, and widely used in the industry for various applications, which can be made renewable if sustainable energy is used in the future.
8. Reduce derivatives	No derivatives used for this reaction
9. Catalysis	The reaction is auto catalytic without requiring additional catalysts
10. Design for degradation	The polymer has high stability under DAC conditions, but would slowly degrade due to oxidation
11. Real-time analysis for pollution prevention	The reaction is a one-pot process without pollution generated
12. Inherently benign chemistry for accident prevention	The reaction generates mild heat which can be easily managed, no volatiles/gases/toxic chemicals produced, eliminating the possibilities of accident

**Table S2** Adsorption kinetics for Class IV, conventional Class I sorbents and commercial Lewatit resin

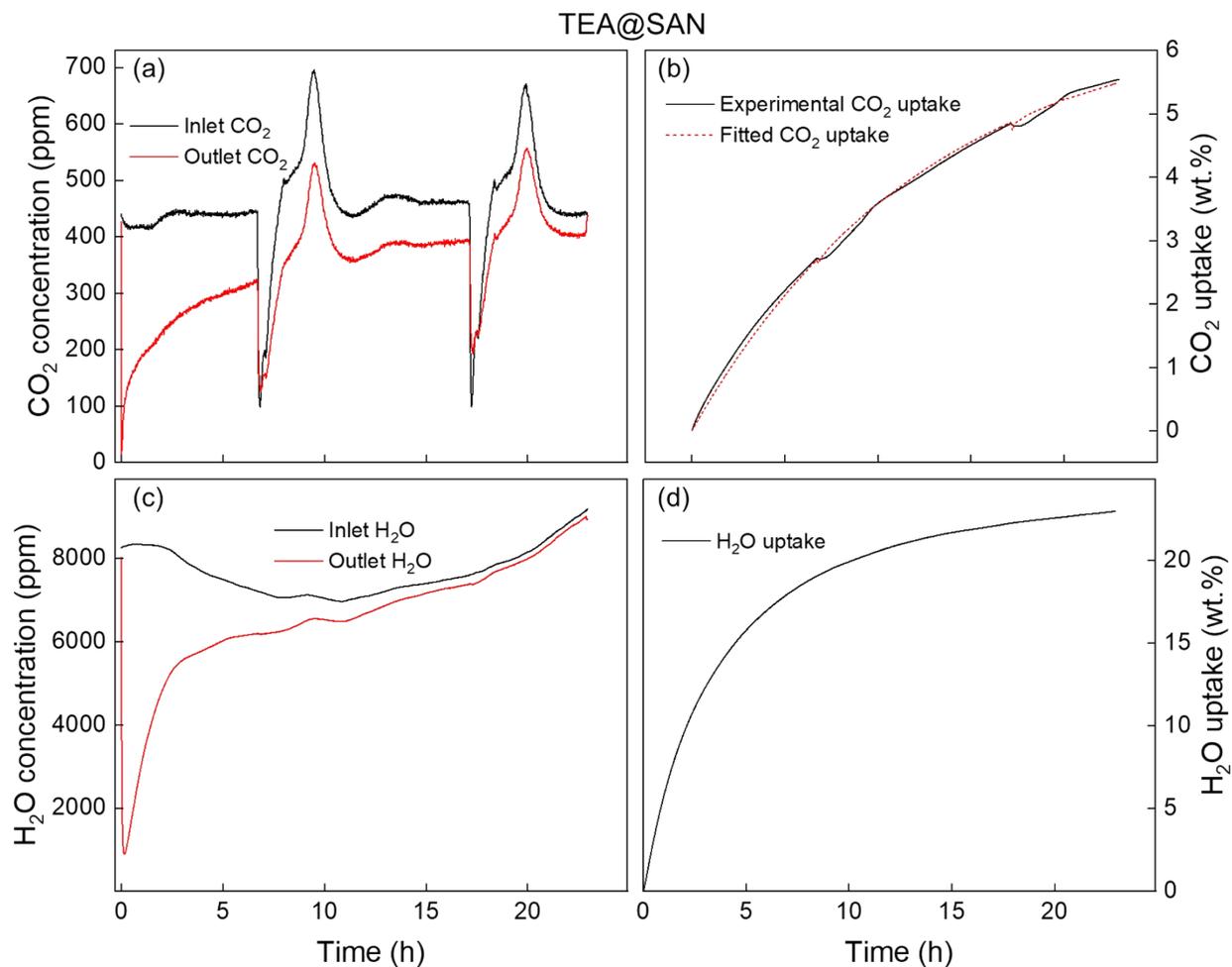
Samples	Kinetic Parameters			
	$q_e$	K	$k_1$	$R^2$
TEA@SAN	1.51	0.2712	0.079	0.9975
MDEA@SAN	1.65	0.1895	0.116	0.9969
DEA@SAN	4.41	0.1416	0.065	0.9899
0.5DEA&0.5TEA@SAN	3.06	0.3176	0.080	0.9986
0.5DEA&0.5MDEA@SAN	3.25	0.3418	0.090	0.9982
H <sub>2</sub> O@SAN	0.78	1.7364	0.233	0.9955
TEA/SiO <sub>2</sub>	1.57	0.0829	0.003	0.9933
MDEA/SiO <sub>2</sub>	0.32	0.0936	0.022	0.9967
DEA/SiO <sub>2</sub>	1.13	0.1447	0.273	0.9798
PEI800/SiO <sub>2</sub>	2.59	0.5364	0.105	0.9976
Lewatit resin	1.71	0.4704	0.192	0.9889

## Supplementary Figure 1



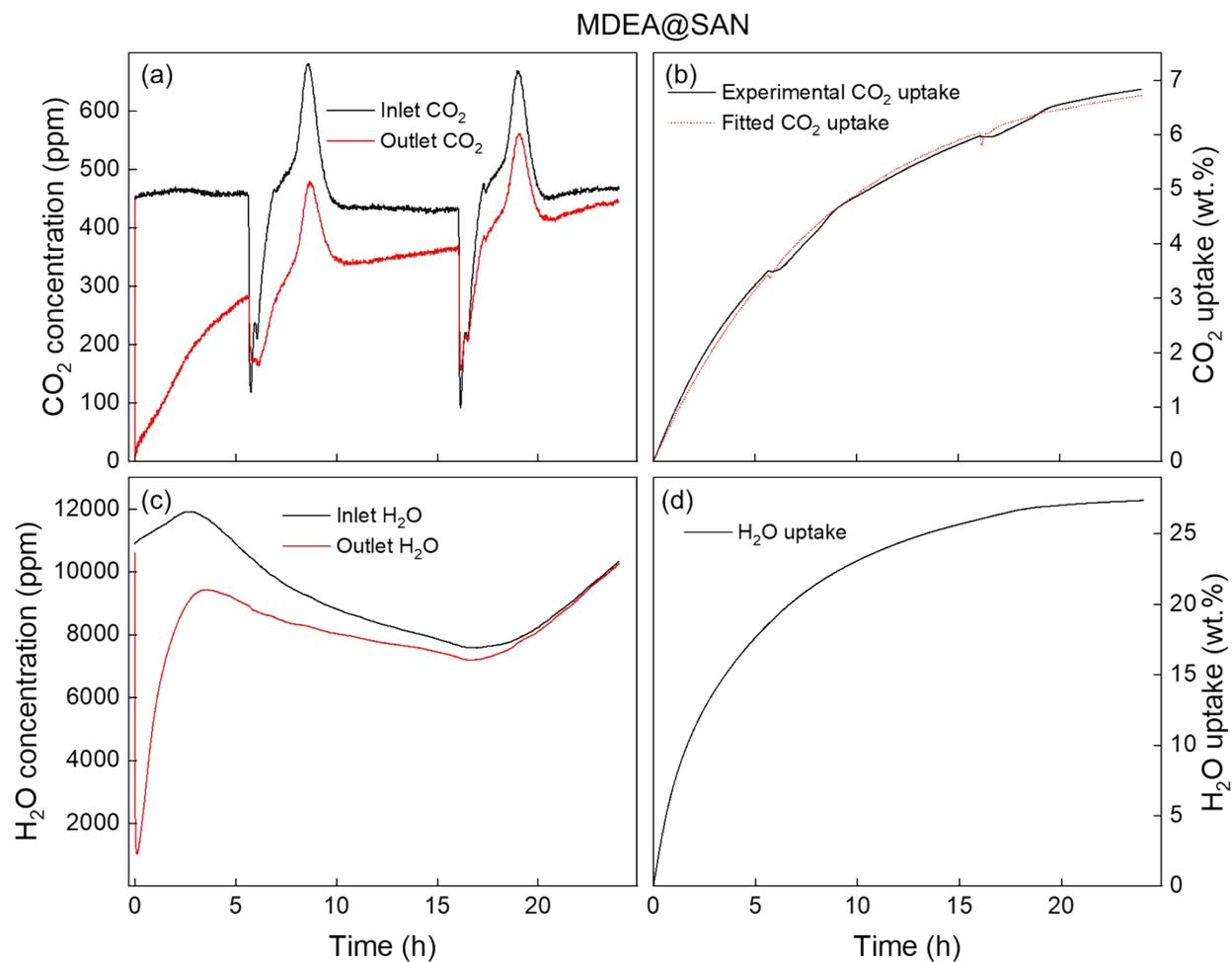
**Figure S1** Schematic of solid amine network with PEI being knitted together by TGIC, which is performed at ambient conditions and completed in 10 minutes.

## Supplementary Figure 2



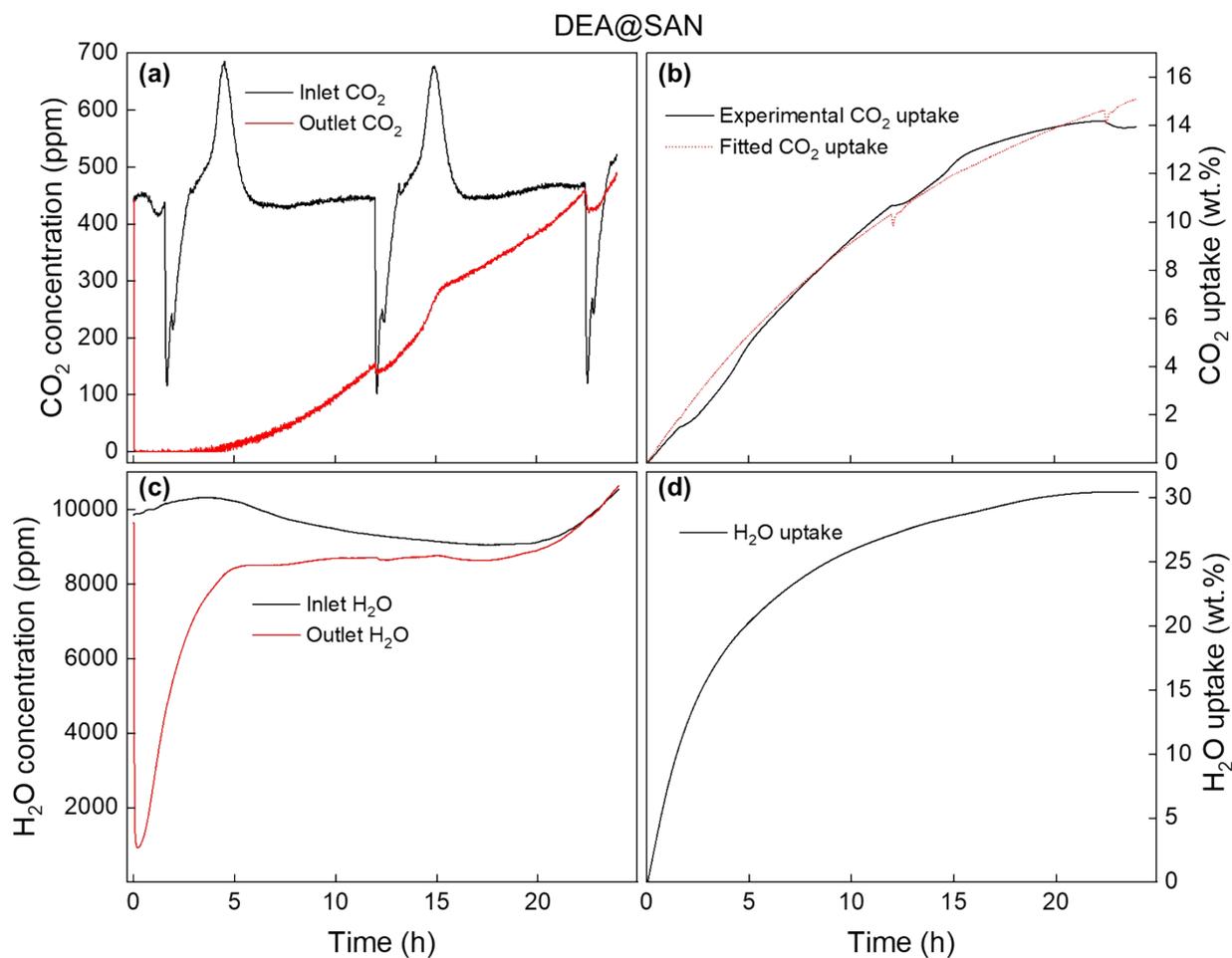
**Figure S2** The dynamic changes in CO<sub>2</sub> and H<sub>2</sub>O concentrations at the inlet and outlet, as well as CO<sub>2</sub> and H<sub>2</sub>O uptakes as a function of time for TEA@SAN during the DAC test: (a) CO<sub>2</sub> concentrations; (b) experimental and fitted CO<sub>2</sub> uptake; (c) H<sub>2</sub>O concentrations; and (d) H<sub>2</sub>O uptake.

### Supplementary Figure 3



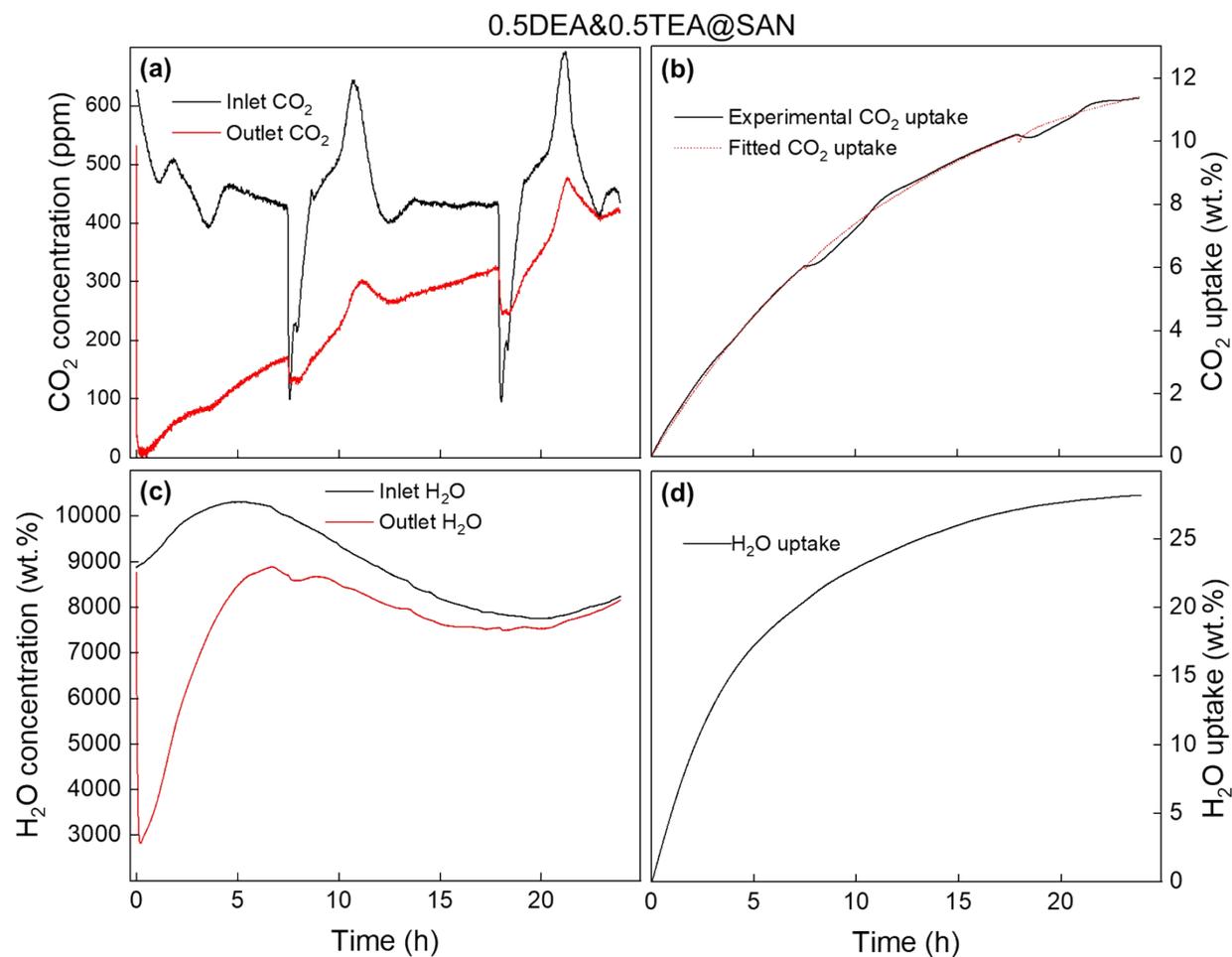
**Figure S3** The dynamic changes in CO<sub>2</sub> and H<sub>2</sub>O concentrations at the inlet and outlet, as well as CO<sub>2</sub> and H<sub>2</sub>O uptakes as a function of time for MDEA@SAN during the DAC test: (a) CO<sub>2</sub> concentrations; (b) experimental and fitted CO<sub>2</sub> uptake; (c) H<sub>2</sub>O concentrations; and (d) H<sub>2</sub>O uptake.

### Supplementary Figure 4



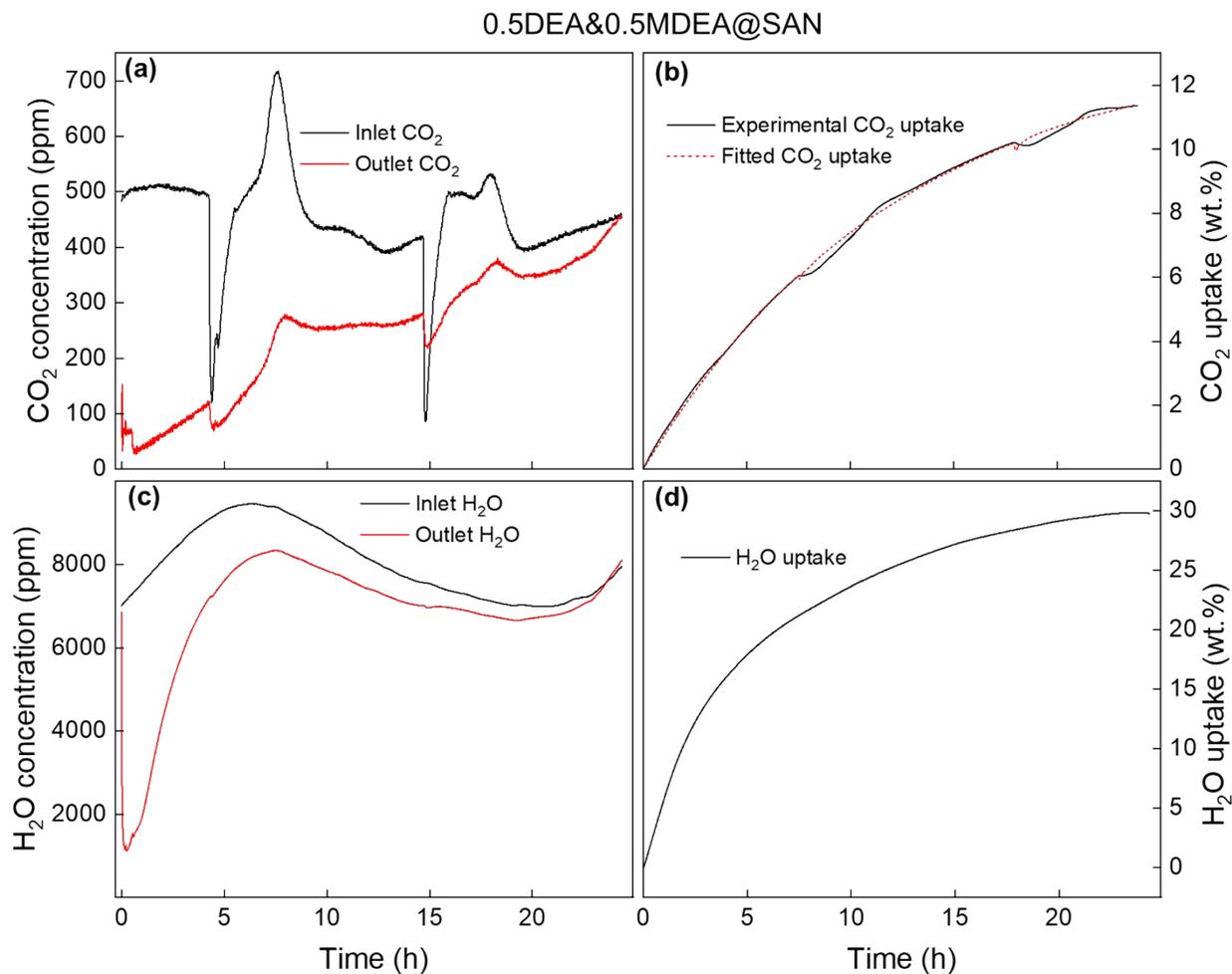
**Figure S4** The dynamic changes in CO<sub>2</sub> and H<sub>2</sub>O concentrations at the inlet and outlet, as well as CO<sub>2</sub> and H<sub>2</sub>O uptakes as a function of time for DEA@SAN during the DAC test: (a) CO<sub>2</sub> concentrations; (b) experimental and fitted CO<sub>2</sub> uptake; (c) H<sub>2</sub>O concentrations; and (d) H<sub>2</sub>O uptake.

## Supplementary Figure 5



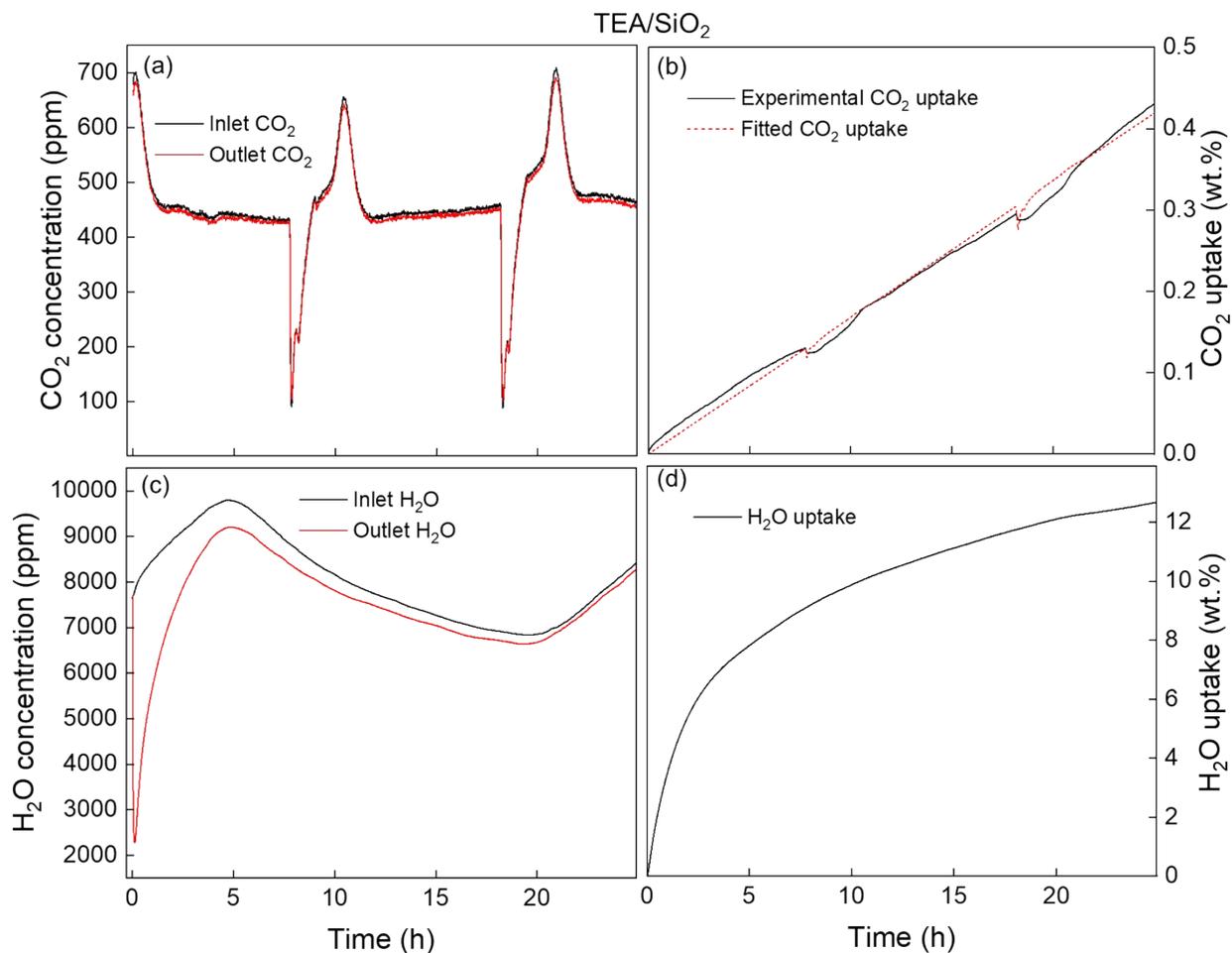
**Figure S5** The dynamic changes in CO<sub>2</sub> and H<sub>2</sub>O concentrations at the inlet and outlet, as well as CO<sub>2</sub> and H<sub>2</sub>O uptakes as a function of time for 0.5DEA&0.5TEA@SAN during the DAC test: (a) CO<sub>2</sub> concentrations; (b) experimental and fitted CO<sub>2</sub> uptake; (c) H<sub>2</sub>O concentrations; and (d) H<sub>2</sub>O uptake.

## Supplementary Figure 6



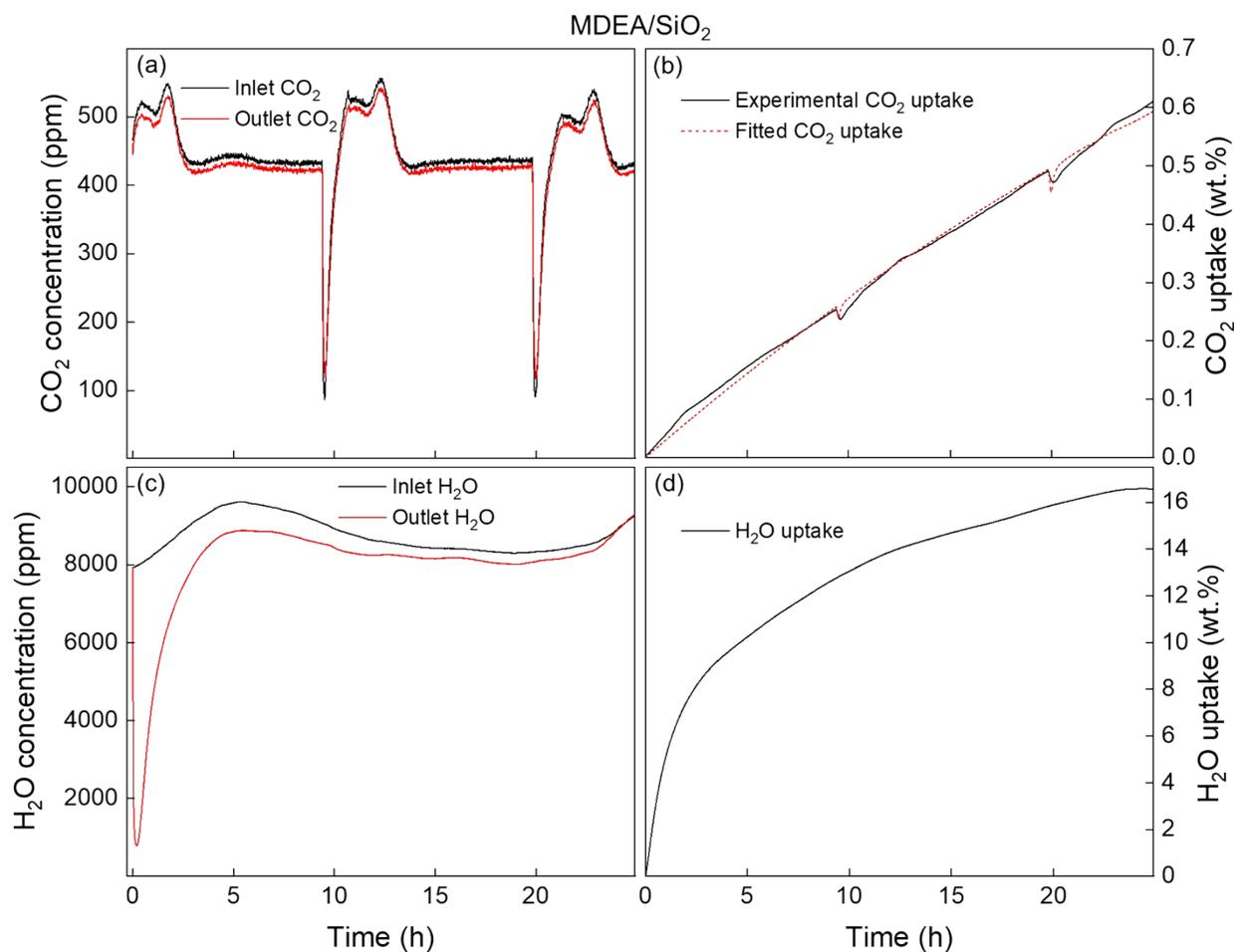
**Figure S6** The dynamic changes in CO<sub>2</sub> and H<sub>2</sub>O concentrations at the inlet and outlet, as well as CO<sub>2</sub> and H<sub>2</sub>O uptakes as a function of time for 0.5DEA&0.5MDEA@SAN during the DAC test: (a) CO<sub>2</sub> concentrations; (b) experimental and fitted CO<sub>2</sub> uptake; (c) H<sub>2</sub>O concentrations; and (d) H<sub>2</sub>O uptake.

## Supplementary Figure 7



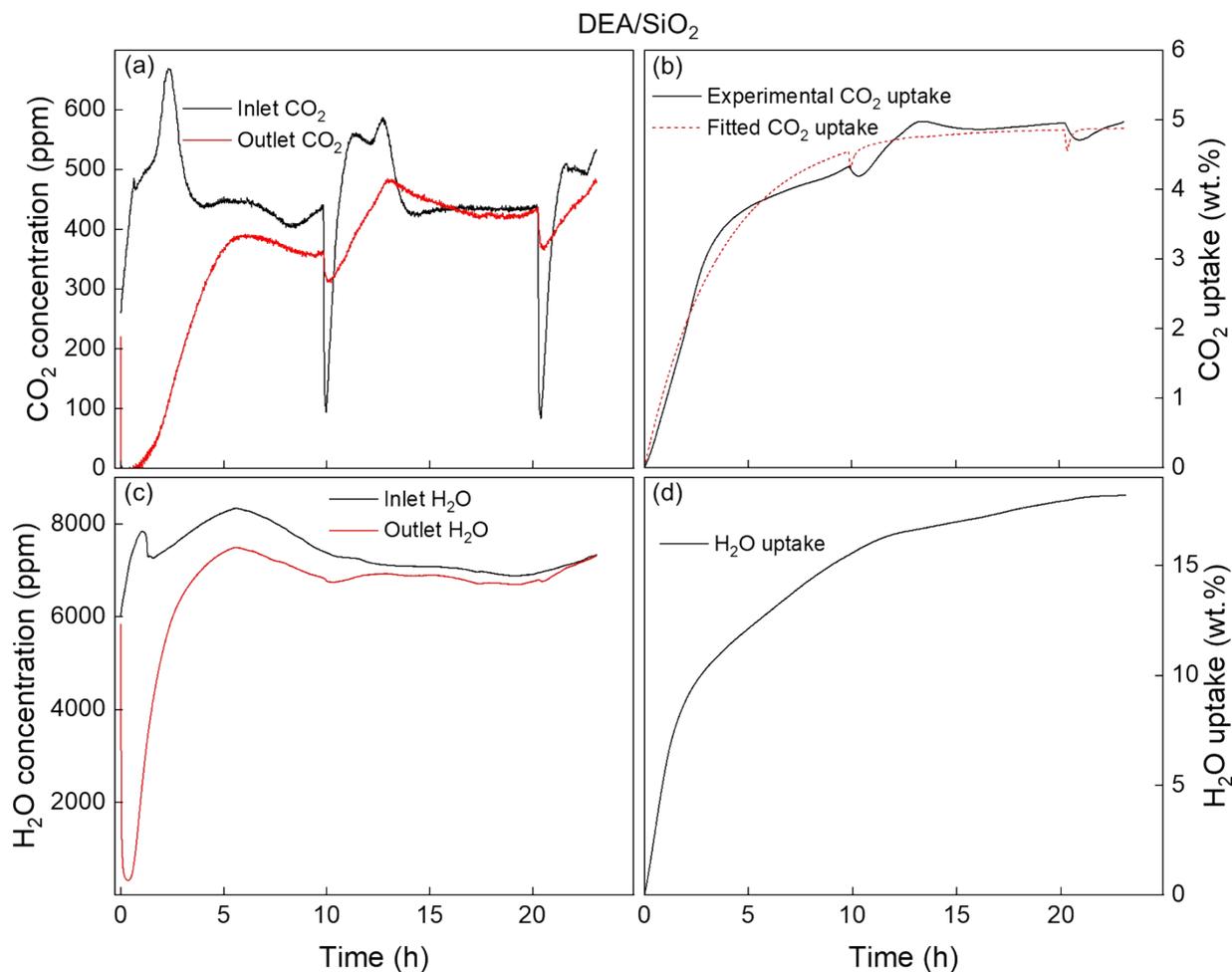
**Figure S7** The dynamic changes in CO<sub>2</sub> and H<sub>2</sub>O concentrations at the inlet and outlet, as well as CO<sub>2</sub> and H<sub>2</sub>O uptakes as a function of time for TEA/SiO<sub>2</sub> during the DAC test: (a) CO<sub>2</sub> concentrations; (b) experimental and fitted CO<sub>2</sub> uptake; (c) H<sub>2</sub>O concentrations; and (d) H<sub>2</sub>O uptake.

### Supplementary Figure 8



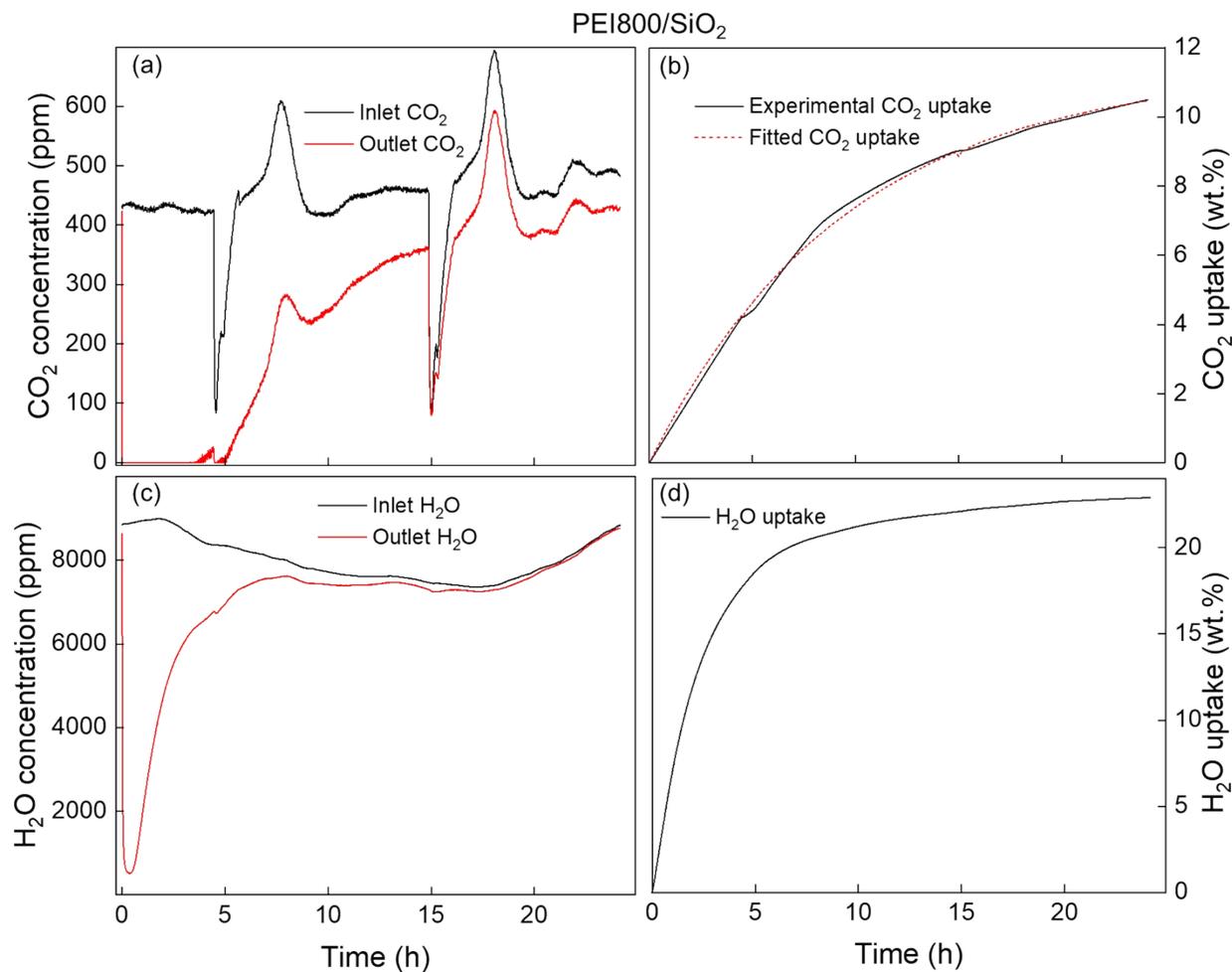
**Figure S8** The dynamic changes in CO<sub>2</sub> and H<sub>2</sub>O concentrations at the inlet and outlet, as well as CO<sub>2</sub> and H<sub>2</sub>O uptakes as a function of time for MDEA/SiO<sub>2</sub> during the DAC test: (a) CO<sub>2</sub> concentrations; (b) experimental and fitted CO<sub>2</sub> uptake; (c) H<sub>2</sub>O concentrations; and (d) H<sub>2</sub>O uptake.

## Supplementary Figure 9



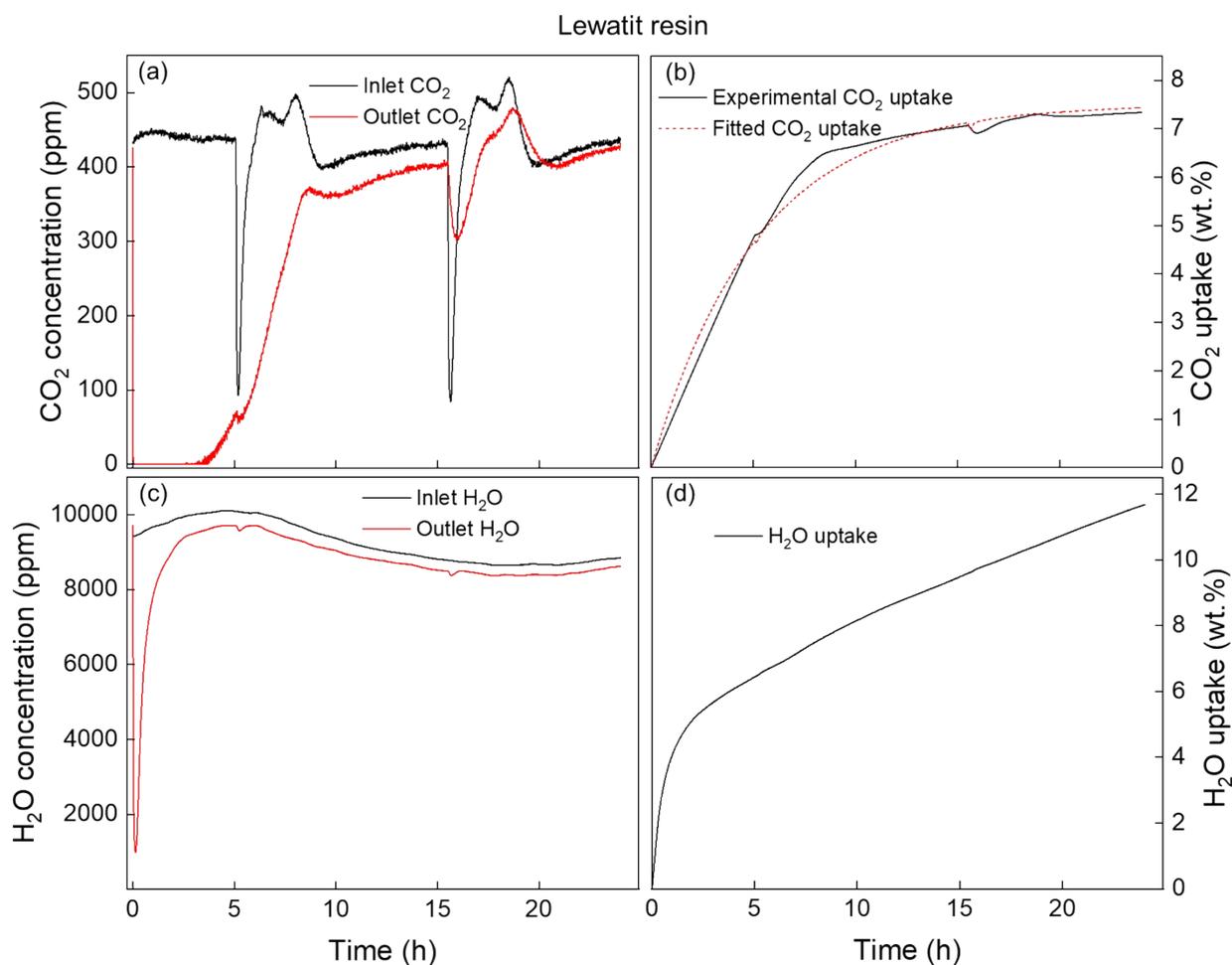
**Figure S9** The dynamic changes in CO<sub>2</sub> and H<sub>2</sub>O concentrations at the inlet and outlet, as well as CO<sub>2</sub> and H<sub>2</sub>O uptakes as a function of time for DEA/SiO<sub>2</sub> during the DAC test: (a) CO<sub>2</sub> concentrations; (b) experimental and fitted CO<sub>2</sub> uptake; (c) H<sub>2</sub>O concentrations; and (d) H<sub>2</sub>O uptake.

## Supplementary Figure 10



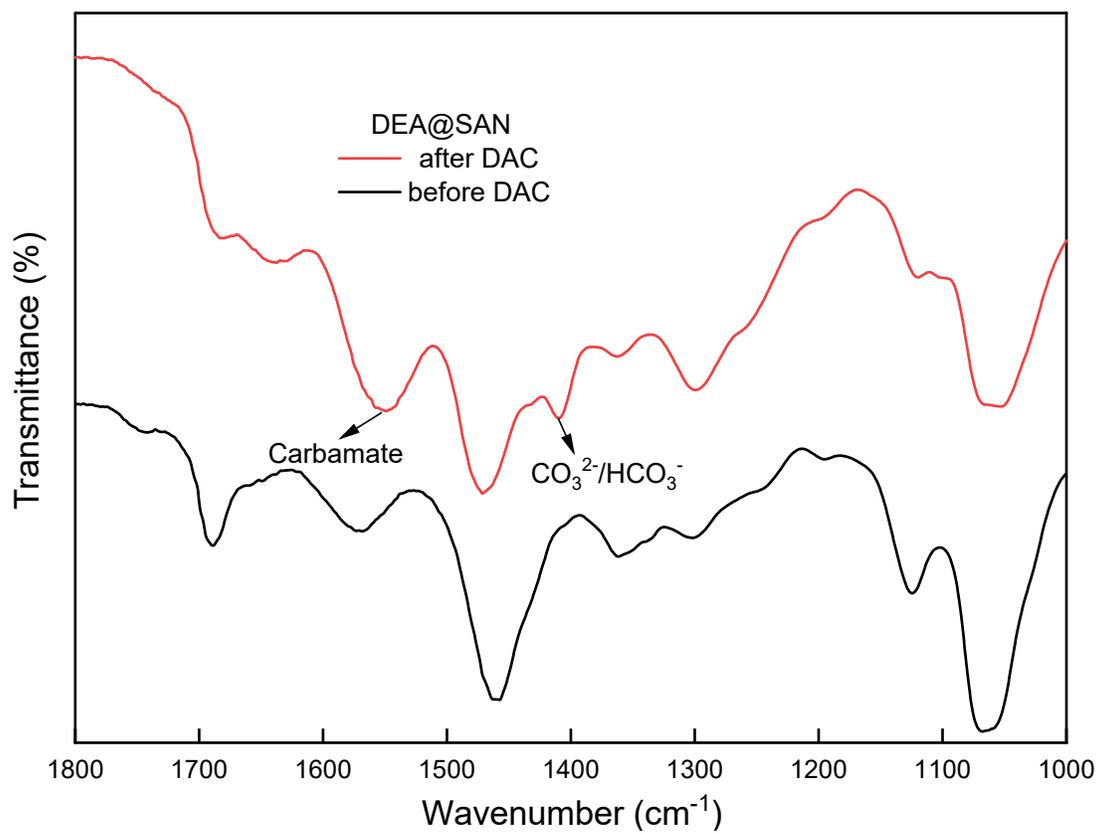
**Figure S10** The dynamic changes in CO<sub>2</sub> and H<sub>2</sub>O concentrations at the inlet and outlet, as well as CO<sub>2</sub> and H<sub>2</sub>O uptakes as a function of time for PEI800/SiO<sub>2</sub> during the DAC test: (a) CO<sub>2</sub> concentrations; (b) experimental and fitted CO<sub>2</sub> uptake; (c) H<sub>2</sub>O concentrations; and (d) H<sub>2</sub>O uptake.

## Supplementary Figure 11



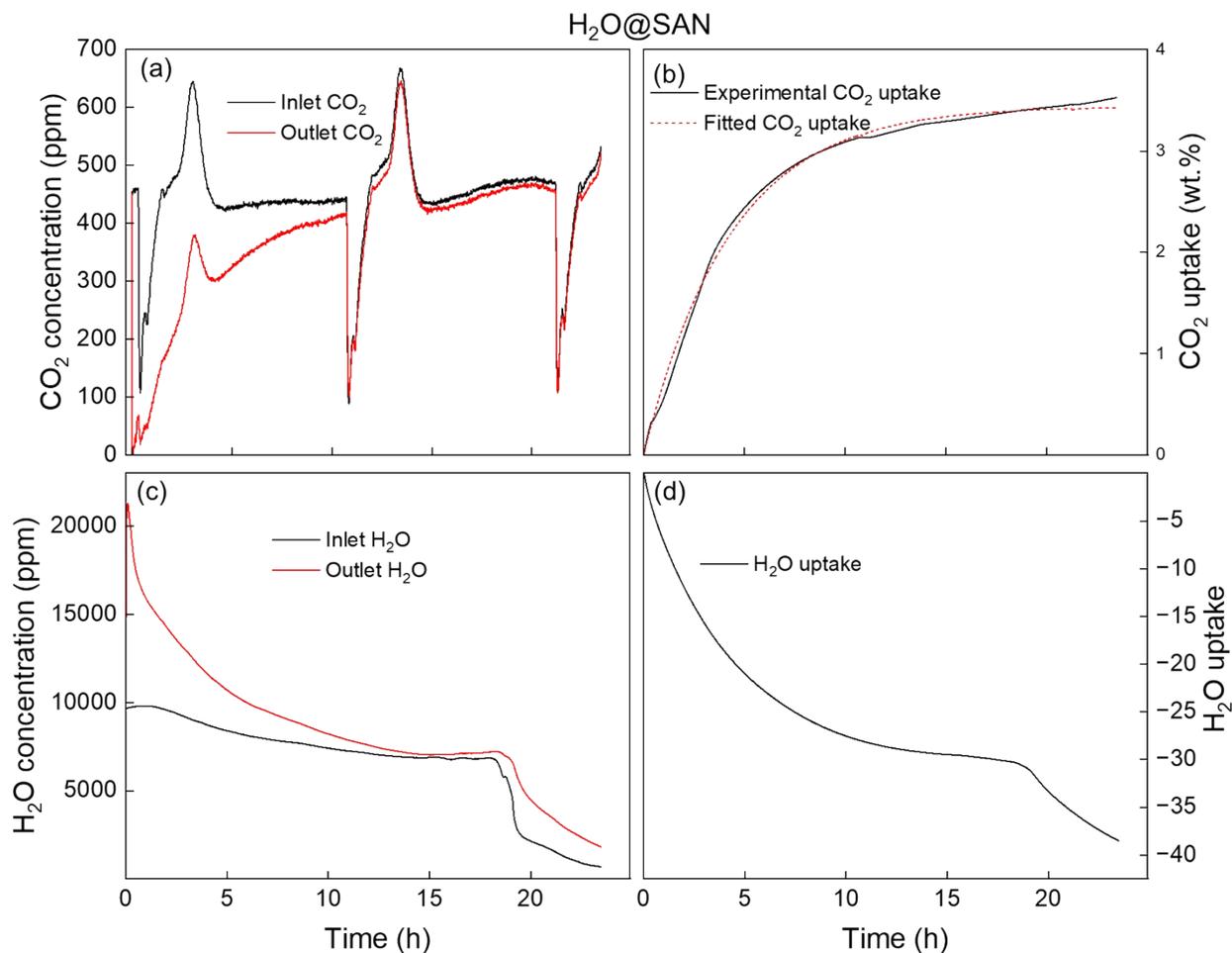
**Figure S11** The dynamic changes in CO<sub>2</sub> and H<sub>2</sub>O concentrations at the inlet and outlet, as well as CO<sub>2</sub> and H<sub>2</sub>O uptakes as a function of time for Lewatit resin during the DAC test: (a) CO<sub>2</sub> concentrations; (b) experimental and fitted CO<sub>2</sub> uptake; (c) H<sub>2</sub>O concentrations; and (d) H<sub>2</sub>O uptake.

## Supplementary Figure 12



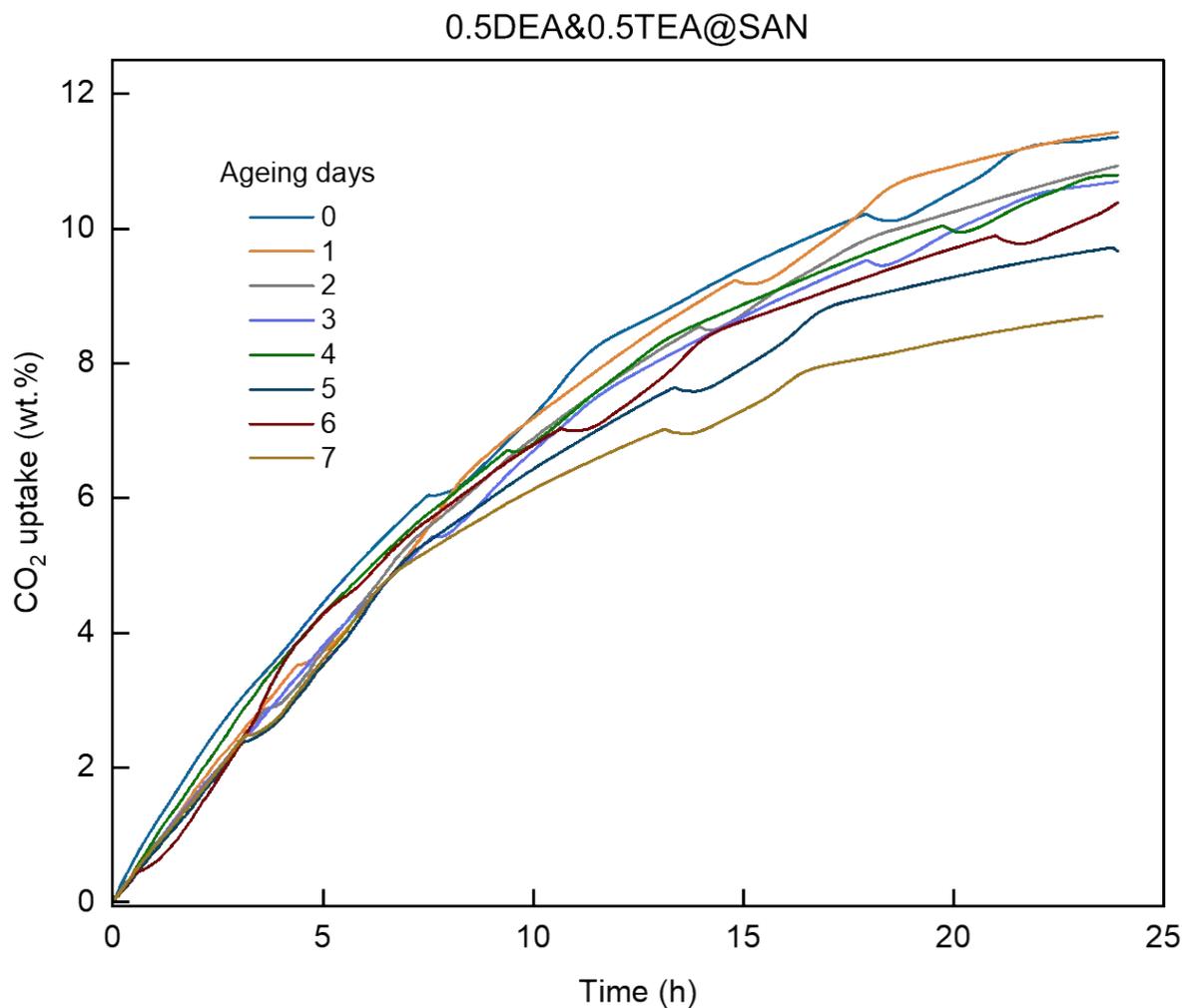
**Figure S12** IR spectra to identify new species formed after DAC test for DEA@SAN.

### Supplementary Figure 13



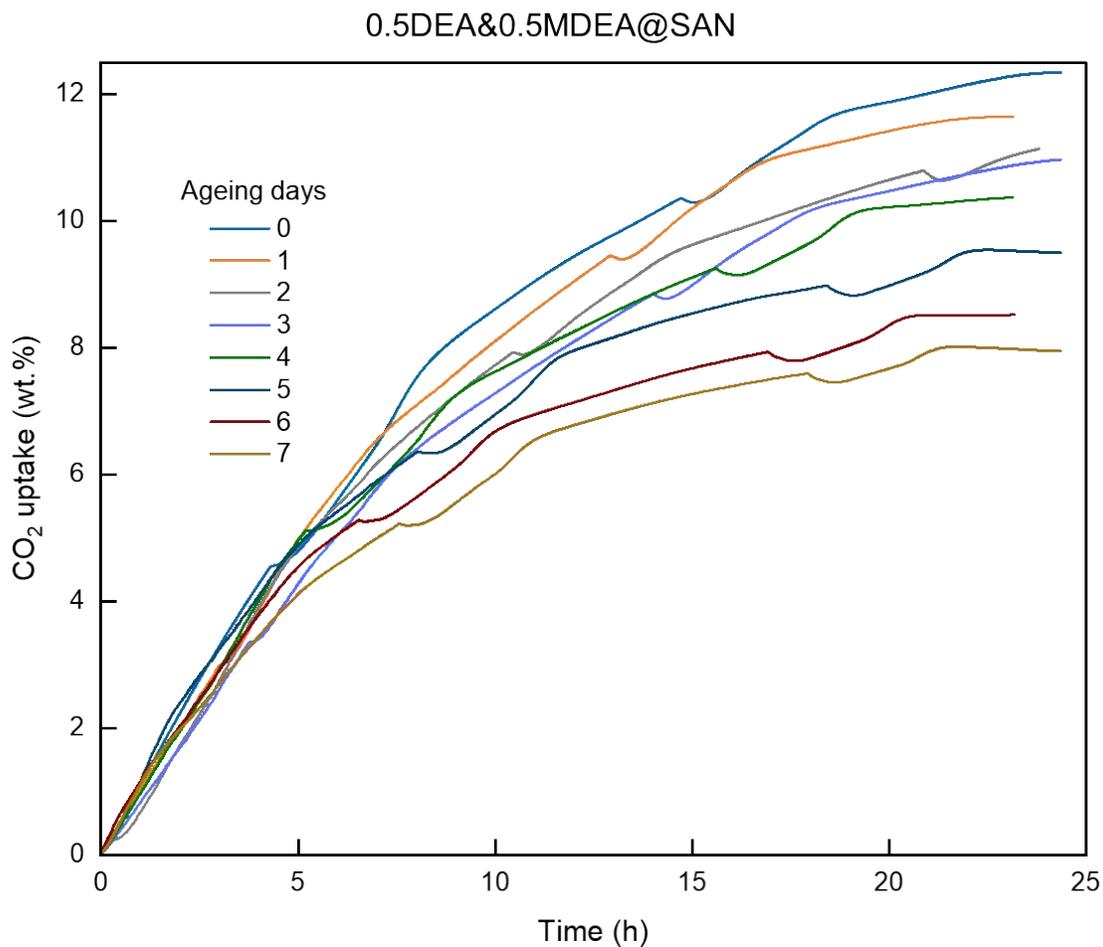
**Figure S13** The dynamic changes in CO<sub>2</sub> and H<sub>2</sub>O concentrations at the inlet and outlet, as well as CO<sub>2</sub> and H<sub>2</sub>O uptakes as a function of time for H<sub>2</sub>O@SAN during the DAC test: (a) CO<sub>2</sub> concentrations; (b) experimental and fitted CO<sub>2</sub> uptake; (c) H<sub>2</sub>O concentrations; and (d) H<sub>2</sub>O uptake.

### Supplementary Figure 14



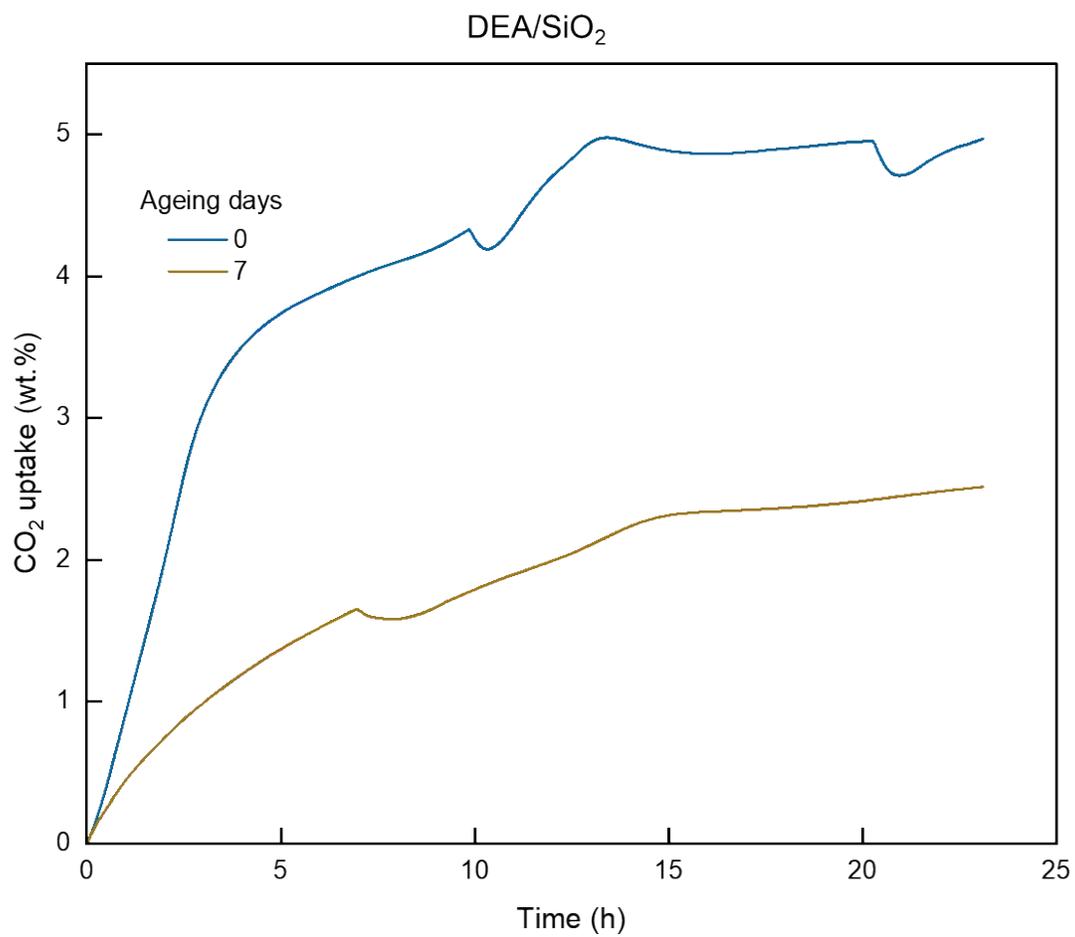
**Figure S14** CO<sub>2</sub> uptake as a function of time for 0.5DEA&0.5TEA@SAN at different ageing days. Accelerated ageing conditions: 85 °C in air.

## Supplementary Figure 15



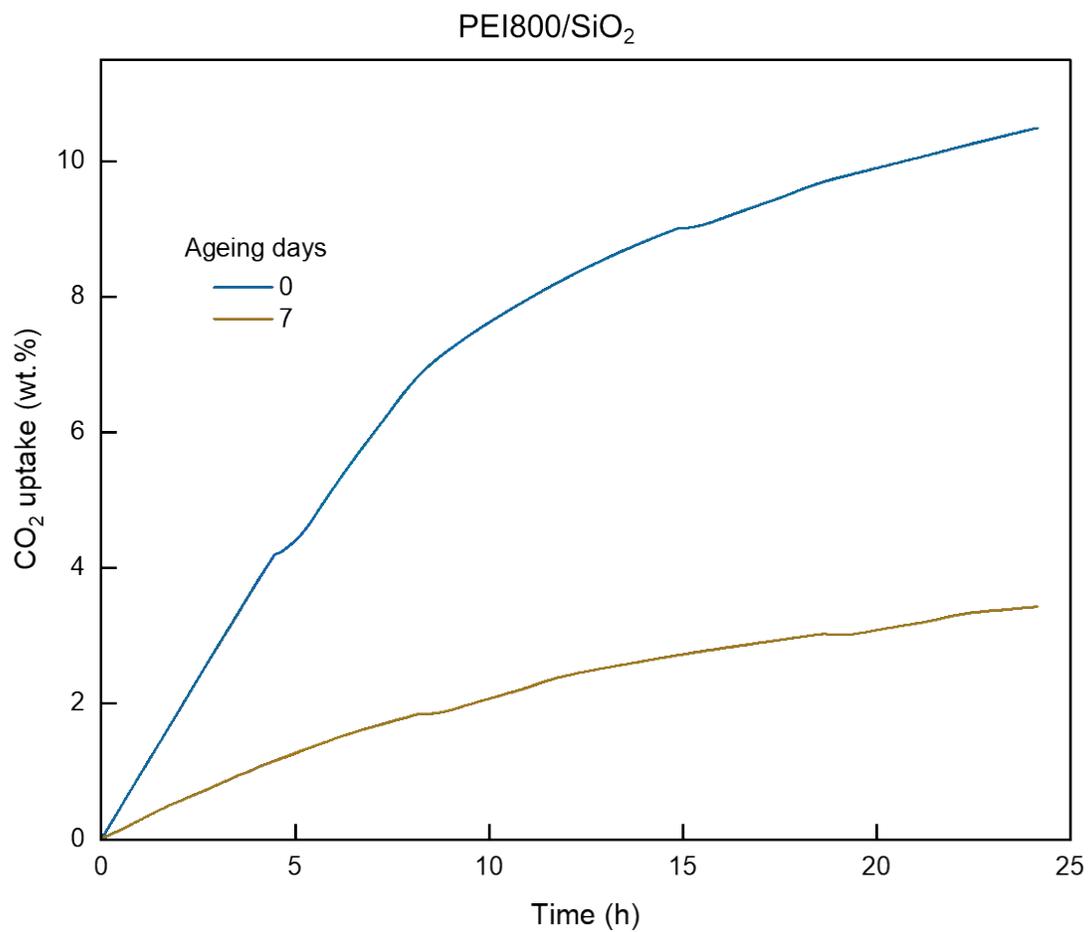
**Figure S15** CO<sub>2</sub> uptake as a function of time for 0.5DEA&0.5MDEA@SAN at different ageing days. Accelerated ageing conditions: 85 °C in air.

## Supplementary Figure 16



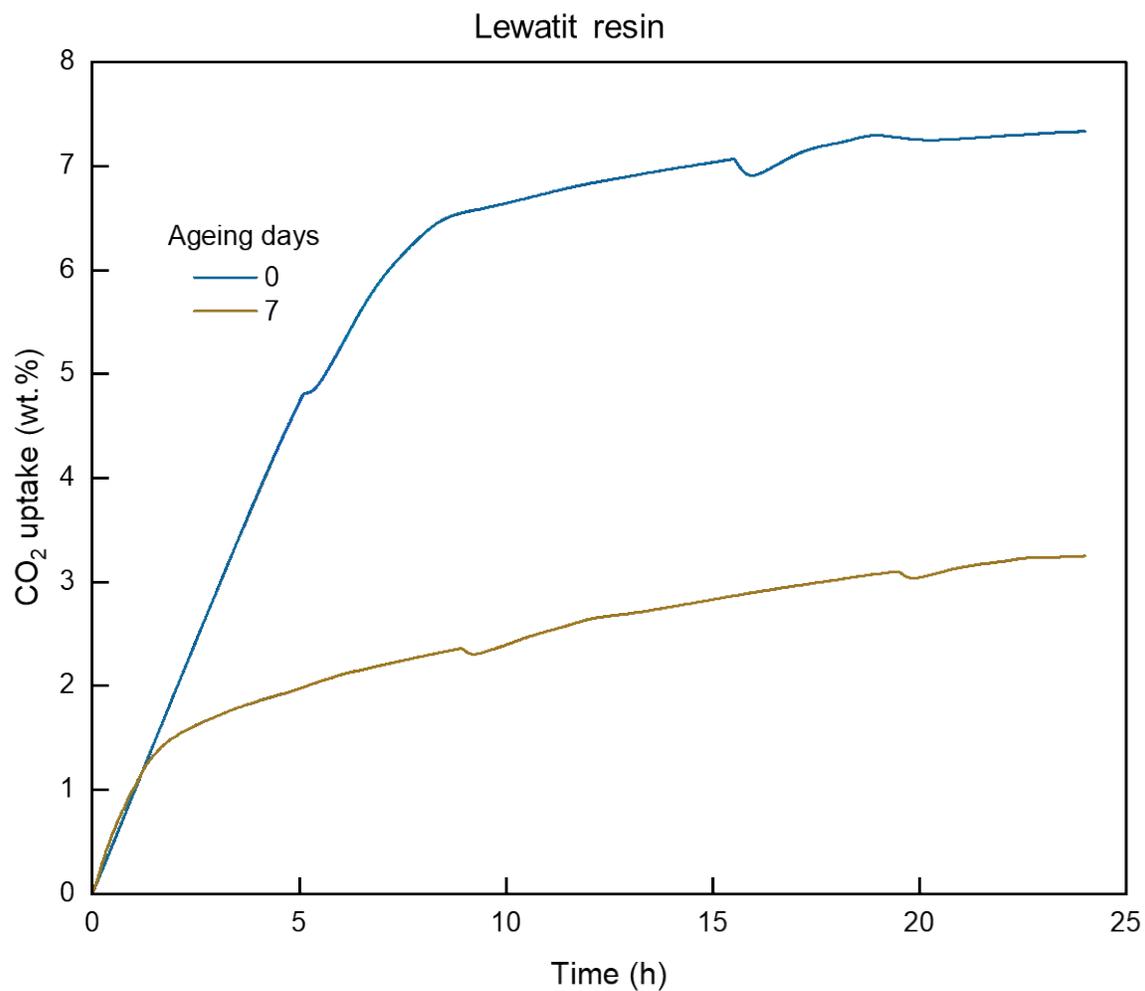
**Figure S16** CO<sub>2</sub> uptake as a function of time for DEA/SiO<sub>2</sub> at different ageing days. Accelerated ageing conditions: 85 °C in air.

### Supplementary Figure 17



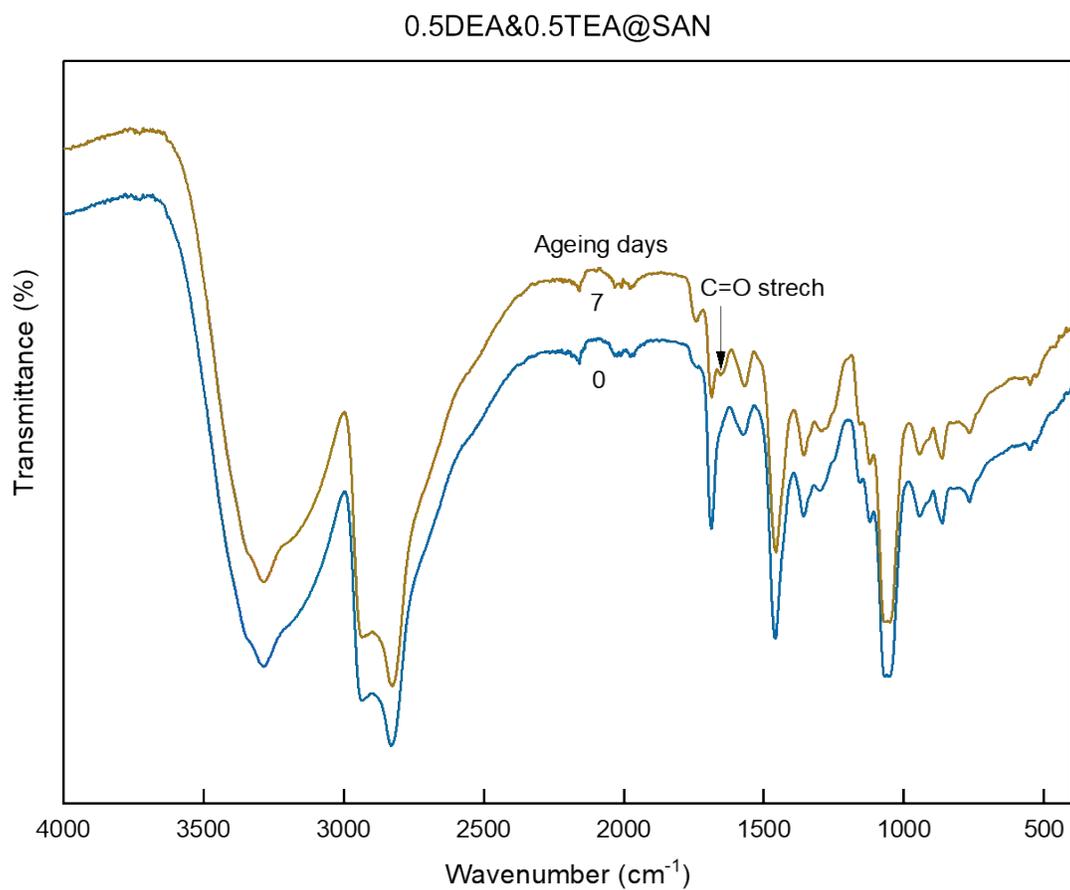
**Figure S17** CO<sub>2</sub> uptake as a function of time for PEI800/SiO<sub>2</sub> at different ageing days. Accelerated ageing conditions: 85 °C in air.

### Supplementary Figure 18



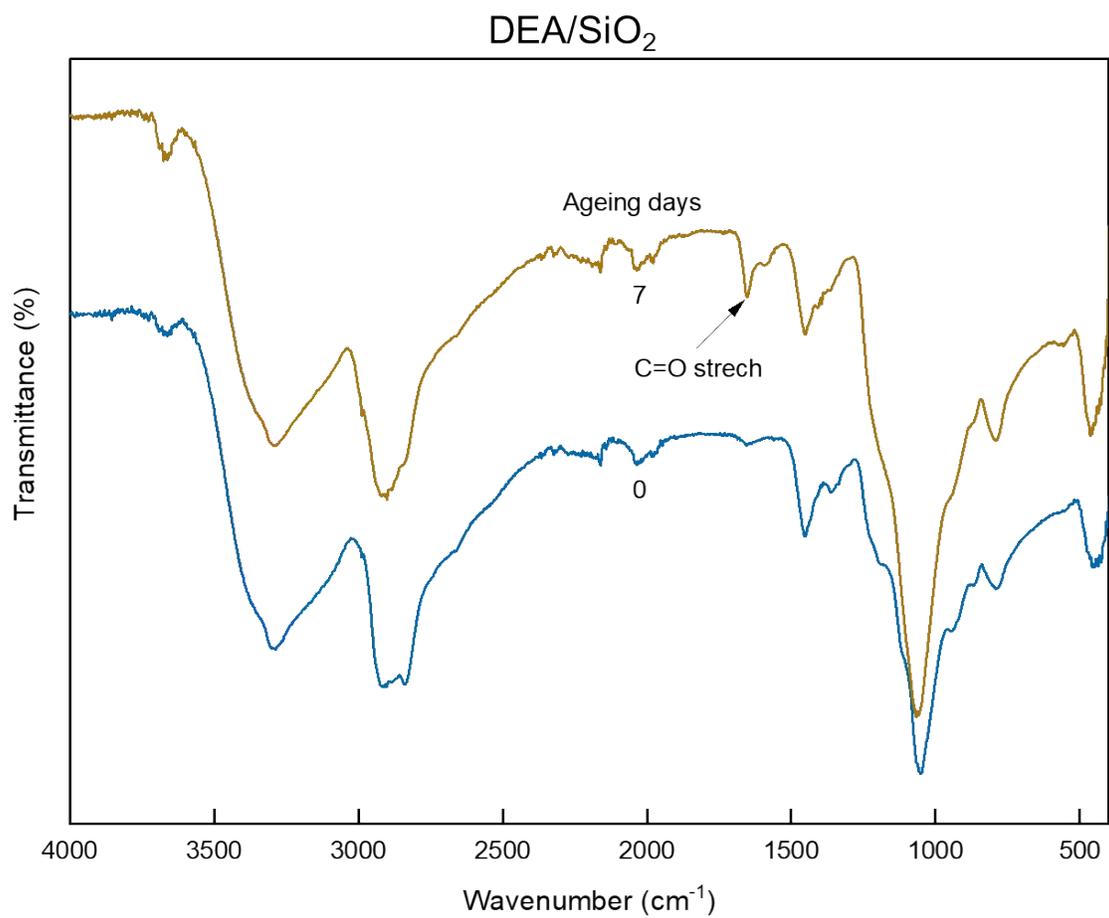
**Figure S18** CO<sub>2</sub> uptake as a function of time for Lewatit resin on different ageing days. Accelerated ageing conditions: 85 °C in air.

## Supplementary Figure 19



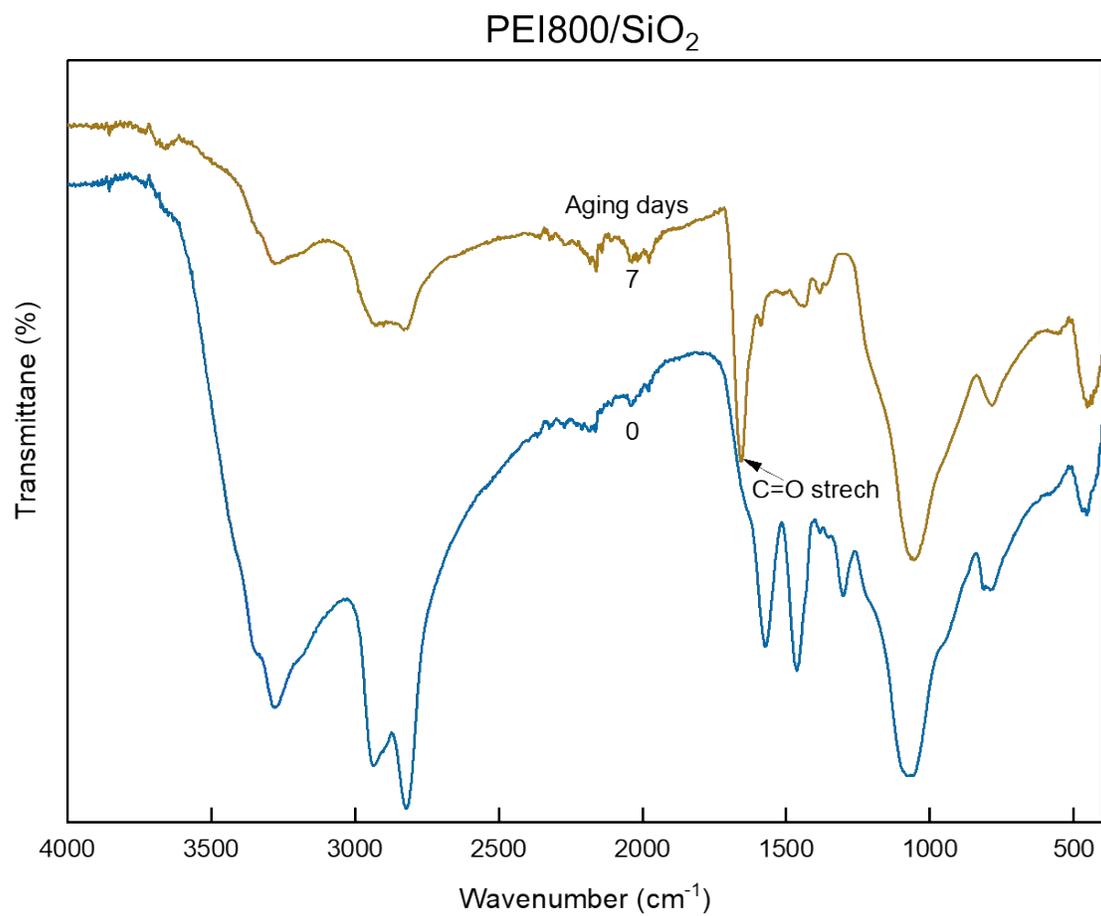
**Figure S19** Changes in IR absorption for 0.5DEA&0.5TEA@SAN at different ageing days. Accelerated ageing conditions: 85 °C in air.

Supplementary Figure 20



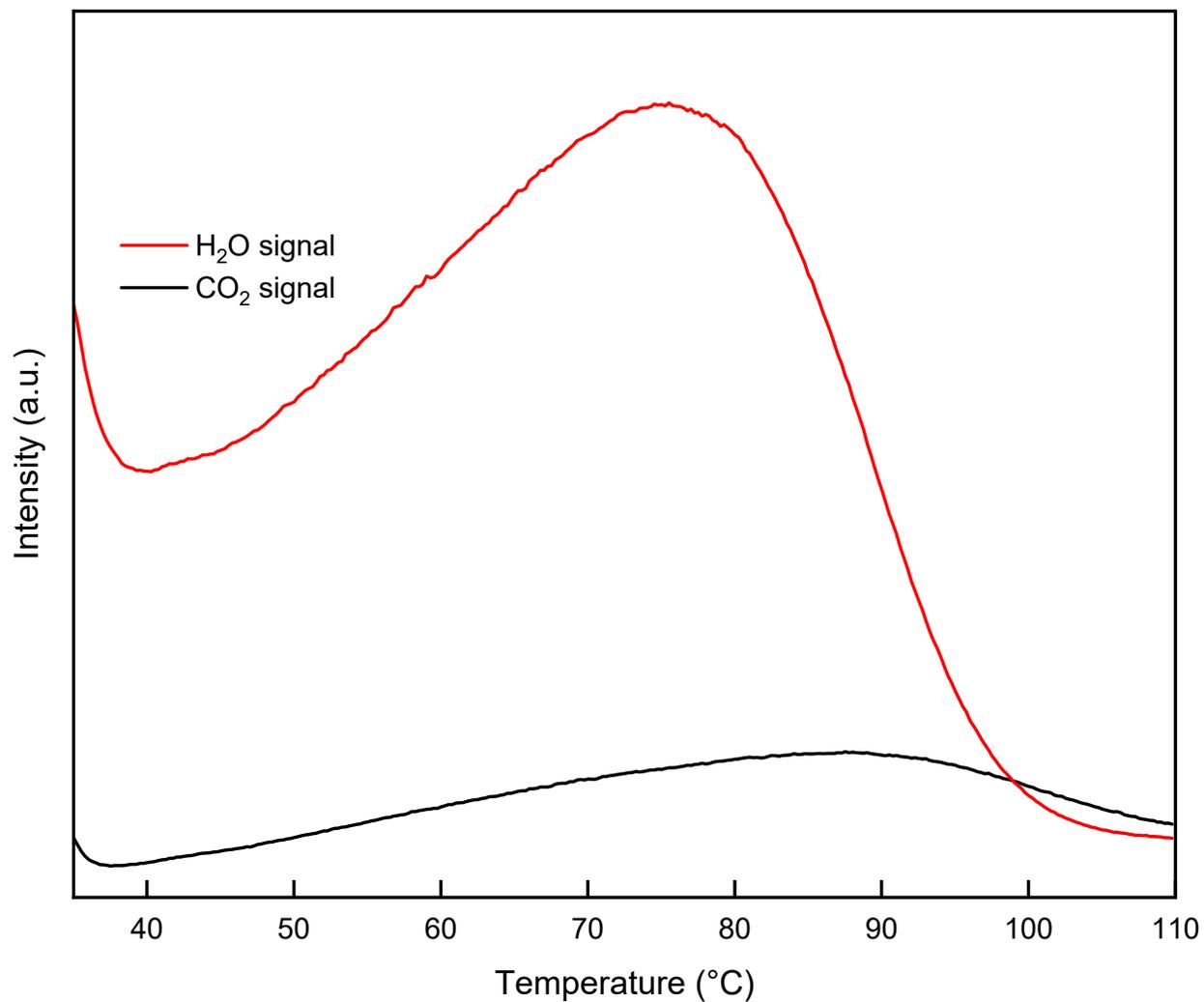
**Figure S20** Changes in IR absorption for DEA/SiO<sub>2</sub> at different ageing days. Accelerated ageing conditions: 85 °C in air.

## Supplementary Figure 21



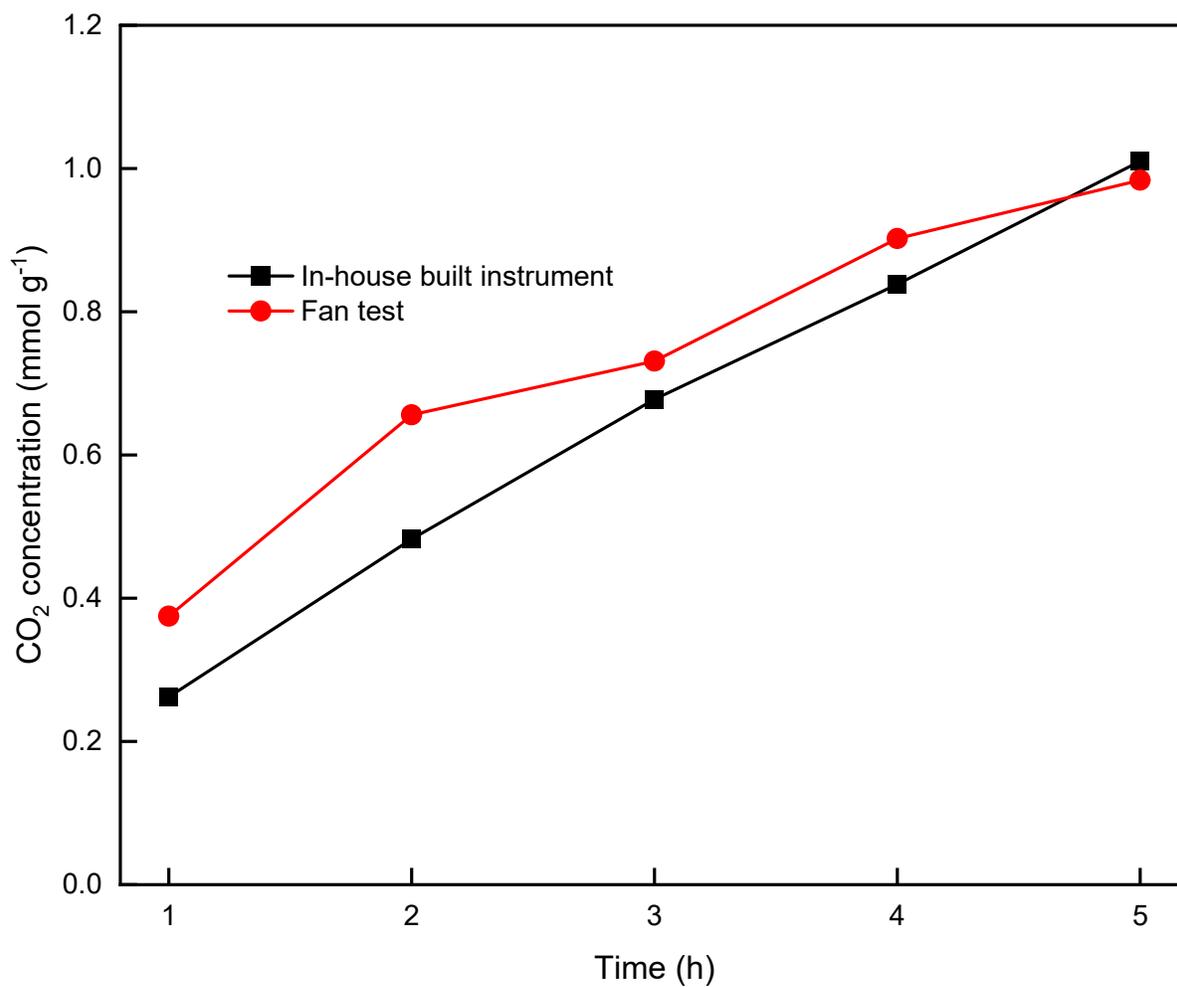
**Figure S21** Changes in IR absorption for PEI800/SiO<sub>2</sub> at different ageing days. Accelerated ageing conditions: 85 °C in air.

### Supplementary Figure 22



**Figure S22** TGA-MS analysis to measure the CO<sub>2</sub> and H<sub>2</sub>O desorption profiles for 0.5DEA&0.5TEA@SAN.

### Supplementary Figure 23



**Figure S23** CO<sub>2</sub> uptake from the in-house built instrument compared with the amount of CO<sub>2</sub> desorbed from samples being exposed to outdoor ambient air at different durations. The sample used was 0.5DEA&0.5TEA@SAN.