Post Combustion Carbon Dioxide Capture Using Aqueous Amine Solvents and Amine Impregnated Mesoporous Silica Adsorbents

Satyajit Mukherjee
Supervisor: Prof. Amar N. Samanta

Carbon Capture and Sequestration (CCS) in the energy sector is seen as a bridge technology for CO_2 mitigation, due to the ever-growing environmental impact of anthropogenic emitted greenhouse gases. Aqueous solutions of alkanolamines are the most widely used solvents for regenerative chemical absorption for high volume CO_2 removal. Solid adsorbent based CCS technology has gained momentum as an alternative to the existing amine absorption process for CO_2 removal in specific closed environments' applications (i.e., submarines, space stations, space shuttles). Therefore, the development of amine solvents with high CO_2 absorption rate and capacity with lower regeneration energy and efficient solid adsorbents are needed in CCS application.

In this work, benzylamine (BZA) and 2-(2-aminoethyl-amino)-ethanol (AEEA) based single and blended aqueous solvents are studied in the wide range of temperature and composition to develop effective solvents for the post-combustion CO_2 capture by evaluating the absorption properties (physico-chemical property, kinetics, vapour-liquid equilibrium and absorption enthalpy). Experiential Physico-chemical (density, viscosity, and physical solubility) properties of aqueous amine solvents are correlated using suitable models (Redlich-Kister, Grunberg and Nissan and Jouyban-Acree models for density and viscosity, O'Connell's model for physical solubility). The kinetics of CO_2 absorption is investigated under pseudo-first-order reaction condition using a reaction calorimeter set-up. Second-order rate constants for (BZA-CO_2-H_2O) and (AEEA-CO_2-H_2O) reaction systems are determined using kinetic models based on the zwitterion and termolecular mechanism which are comparable and higher than that of the benchmark MEA solvent. Vapour-liquid equilibrium (VLE) of CO_2 in the aqueous BZA, AEEA and blended (BZA+AEEA) solvent systems are measured using a stirred custom-made equilibrium cell set-up and correlated using Electrolyte Non-Random Two Liquid (ENRTL) and Kent-Eisenberg models. The loading capacity of aqueous BZA and AEEA is found to be comparable and much higher than that of in MEA respectively. The heat of absorption of CO_2 in aqueous BZA, AEEA and their blends are measured to estimate desorption energy using a commercial automated reaction calorimeter and correlated using artificial neural network (ANN) model. The enthalpy of CO_2

absorption in aqueous BZA is almost similar than that of the MEA solvent, but for the blended amine (BZA+AEEA) systems absorption enthalpy is lower due to the presence of the low energy solvent (AEEA). The BZA, MEA and AEEA impregnated MCM-41, MCM-48 adsorbents are characterized using BET, TGA, FT-IR, CHNS, FE-SEM and XRD analysis. The materials present good structural and thermal stability. Automated high-pressure isob-HP2 sorption analyser is used to study the CO_2 adsorption. The MCM-41-40%AEEA and MCM-48-30%AEEA materials are exhibited the highest CO_2 adsorption capacity. The Freundlich, Langmuir, Sips and Toth isotherm models are proposed to correlate the experimental CO_2 adsorption data. The calculated isosteric heat of adsorption using Van't Hoff plot describes the degree of heterogeneity and the surface energy distribution of the adsorbents. This study indicates the potential application of the aqueous BZA and AEEA based absorbents and impregnated mesoporous adsorbents for efficient CO_2 capture.

Key Words: CO_2 capture, VLE, Kinetics, Enthalpy, BZA, AEEA, Adsorption, MCM-41, MCM-48.