

Electronic Supplementary Information

Carbon Capture with Polyethylenimine hydrogel beads (PEI HBs)

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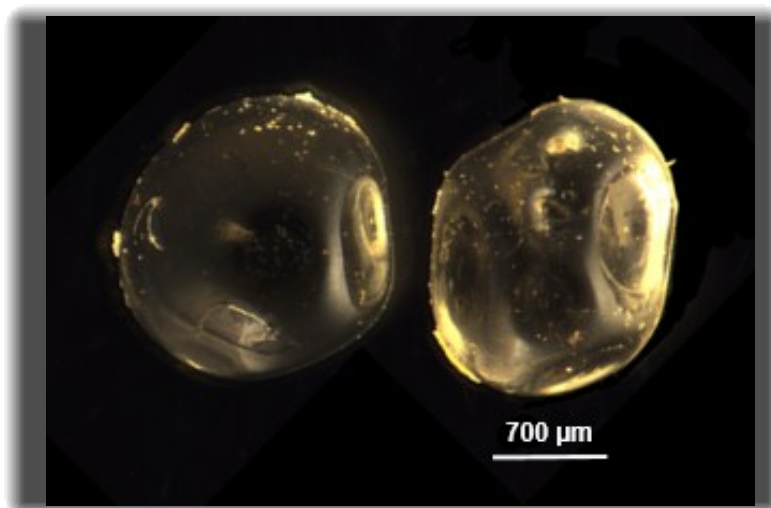


Fig. S1 Optical image of the dry PEI HBs with an EPC mass concentration of 4.0%.

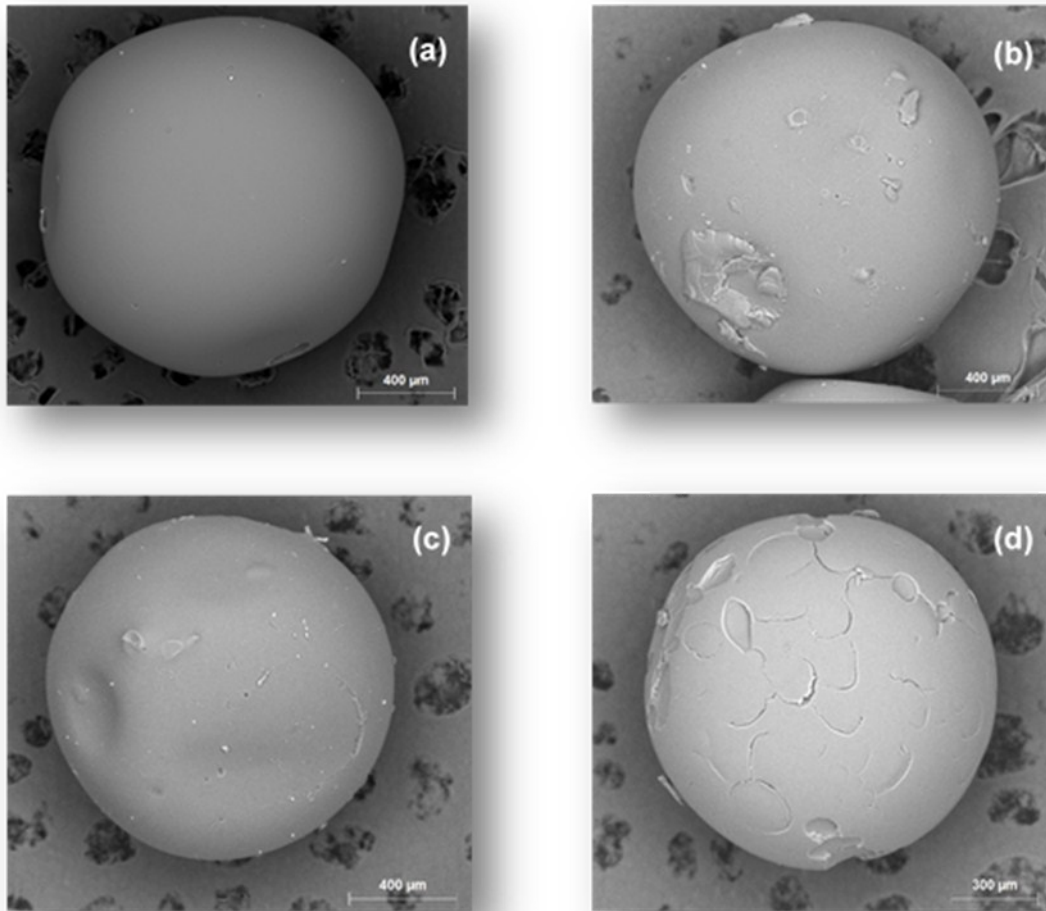
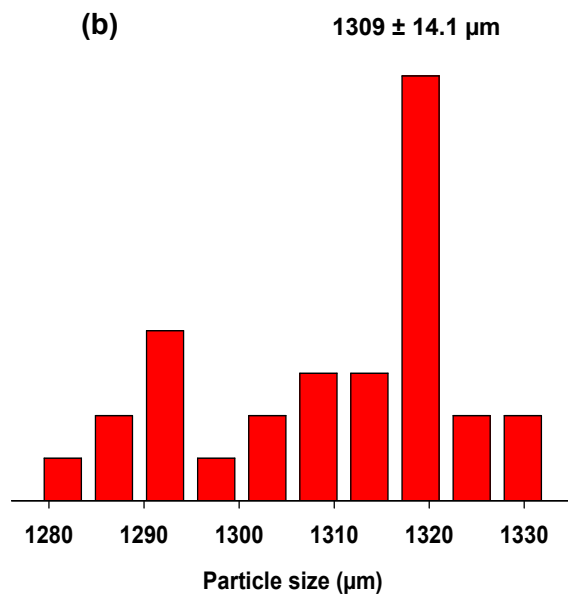
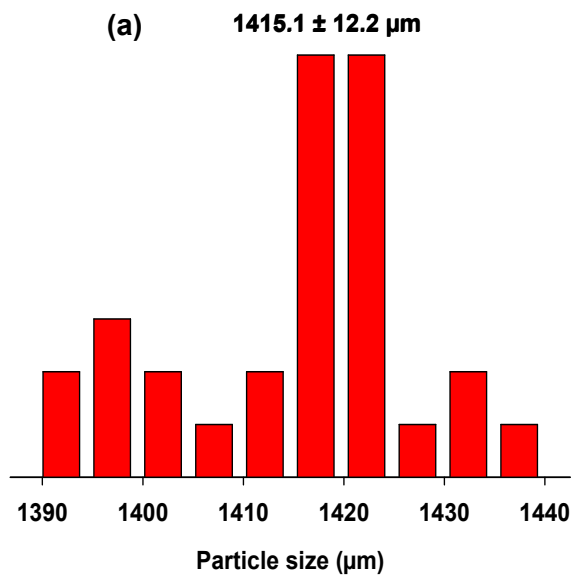


Fig. S2 Typical SEM images of PEI HBs with 2.0 wt% (a), 4.0 wt% (b), 6.0 wt% (c), and 8.0 wt% (d) of EPC.



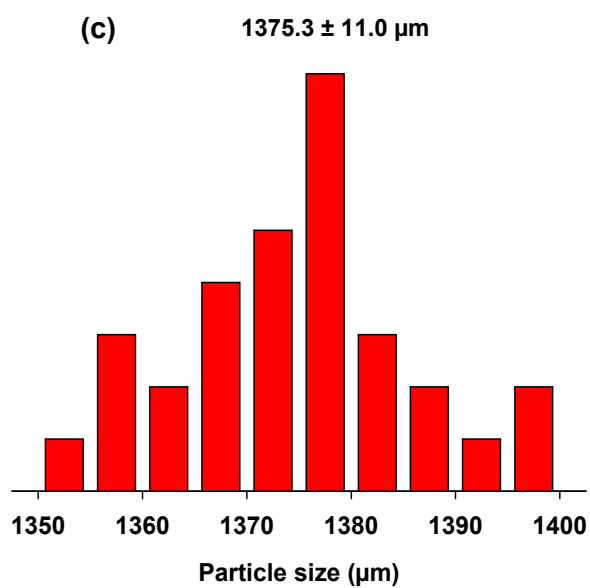


Fig. S3 Particle size distribution of PEI HBs with various amounts of EPC. (a) 4.0 wt%; (b) 6.0 wt%; (c) 8.0 wt%. The sizes of PEI HBs were analysed by the software Image J counting 100 particles.

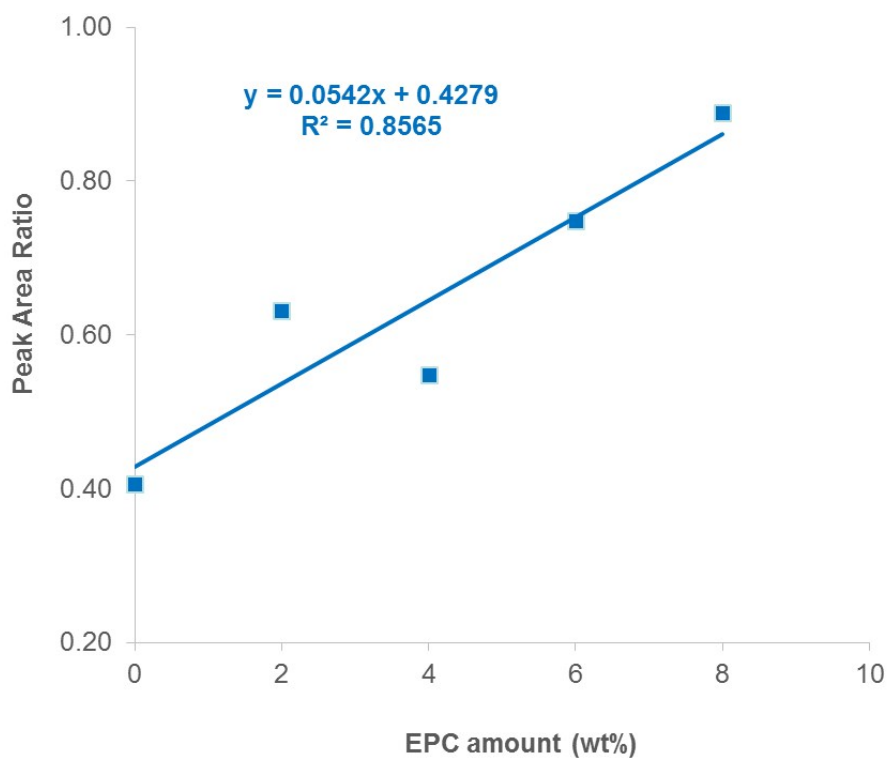
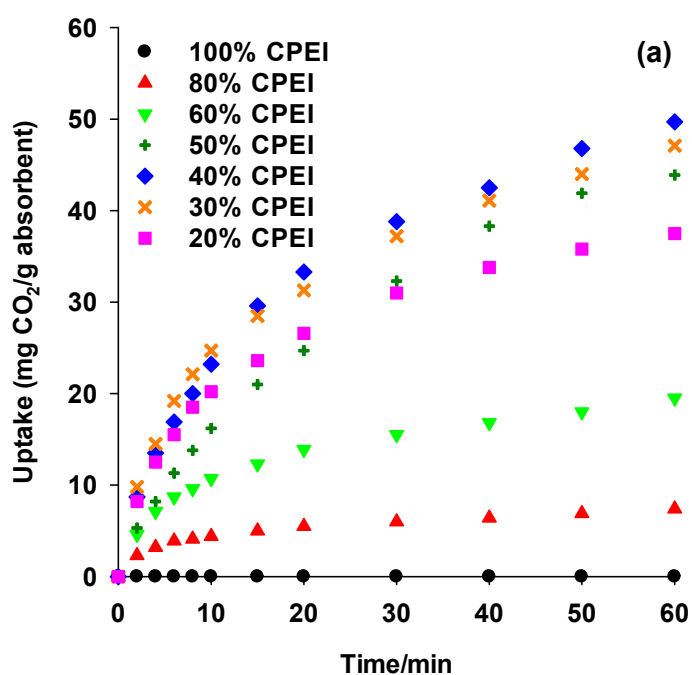
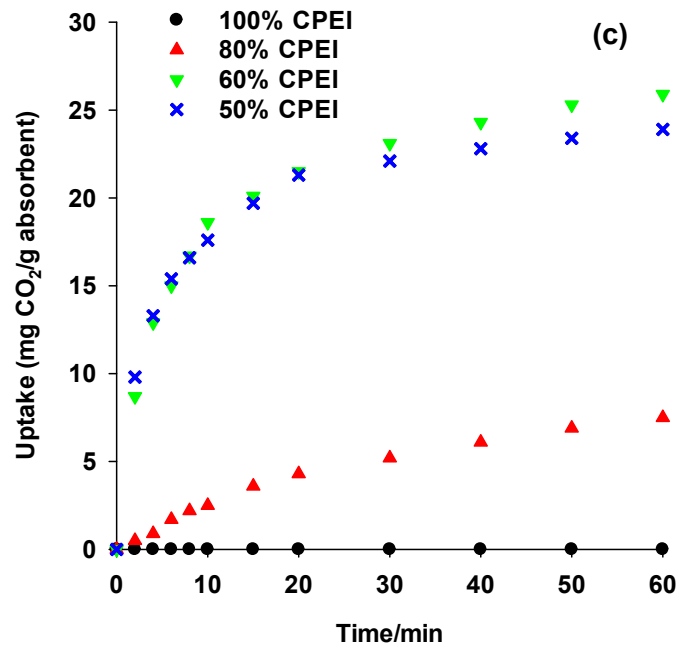
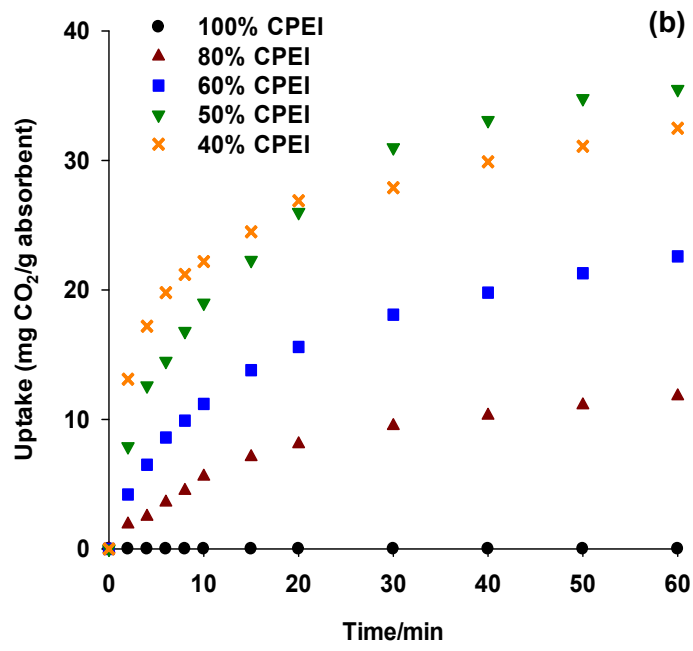
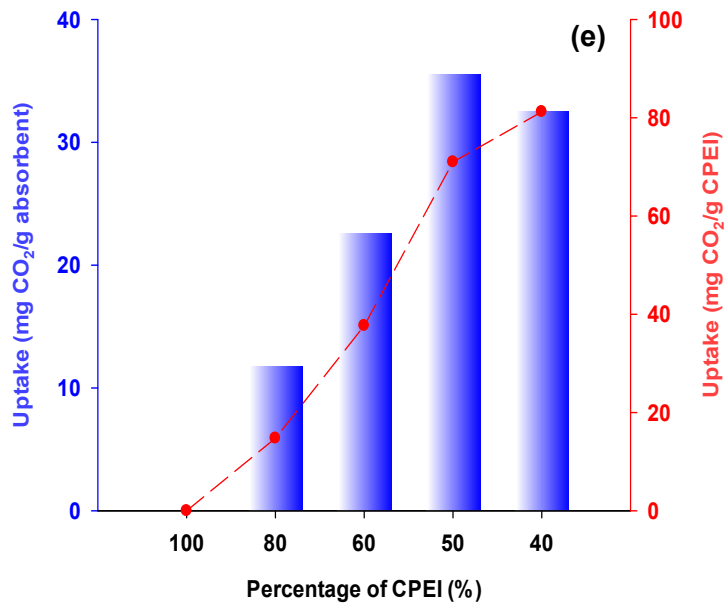
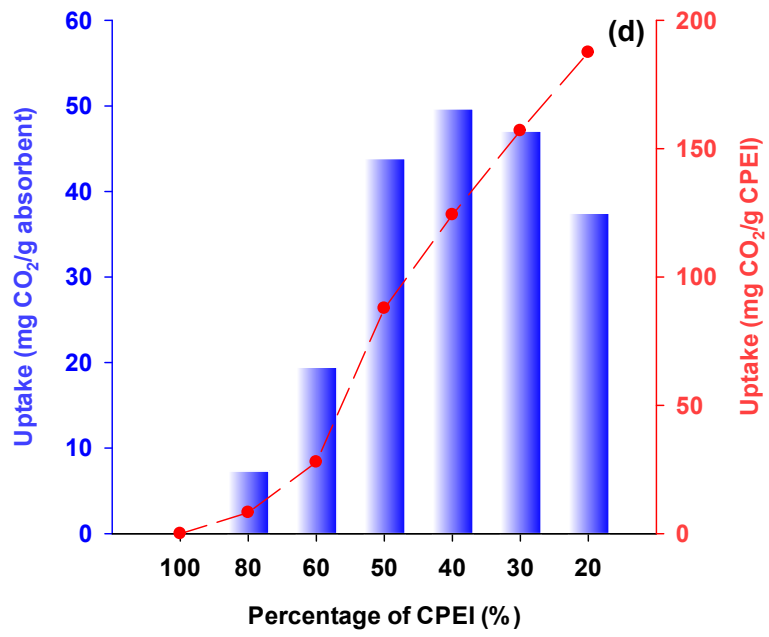


Fig. S4 Dependence of (C-N/C-H) ratio on cross-linker amount. The peaks corresponding to the C-H and C-N stretching vibrations were integrated and the ratio (the C-N band at $\sim 1100\text{ cm}^{-1}$ versus the C-H band at $\sim 3000\text{ cm}^{-1}$) was determined in order to see how the number of C-N bonds in the hydrogel is changing with cross-linker amount. It is evident that the proportion of the C-N band increases relative to the C-H band and that the change is almost linear ($R^2 \sim 0.85$). This confirms that the cross-linker is reacting with the PEI.







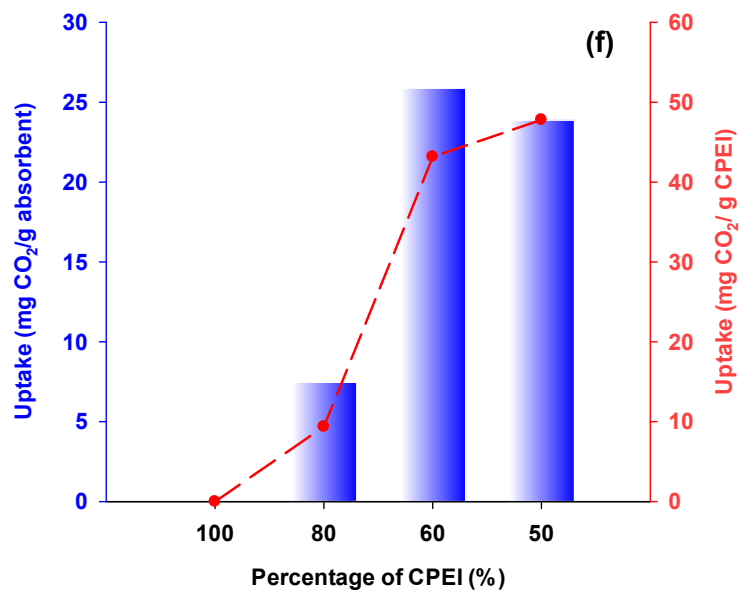


Fig. S5 CO₂ uptake of PEI HBs with various EPC amounts. (a) 2.0 wt%; (b) 6.0 wt%; (c) 8.0 wt %. Summary of the working capacity based on both absorbent mass and PEI mass of PEI HBs with various EPC amounts. (d) 2.0 wt%; (e) 6.0 wt%; (f) 8.0 wt%.

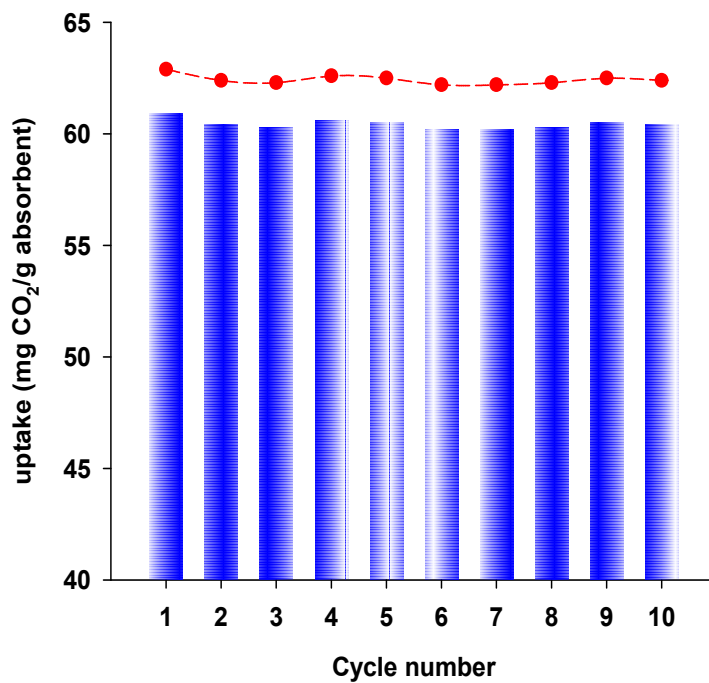


Figure S6. Recyclability of PEI HBs-4.0 % EPC with 70% water (regenerated by convective oven at 160°C).

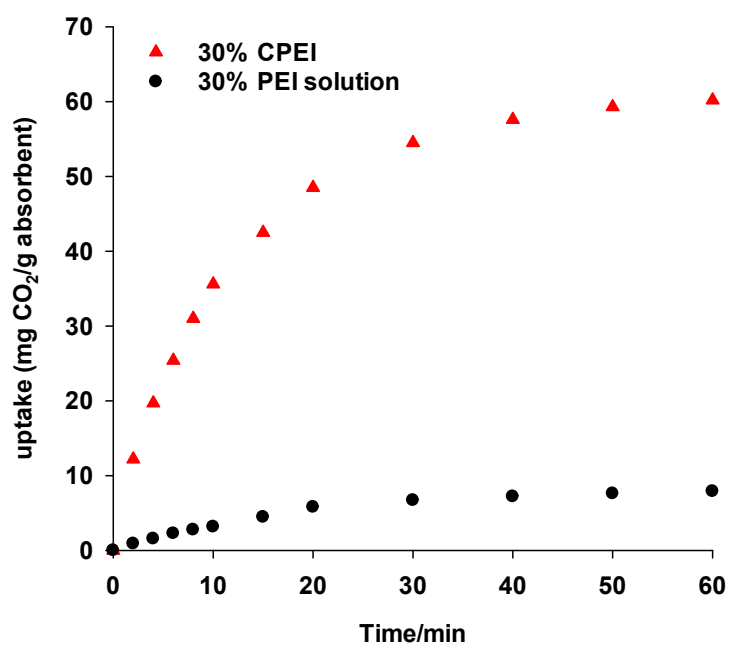


Fig. S7 CO₂ uptake of wet PEI HBs-4.0 wt% EPC with a CPEI concentration of 30% and PEI solution with a weight concentration of 30%.

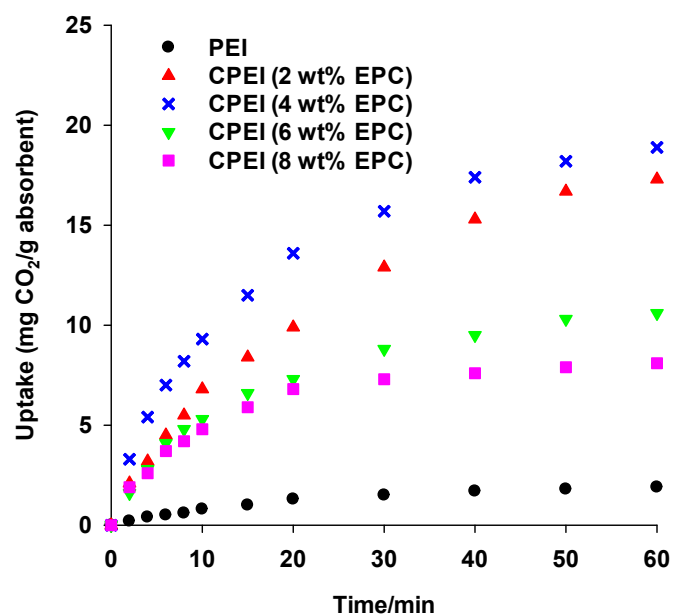


Fig. S8 CO₂ uptakes of PEI solution and PEI HBs under CO₂/N₂ mixture (15% CO₂ and 85% N₂ by volume). The PEI solution has a concentration of 50 wt%. PEI HBs contain 50 wt% of CPEI with various cross-linker amounts.

Table S1 FTIR band assignments for the absorbed carbon dioxide into PEI HBs-4.0 % EPC with a water content of 70% (Peaks are indicated in Fig. 5b)

Peak	Wavenumber (cm^{-1})	Assignment	Reference
1	1553	COO ⁻ stretch (carbamate)	[1]
2	1472	COO ⁻ stretch or HN-C=O (carbamate)	[1, 2]
3	1414	C-N stretch / NCOO ⁻ skeletal vibration (carbamate)	[1]
4	1365	CO ₃ ²⁻ /HCO ₃ ⁻ (carbonate/bicarbonate)	[2]
5	1292	NCOO ⁻ skeletal vibration (carbamate)	[1, 2]

References

[1] Wilfong, W.C., Srikanth, C.S., and Chuang, S.S.C., In situ ATR and DRIFTS studies of the nature of adsorbed CO₂ on tetraethylenepentamine films, *ACS Applied Materials and Interfaces*, 2014, 6(16), 13617-13626.

[2] Jackson, P., Robinson, K., Puxty, G., and Attalla, M., In situ Fourier Transform-Infrared (FT-IR) analysis of carbon dioxide absorption and desorption in amine solutions, *Energy Procedia*, 2009, 1, 985-994.