Porous Carbon Pellets for Physical Adsorption of CO2: Size and Shape Effect

Baljeet Singh,* Marianna Kemell, Timo Repo^{*} Department of Chemistry, University of Helsinki, FI-00014 Helsinki, Finland <u>baljeet.singh@helsinki.fi</u> <u>timo.repo@helsinki.fi</u>

Materials and Methods:

Materials

The chemicals used in this study were analytic grade. Porous activate charcoal (242276, DARCO, 100 mesh particle size, powder), CaCl₂, polyacrylic acid and sodium alginate were purchased from Sigma Aldrich and used as received.

Characterization

The surface morphology of the samples was studied using a field emission SEM (Hitachi S-4800).

Thermal stability of pellets was measured using by control heating pellets from 25 to 300 $^{\circ}$ C using rate of 10 $^{\circ}$ C min⁻¹ under the flow of air.

For CO₂ capture (from 15% CO₂ in N₂), the Mettler-Toledo TGA/DSC-3+ thermal analysis system was used. Initially, samples were activated under the flow of N₂ gas (50 mL min⁻¹) for 40 min at 100 °C (5 min RT stabilizing period, 25 to 100 °C heating with rate of 10 °C min⁻¹, isotherm at 100 °C for 15 min, followed by 100 to 25 °C cooling. Before exposing to CO₂, samples were stabilized for 5 min stabilizing. Initially, CO₂ flow was 50 mL min⁻¹ (otherwise mentioned), and desorption was performed at 60 °C for 15 min, under N₂ as a sweep gas (flow: 50 mL min-1).

The specific surface area and textural properties were measured by N2 sorption at 77K using a Quantachrome analyser.

Spherical silica pellet design: Pellets/beads were designed using sodium alginate as a binder. A stock solution of sodium alginate was prepared by dissolving 20 mg mL⁻¹ in water. While curing solution was prepared by dissolving 188 mg CaCl₂ in 30 mL of 50 mmol L⁻¹ polyacrylic acid aq. solution. 1 g carbon powder mixed properly with 4 ml sodium alginate solution and suspension was dropped in a curing solution. After shaping the pellets were collected, washed with water and ethanol, and dried under vacuum at RT.

Cylindrical pellet design: Same proportion (a standard mixture of carbon powder and alginate solution) was used as spherical pellet design and instead of dropping spherical drop in curing solution, a plastic syringe was used to prepare solid cylindrical pellets. After shaping into desired shape, pellets were soaked into curing solution overnight. Pellets were separated washed with ethanol and dried under vacuum at RT.

Hollow cylindrical pellet design: A hole of varying inner diameter was made manually using a stainless-steel needle of varying diameter through the solid cylindrical pellets before curing. After shaping into desired shape hollow cylindrical shape, pellets were soaked into curing solution overnight. Pellets were separated washed with ethanol and dried under vacuum at RT.

Flack's design: A mixture of porous carbon, and sodium alginate was kept for RT drying and a thin layer later separated from glass vial, soaked in curing solution and dried under vacuum. Thickness was varied by using different volume of mixture of porous carbon and sodium alginate standard solution as used in spherical pellets design.

Selectivity calculation: Selectivity was calculated based on the amounts of CO_2 and N_2 capture under the same adsorption conditions using following formula:

Selectivity (%) = $[CO_2 \text{ adsorption capacity (wt%)}/N_2 \text{ adsorption capacity (wt%)}]/(P1/P2)$

P1 and P2 are partial pressure of CO_2 (0.15 atm) and N_2 (0.85 atm)



Figure S1. Vernier clipper images with pellets size.



Figure S2. N₂ sorption isotherm of P3 pellets



Figure S3. DFT pore size distribution of P3 pellets.



Figure S4. CO₂ adsorption TGA profile of P1.



Figure S5. CO₂ adsorption TGA profile of P2.



Figure S6. CO₂ adsorption TGA profile of P3.



Figure S7. CO₂ adsorption TGA profile of P4.



Figure S8. CO₂ adsorption TGA profile of P5.



Figure S9. CO₂ adsorption TGA profile of P6.



Figure S10. CO₂ adsorption TGA profile of P7.



Figure S11. CO₂ adsorption TGA profile of P8.



Figure S12. CO₂ adsorption TGA profile of P9.



Figure S13. CO₂ adsorption TGA profile of P10.



Figure S14. Vernier caliper images with thickness and sizes of different pellets.



Figure S15. CO₂ adsorption TGA profile of P11.



Figure S16. CO₂ adsorption TGA profile of P12.



Figure S17. CO2 adsorption TGA profile of P13.



Figure S18. CO₂ adsorption TGA profile of P14.



Figure S19. CO₂ adsorption TGA profile of P15.



Figure S20. CO₂ adsorption TGA profile of P16.



Figure S21. CO₂ adsorption TGA profile of P17.



Figure S22. CO₂ adsorption TGA profile of P18.



Figure S23. CO₂ adsorption TGA profile of powder sample.



Figure S24. Pseudo first order fitting of powder sample CO₂ adsorption profile.



Figure S25. Adsorption temperature Optimization at constant flow rate (50 ml/min)



Figure S26. CO₂ flow rate optimization at fixed adsorption temperature (30 °C).



Figure S27. CO₂ adsorption -desorption 20 cycles of powder sample (Adsorption at 30 °C, and desorption at 60 °C).



Figure S28. N₂ adsorption -desorption 20 cycles of powder sample (Ads at 30 °C, adsorption desorption at 60 °C).