# Supporting Information

# **Carbon Dioxide Capture from the Air Using a Polyamine Based Regenerable Solid Adsorbent**

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#### **Reagents and supports**

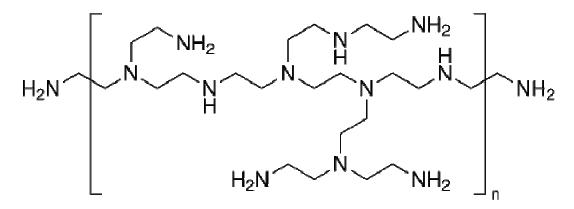
PEI (HMW) high molecular weight (weight average molecular weight  $M_w$  ca. 25000, determined by LS,  $M_n$  ca 10000 determined by GPC) was purchased from Aldrich. Fumed silica Aerosil® 380 (average primary particle size 7 nm) was obtained from Evonik (formerly Degussa).

### **Preparation of adsorbents**

The following example illustrates the preparation of a supported amine sorbent composed of 50 wt. % polyethylenimine and 50 wt. % fumed silica.

25 g of polyethylenimine (HMW) was dissolved in 300 mL of methanol. This solution was then added stepwise under stirring to 25 g of fumed silica suspended in 800 mL methanol to ensure a good dispersion of the polyethylenimine on the support. The solution was then mixed for an additional three hours. After that, the solvent was removed from the mixture by heating at 50°C under vacuum on a rotovapor followed by vacuum treatment overnight (< 1 mm Hg). The supported amine sorbent was obtained as a white solid.

PEI (HMW) structure



Measurement of CO<sub>2</sub> adsorption and desorption capacity

CO<sub>2</sub> adsorption data were obtained using an all-glass grease free flow system. The adsorbent, generally 3 g was placed in a U-tube between two glass wool plugs. The inner diameter of the tube was 8 mm. The height of the adsorbent bed for FS-PEI-50 was about 10 cm. The U-tube was then evacuated (~65 mTorr) at 85°C for 3 hours. The weight of the adsorbent after this treatment was measured. The weight loss due to CO<sub>2</sub> and water desorption, depending on the adsorbent, was generally between 2 and 10%. The FS-PEI-50 used to obtain Figure 1 had for example a weight of 2.72 g after vacuum treatment due to a weight loss of 9.45%. The adsorbent weight after pretreatment was used for the later calculation of the CO<sub>2</sub> adsorption capacities. After pretreatment the adsorbent containing U-tube was placed in a thermostated oil bath at 25 °C. Air from the laboratory atmosphere was dried (over silica gel), filtered and compressed to 150 psi and used directly for the adsorption measurements. For the adsorption measurements a Horiba VIA-510 CO<sub>2</sub> analyzer equipped with an IR detector specifically intended for CO<sub>2</sub> measurements was placed in-line with the adsorption setup. The concentration range used was 0-2000 ppm. Before each run, the analyzer was calibrated with reference gases; CO<sub>2</sub> in air and ultra zero grade air for the zero. After calibration, the initial CO<sub>2</sub> concentration in air was determined and was generally between 410 and 420 ppm; somewhat higher than the average global atmospheric CO<sub>2</sub> concentration of 390 to 395 ppm. The air flow (335 mL/min) was then opened on the adsorbent bed. Almost immediately the CO<sub>2</sub> concentration in the gas outlet fell to 0 ppm, signaling complete CO<sub>2</sub> adsorption from the air. The CO<sub>2</sub> concentration was recorded as a function of time via LabView 8.6. After saturation of the adsorbent, when the CO<sub>2</sub> concentration reached the inlet value (410-420 ppm), the gas flow was stopped.

Desorption was performed using two different methods:

a) By applying vacuum (~65 mTorr) at 85 °C for 3 hours

b) By heating the adsorbent containing U-tube to 85 °C and then passing a flow of air (335 ml/min) through it. In this case the outlet gas was analyzed on a Horiba VIA-510 CO<sub>2</sub> analyzer with a range of 0-20% CO<sub>2</sub>. The CO<sub>2</sub> concentration was recorded as a function of time via LabView 8.6. Immediately after opening of the air flow onto the saturated adsorbent the concentration in CO<sub>2</sub> spiked to 3-5% CO<sub>2</sub> and then slowly decreased until reaching the inlet CO<sub>2</sub> concentration (410-420 ppm).

For CO<sub>2</sub> adsorption measurements under humid conditions, a part of the air feed was saturated with water by bubbling through a water-containing saturator thermostated at 25 °C. This moisture saturated feed was then combined with a dry air feed in order to obtain a gas flow with a relative humidity of 67%. The obtained humid air was flown through the adsorbent.

## Surface area and pore volume analysis

The supports were characterized by  $N_2$  adsorption/desorption isotherm measurements on a Quantachrome NOVA 2200e instrument. The surface area was determined by the multipoint BET method. The total pore volume was evaluated at a P/P<sub>0</sub> close to 0.995.