## **Supporting Information**

## for

## Impregnation of Polyethylenimine in Mesoporous Multilamellar Silica Vesicles

for CO<sub>2</sub> Capture: A Kinetic Study

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Figure S1 (a) TG profile (MMSV(a)-PEI-60%) for dry CO<sub>2</sub> adsorption and desorption at 90 °C and (b) TG profile (MMSV(a)-PEI-60%) for humid CO<sub>2</sub> (30%RH) adsorption and desorption at 90 °C



Figure S2 SEM image of MMSV(c)



Figure S3 (a) TG and (b) DTG curves for MMSV(a), DTAB, DHDAB and MMSV(c) with a temperature ramp of 10  $^{\circ}$ C/min in pure N<sub>2</sub>



Figure S4 TG (a) and DTG (b) curves for MMSV(c)/PEIs with a temperature ramp of 10 °C/min in pure  $N_2$ 



Figure S5 Nitrogen adsorption/desorption isotherms of MMSV-PEIs



Figure S6 Repeating demonstrations of  $CO_2$  dynamic adsorption on MMSV(a)-PEI-60%

The CO<sub>2</sub> capacities are 4.68, 4.73, and 4.59 mmol/g, respectively, and the standard deviation is calculated as  $\pm 0.058$ .



Figure S7  $CO_2$  adsorption capacity of MMSV(a)/PEIs with different amine loadings, Pure PEI and MMSV(a) at 90 °C under 20 mL/min flow rate of pure  $CO_2$  in TGA



Figure S8 (a) Comparison of dynamic CO<sub>2</sub> adsorption in isothermal conditions for MMSV-PEI-60% at adsorption temperatures of 60, 75, 90 and 100 °C under 20 mL/min flow rate of pure CO<sub>2</sub> in TGA, (b) CO<sub>2</sub> adsorption capacity of MMSV(a)/PEIs with different amine loadings at adsorption temperatures of 60, 75, 90 and 100 °C under 20 mL/min flow rate of pure CO<sub>2</sub> in TGA and (c) Fitting plots of  $t/q_t$  against t as predicted by the second-order rate law



Figure S9 Cyclic adsorption/desorption of MMSV(a)-PEI-60% under dry pure CO<sub>2</sub> at 90  $^{\circ}\text{C}$ 

Samples	$S_{BET}(m^2/g)$	Volume(cm <sup>3</sup> /g)
MSMV(c)	797.7	0.857
MSMV(c)-PEI-40%	39.7	0.13
MSMV(c)-PEI-50%	16.9	0.034
MSMV(c)-PEI-60%	9.5	0.016
MSMV(c)-PEI-70%	3.4	0.005
MSMV(a)	43.9	0.192
MSMV(a)-PEI-40%	10.9	0.049
MSMV(a)-PEI-50%	2.37	0.001
MSMV(a)-PEI-60%	0.09	

Table S1 Pore properties of MMSV-PEIs

Materials	CO <sub>2</sub> capacity (mmol/g)	Amine efficiency* (%)	Refs.
MCM-41/75%PEI	3.02	37.1	[1]
MCM-48/50%PEI	2.70	46.4	[2]
SBA-15/55%PEI	3.93	61.4	[3]
SBA-16/50%PEI	2.93	50.4	[2]
KIT-6/50%PEI	3.07	52.8	[2]
HMS/60%PEI	4.18	59.9	[4]
PE-MS/70%PEI	4.95	60.8	[5]
MCF/50%PEI	4.09	70.4	[6]
Monolith/65%PEI	4.77	63.1	[7]
Mesoporous capsules/83%PEI	5.70	59.1	[8]
Silica foam/83%PEI	5.80	60.1	[9]
Spherical silica foams/56.5%	4.27	65.0	[10]
MMSV(a)-PEI-60%	4.73	67.8	This work

**Table S2** CO<sub>2</sub> adsorption capacity and amine efficiency of various sorbents of porous silica with PEI impregnated

\* The amine efficiency was defined as the ratio of mol  $CO_2$  captured per 2 mol of amino groups in the sorbent and expressed as a % of the maximum adsorption capacity

Sample	CO <sub>2</sub> capacity (mmol/g)	Amine efficiency (%)
MMSV(a)-PEI-40%	2.57	55.3
MMSV(a)-PEI-50%	3.14	54.6
MMSV(a)-PEI-60%	3.85	55.1
MMSV(a)-PEI-70%	1.95	23.8

Table S3 CO<sub>2</sub> capacity of MMSV(a)/PEIs at the simulated flue gas (with 15:85 v/v  $CO_2/N_2$  and dry) at 90 °C

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