

## **Abstract**

Climate change due to the ever-increasing emission of anthropogenic greenhouse gases arising from the use of fossil fuels for power generation and most industrial processes is now a global challenge. It is therefore imperative to develop strategies or modern technologies that could mitigate the effect of global warming due to the emission of CO<sub>2</sub>. Carbon capture and storage (CCS) is a viable option that could ensure the sustainable use of cheap fossil fuels for energy generation with less CO<sub>2</sub> emission. Amongst existing CCS technologies, absorption technology using monoethanolamine (MEA) is very mature and widely embraced globally. However, the absorption technology has a lot of challenges such as, low CO<sub>2</sub> loading, high energy requirement for solvent regeneration, corrosive nature etc. On this note, the adsorption technology using solid sorbents is being considered for CO<sub>2</sub> capture due to its competitive advantages such as flexibility, low energy requirement for sorbent regeneration, non-corrosive nature etc. On the other hand, adsorbents have a very vital role to play in adsorption technology and there is need to understand the behaviour of adsorbents for CO<sub>2</sub> capture under different operating conditions in order to adapt them for wider applications. On this note, the study contained in this dissertation investigated the adsorption behaviour of a novel polymer-based adsorbent (polyaspartamide) during post-combustion CO<sub>2</sub> capture using experimental study and mathematical modelling approach.

Polyaspartamide is an amine-rich polymer widely used in drug delivery. In addition, its rich amine content increases its affinity for CO<sub>2</sub>. Its porosity, thermal stability and large surface area make it a promising material for CO<sub>2</sub> capture. In view of this, polyaspartamide was used as the adsorbent for post-combustion CO<sub>2</sub> capture in this study. This dissertation investigated the kinetic behaviour, the diffusion mechanism and rate limiting steps (mass transfer limitation) controlling the CO<sub>2</sub> adsorption behaviour of this adsorbent. Furthermore, effect of impurities such as moisture and other operating variables such as temperature, pressure, inlet gas flow rate etc. on the CO<sub>2</sub> adsorption behaviour of polyaspartamide was also investigated. Existing mathematical models were used to understand the kinetics and diffusion limitation of this adsorbent during CO<sub>2</sub> capture. Popularly used gas-solid adsorption models namely; Bohart-Adams and Thomas model were applied in describing the breakthrough curves in order to ascertain the equilibrium concentration and breakthrough time for CO<sub>2</sub> to be adsorbed onto

polyaspartamide. Lagergren's pseudo 1<sup>st</sup> and 2<sup>nd</sup> order models as well as the Avrami kinetic models were used to describe the kinetic behaviour of polyaspartamide during post-combustion CO<sub>2</sub> capture. Parameter estimations needed for the design and optimization of a CO<sub>2</sub> adsorption system using polyaspartamide were obtained and presented in this study. The Boyd's film diffusion model comprising of the interparticle and intra-particle diffusion models were used to investigate the effect of mass transfer limitations during the adsorption of CO<sub>2</sub> onto polyaspartamide.

Data obtained from continuous CO<sub>2</sub> adsorption experiments were used to validate the models in this study. The experiments were conducted using a laboratory-sized packed-bed adsorption column at isothermal conditions. The packed bed was attached to an ABB CO<sub>2</sub> analyser (model: ABB-AO2020) where concentrations of CO<sub>2</sub> at various operating conditions were obtained.

The results obtained in this study show that temperature, pressure and gas flow rate had an effect on the adsorption behaviour of polyaspartamide (PAA) during CO<sub>2</sub> capture. Polyaspartamide exhibited a CO<sub>2</sub> capture efficiency of 97.62 % at the lowest temperature of 303 K and pressure of 2 bar. The amount of CO<sub>2</sub> adsorbed on polyaspartamide increased as the operating pressure increased and a decrease in the adsorption temperature resulted in increased amount of CO<sub>2</sub> adsorbed by polyaspartamide. The amounts of CO<sub>2</sub> adsorbed on polyaspartamide were 5.9, 4.8 and 4.1 mol CO<sub>2</sub>/kg adsorbent for adsorption temperatures of 303, 318 and 333 K, respectively.

The maximum amount of CO<sub>2</sub> adsorbed by polyaspartamide at different flow rates of 1.0, 1.5 and 2.5 ml/s of the feed gas were 7.84, 6.5 and 5.9 mmol CO<sub>2</sub>/g of adsorbent. This shows that higher flow rates resulted in decreased amount of CO<sub>2</sub> adsorbed by polyaspartamide because of low residence time which eventually resulted in poor mass transfer between the adsorbent and adsorbate. Under dry conditions, the adsorption capacity of polyaspartamide was 365.4 mg CO<sub>2</sub>/g adsorbent and 354.1 mgCO<sub>2</sub>/g adsorbent under wet conditions. Therefore, the presence of moisture had a negligible effect on the adsorption behaviour of polyaspartamide. This is very common with most amine-rich polymer-based adsorbents. This could be attributed to the fact that CO<sub>2</sub> reacts with moisture to form carbonic acid, thereby enhancing the CO<sub>2</sub> adsorption capacity of the material.

In conclusion, this study confirmed that the adsorption of CO<sub>2</sub> onto polyaspartamide is favoured at low temperatures and high operating pressures. The adsorption of CO<sub>2</sub> onto polyaspartamide was governed by film diffusion according to the outcome of the Boyd's film diffusion model. It was also confirmed that intra-particle diffusion was the rate-limiting step controlling the adsorption of CO<sub>2</sub> onto polyaspartamide. According to the results from the kinetic study, it can be inferred that lower temperatures had an incremental effect on the kinetic behaviour of polyaspartamide, external mass transfer governed the CO<sub>2</sub> adsorption process and the adsorption of CO<sub>2</sub> onto polyaspartamide was confirmed to be a physicochemical process (both physisorption and chemisorption).