Supporting Information

Applicability of linear polyethylenimine supported on silica for the adsorption of CO_2 from various sources including dry air.

Table S1. LPEI content in FS-LPEI(x) for the study of the effect of molecular weight on the adsorption characteristics and heat of adsorption/desorption measurements by DSC.

	% LPEI in FS-LPEI(x)
LPEI(2500)	45.5
LPEI(5000)	47.8
LPEI(10000)	48.7
LPEI(25000)	44.5

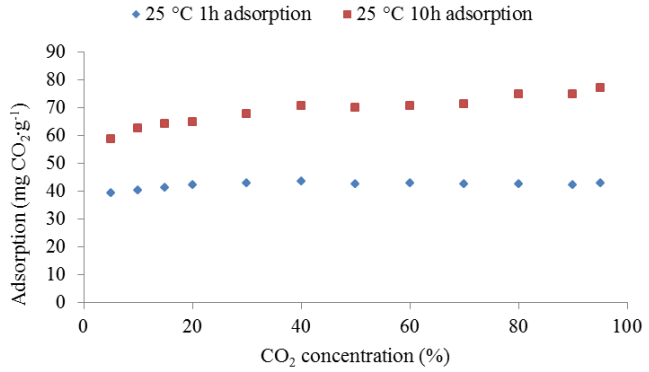

Figure S1. Effect of adsorption time on the adsorption capacity at 25 °C on FS-LPEI(5000)-45.

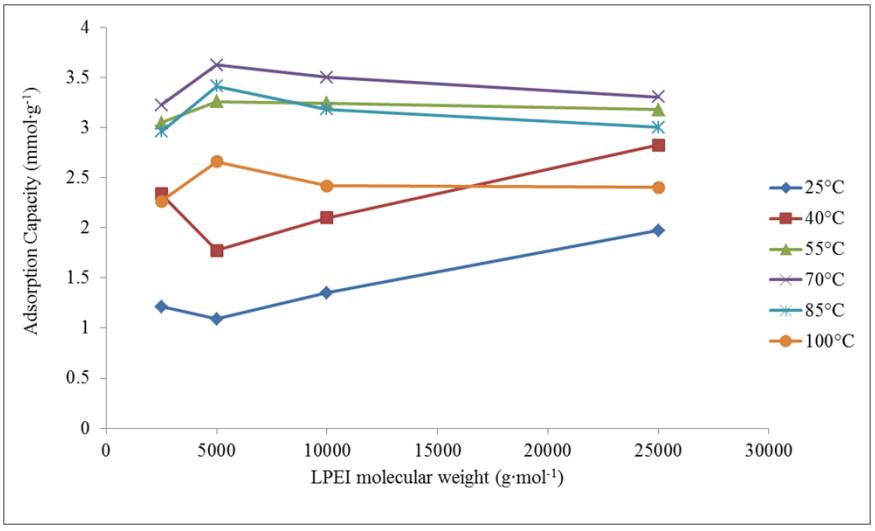

Figure S2. Effect of LPEI molecular weight on the CO_2 adsorption capacity from a gas mixture containing 95% CO_2 in N_2.

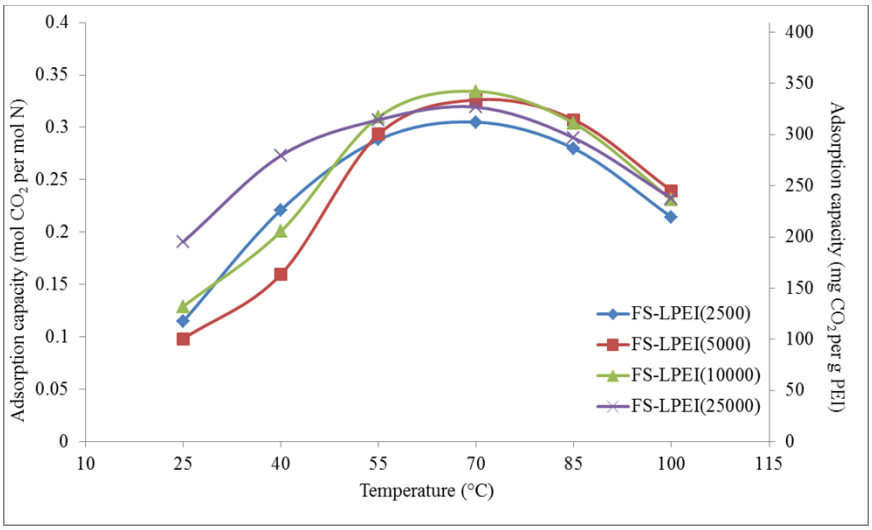

Figure S3. Efficiency of PEI utilization for CO_2 adsorption from a gas mixture containing 95% CO_2 in N_2 on FS-LPEI with various LPEI molecular weights.

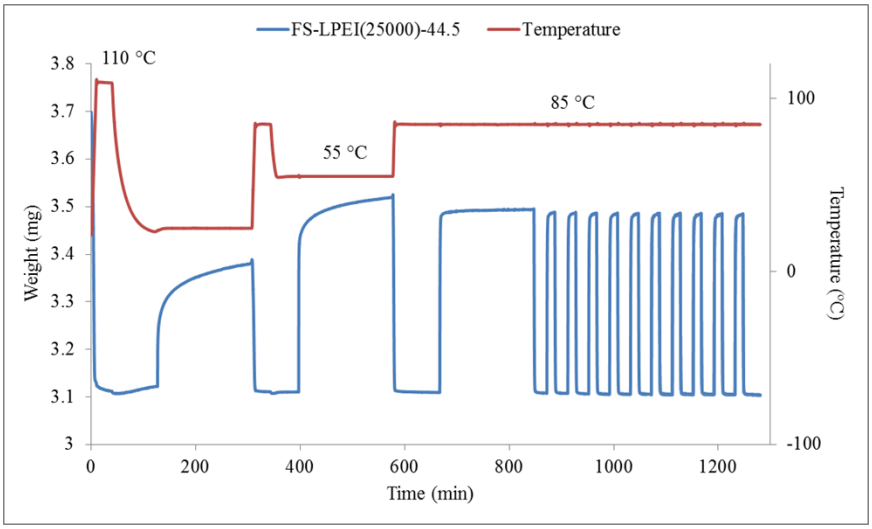

Figure S4. Example of TGA experiment for CO_2 adsorption on FS-LPEI(25000)-44.5. Adsorption under 95% CO_2 at 25, 55, and 85 °C for 3 h. Desorption under pure nitrogen at 85 °C. 10 short cycles of adsorption/desorption followed (adsorption under 95% CO_2 for 15 min, desorption under pure N_2 for 25 min).

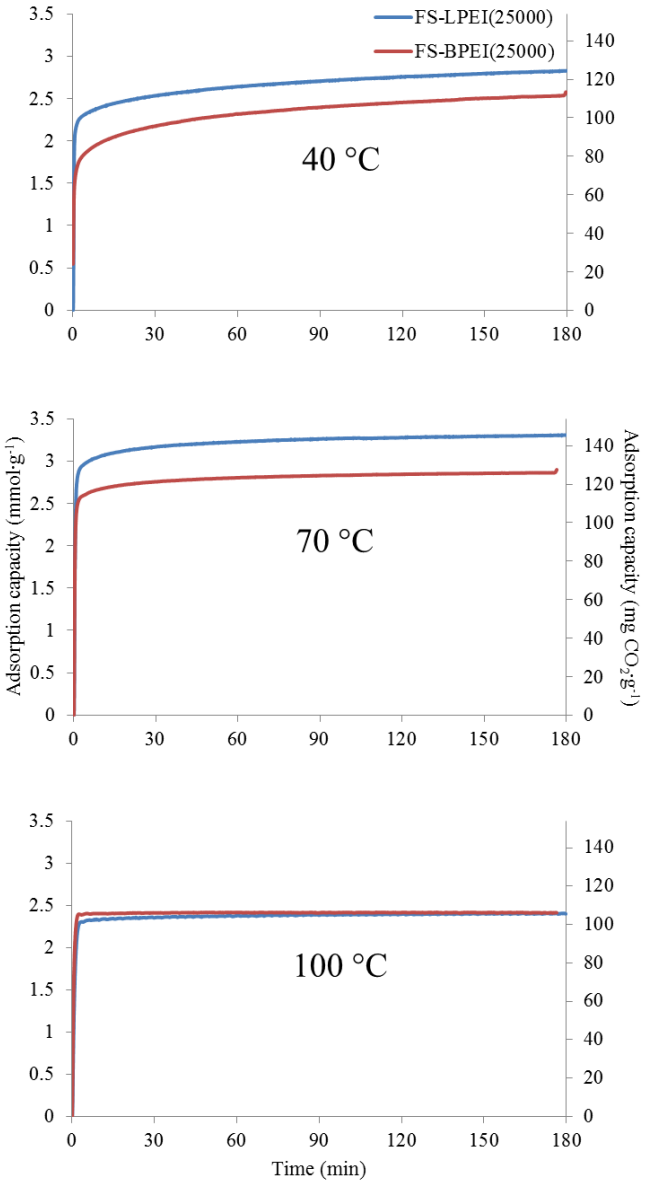

Figure S5. Comparison of CO_2 adsorption kinetics on linear and branched FS-PEI(25000) at 40, 70, and 100 °C.

Table S2. Adsorption capacities at various temperatures based on LPEI and BPEI

	25 °C	40 °C	55 °C	70 °C	85 °C	100 °C
FS-LPEI	83.01	124.31	131.09	145.63	123.40	105.76
FS-BPEI	88.84	111.51	118.84	126.04	125.35	105.66

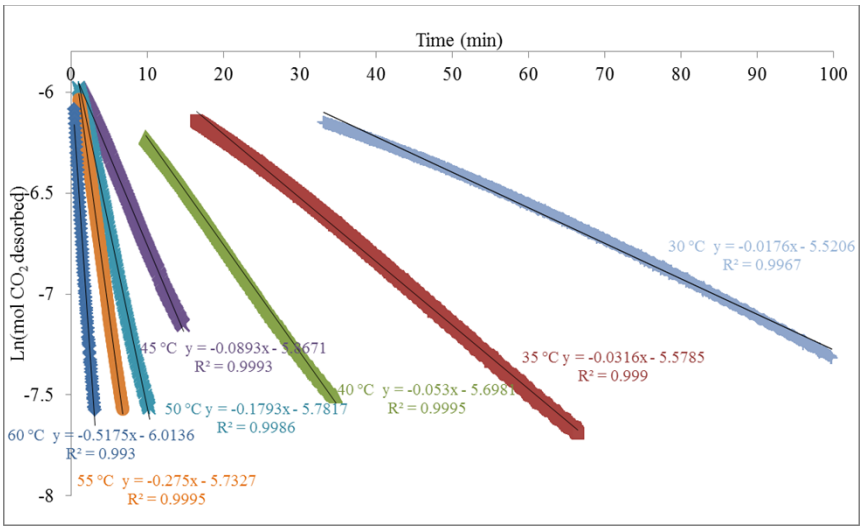

Figure S6. Ln of the amount of CO_2 desorbed as a function of time at various temperatures on FS-LPEI(5000)-48.

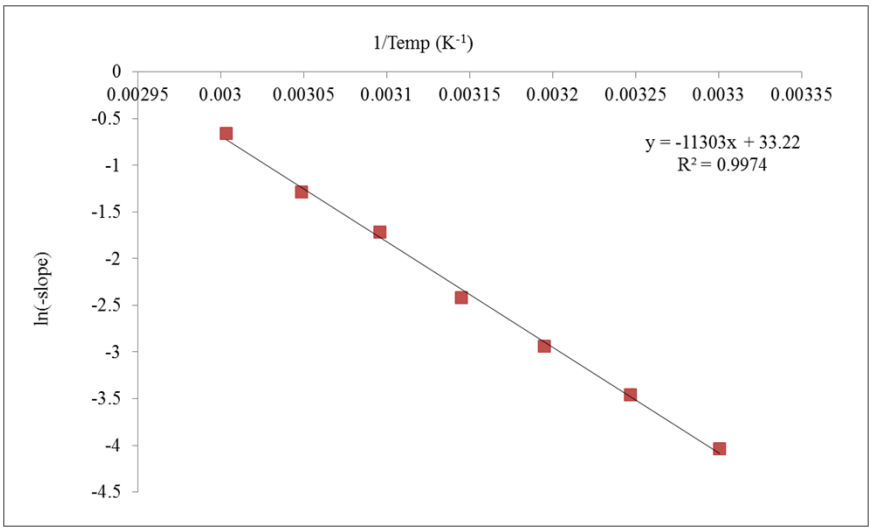

Figure S7. Arrhenius plot for the CO_2 desorption on FS-LPEI(5000)-48.

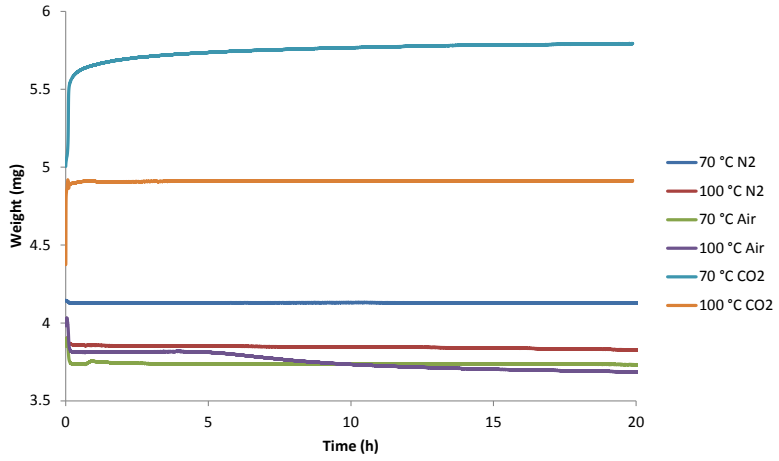

Figure S8. Weight of FS-LPEI(5000)-47.8 samples as a function of time under an atmosphere of N_2, air or CO_2 at 70 and 100 °C (raw data from the TGA).

Physical characteristics of FS-LPEI(5000)

The physical characteristics of one of the adsorbents containing linear PEI, namely FS-LPEI(5000) was measured. Nitrogen adsorption/desorption isotherms were measured at 77 K with a Quantachrome NOVA 2200e surface area and pore volume analyzer. The specific surface area was calculated by the multipoint Brunauer–Emmett–Teller (BET) method. The total pore volume was evaluated at a P/P_0 close to 0.995.The Barrett–Joyner–Halenda (BJH) method was used to calculate the pore volume and pore size distribution by using the desorption branch of the isotherm.

While fumed silica did not have well defined pore structure,[1, 2] FS-LPEI(5000) showed a more defined pore distribution with a peak in pore diameter around 350 Å. This distribution was very similar to the one observed earlier with branched PEI.[2]

Surface area (BET)= 34.1 m^2/g

Total pore volume = 0.445 cm^3/g

Average pore diameter (BJH, desorption branch): 354 Å

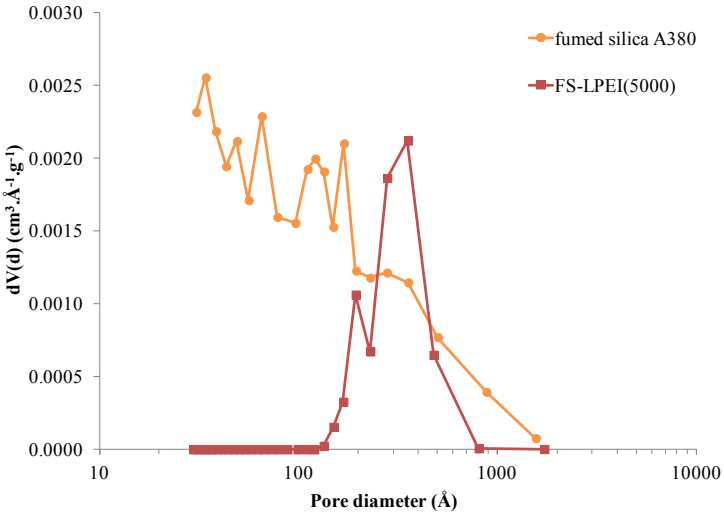

Figure S9. Pore size distribution in fumed silica and FS-LPEI(5000)-47.8 (BJH, desorption branch).

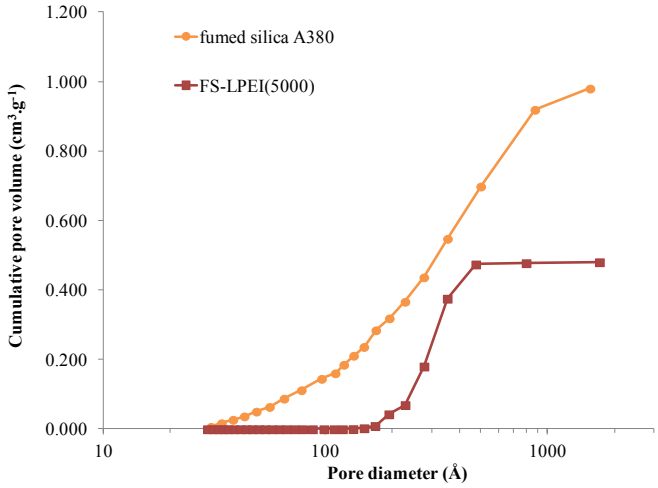

Figure S10. Cumulative pore volume as a function of pore diameter in fumed silica and FS-LPEI(5000)-47.8 (BJH, desorption branch).

Melting points and other characteristics of LPEI(x)

LPEI(2500), bought from Aldrich (product #764604, Batch # MKBL7818V), white powder, melting point: 59-64 °C, poly dispersity (PDI, MW/MN) of 1.1, molecular number (GPC): 2190.
LPEI(5000), bought from Aldrich (product #764582, Batch # MKBL7822), white chunks, melting point: 54-59 °C, poly dispersity (PDI, MW/MN) of 1.1, molecular number (GPC): 5015.
LPEI(10000), bought from Aldrich (product #765090, Batch # MKBL7820), white chunks, melting point: 48-53 °C, poly dispersity (PDI, MW/MN) of 1.2, molecular number (GPC): 9688.
LPEI(25000), bought from Alfa Aesar (product # 43896, lot B04Z069), melting point: 59-60 °C
MW of 25000, (PDI, MW/MN) ≤ 1.3.

References

1 G. A. Olah, A. Goeppert, S. Meth and G. K. S. Prakash, *Energ. Environ. Sci.*, 2010, **3**, 1949-1960.
2 A. Goeppert, H. Zhang, M. Czaun, R. B. May, G. K. S. Prakash, G. A. Olah and S. R. Narayanan, *ChemSusChem*, 2014, **7**, 1386-1397.