

Abstract

The need to mitigate climate change by reducing the emission of greenhouse gases such as CO₂ has never been more urgent. The increase in CO₂ emissions is driven mainly by the combustion of fossil fuels for electricity generation. Capturing it from point emitters such as power plants would ensure the continued exploration of fossil fuels such as coal. Recent efforts continue to focus on reducing the cost associated with CO₂ capture.

Membranes as a tool for CO₂ separation from flue gas have been investigated through experimental work and modelling. The main challenge to the large scale implementation of membranes for CO₂ capture from coal plants is the low driving force because of the low CO₂ composition in the flue gas. This necessitates the use of compressors for driving force generation and large membrane area, which results in high operational and capital cost. Simulation and optimisation studies continue to assess the feasibility of membrane application for CO₂ capture. Optimisation models have been based on constant CO₂ permeance values. However, the CO₂ permeance in facilitated transport membranes such as fixed-site carrier membranes varies with the partial pressure, which is proportional to the pressure and concentration of the target gas. The membrane also requires humid conditions for optimal permeance to be realised. In this work, superstructure based multistage membrane optimisation of the CO₂ capture process by a facilitated transport membrane from power plants has been carried out. A mixed-integer nonlinear program model (MINLP) is developed that has a variable CO₂ permeation model for a fixed site carrier (FSC) membrane is developed. The superstructure embeds numerous potential process routes and involves the treatment of a multicomponent humidified flue gas for CO₂ capture by membranes. The superstructure involves the option of vacuum only operation and sweep gas operation on the permeate side. The model simultaneously optimises the feed humidity, the energy consumption of the compressors, the membrane area required, the flowrate and gives the optimum process flowsheet. The objective of the model is to minimise the annualised cost of CO₂ capture. The benefits of the optimisation model are explored by analysing the results of different scenarios. Next, a sensitivity analysis of the CO₂ capture process is carried out. High separation targets of up to 100% recovery are also studied.

The results show that the multistage optimisation leads to a decrease in the membrane area of 39% and reduces the annual specific cost of capture by 14% compared to a predetermined two stage membrane process flowsheet. The lower cost of capture is attributed to the smaller

membrane area at relatively unchanged power consumption. The use of combining vacuum only and sweep driven stages can further reduce the cost of capture by 3%.

The model is also applied to a South African context. The simulation of the supercritical boiler power plant takes into account the high average atmospheric temperature (32.3 °C) of the location and low pressure (0.81 bar) values of Limpopo as well as air cooling, which is peculiar to the power plant. The type of coal specific to the area and the coal characteristics are also taken into account. The capture process would reduce the power output of the plant from 727.3 MW to 515.9 MW which is an efficiency penalty of 10.6%. The Carbon Capture and Storage (CCS) system would contribute 40% of the total annual cost of the power plant.

The utilisation of an N₂ selective membrane together with a CO₂ selective membrane in an integrated process system is also studied. An optimisation model is developed based on a superstructure that allows both the retentate and permeate streams to become the residue and product streams. The choice of operating either an N₂ selective or CO₂ selective membrane is an added degree of freedom. At the conventional separation targets of recovery of 90% and purity, 95% the optimised the N₂-CO₂ hybrid process system results in 14% savings in the cost of capture compared to the CO₂ selective membrane process.