

ABSTRACT

South Africa emits large amounts of carbon dioxide (CO₂) due to its reliance on coal. The emission of CO₂ needs to be reduced for clean sustainable energy generation. Research efforts have therefore been devoted to reducing CO₂ emissions by developing cost-effective methods for capturing and storing it. Amine-based absorption using monoethanolamine solvent is the most mature technique for CO₂ capture despite its huge energy consumption, corrosiveness and difficulty in solvent regeneration. However, CO₂ removal by solid adsorbents is a promising alternative because it consumes less energy, and can be operated at moderate temperature and pressure. Metal organic frameworks have received attention as a CO₂ adsorbent because they have large surface areas, open metal sites, high porosity and they require less energy for regeneration.

This research was aimed at optimizing and scaling-up SOD-ZMOF through structural modification for enhanced CO₂ adsorption by impregnating it with chitosan. Scaled-up SOD-ZMOF samples were prepared as described elsewhere and impregnated with Chitosan. Physiochemical properties obtained using X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR), and Nitrogen physisorption showed that SOD-ZMOF and SOD-ZMOF-chitosan were successfully synthesized. Qualitatively, the surface area of the SOD-ZMOF synthesized using the scaled up protocol is lower than the one prepared using the non-scaled-up protocol

XRD pattern of SOD-ZMOF showed that it was crystalline and was in agreement with literature. The XRD peaks of the SOD-ZMOF decreased after chitosan impregnation showing that chitosan was impregnated on SOD-ZMOF. The FTIR spectrum of SOD-ZMOF showed functional groups present in organic linker used to synthesize SOD-ZMOF, and that of the SOD-ZMOF-chitosan revealed the same functional groups but with disappearance of carboxylic acid functional group. N₂ physisorption showed a decrease in BET surface area and pore volume after chitosan impregnation on SOD-ZMOF as well.

Performance evaluation of the material was carried out with a demonstration adsorption set-up using a 15%/85% CO₂/N₂ mixture and as a thermal gravimetric analysis (TGA) using 100% CO₂. For both the packed-bed column and the TGA experiments, evaluation was conducted on SOD-ZMOF and SOD-ZMOF with chitosan for comparison. About 50 mg of the adsorbent

was used at 25 °C, 1 bar and 25 ml/min for the packed-bed column. For the adsorption with the TGA, 11 mg of adsorbent was used at 25 °C, 1 bar and 60 ml/min.

SOD-ZMOF showed improved adsorption capacity after chitosan impregnation. CO₂ adsorption capacity of SOD-ZMOF increased by 16% and 39% using packed-bed column and TGA, respectively, after chitosan impregnation. The increase in adsorption capacity was attributed to the impregnated chitosan that has amine groups that display a high affinity for CO₂.

A traditional approach was used to investigate the effect of adsorption temperature and inlet gas flowrate on the CO₂ adsorption capacity of SOD-ZMOF-chitosan. This was done using both the packed bed column and the TGA. Temperature range of 25-80 °C and inlet gas flowrate range of 25-90 ml/min were investigated. Adsorption capacity increased with a decrease in temperature and inlet gas flowrate. For the packed-bed column, maximum of 781 mg CO₂/ g adsorbent was obtained at 25°C, 1 bar, 25 ml/min and for the TGA a maximum CO₂ adsorption capacity of 23 mg/ g adsorbent at 25 °C, 1 bar, and 60 ml/min was obtained.