Abstract

Direct Air Capture (DAC) is an indispensable technology for mitigating climate effects caused by the increasing CO₂ levels in the earth’s atmosphere. Amongst the different DAC technologies, a solid amine sorbent-based adsorption process is a suitable candidate to enable negative CO₂ emissions and opening pathways to CO₂ utilisation. The field of CO₂ adsorption is under rapid development, both in the field of novel sorbent materials as well as for new process alternatives. Previous work has revealed the potential of a commercially available amine-functionalised sorbent, Lewatit® VP OC 1065, as benchmark sorbent for DAC. This sorbent is also used in this work and can capture 1 – 2 mol CO₂/kg{sorbent}, depending on the prevailing weather conditions. Next to having a sorbent with suitable properties, an efficient process is key to the successful implementation of DAC. This means that both an effective adsorption and regeneration process are required, with this work focussing on the latter. For the regeneration process, the contactor configuration and process conditions are important factors that determine the product CO₂ concentration, specific energy consumption (kJ/kgCO₂) and productivity (kgCO₂/day), but also the stability - and therefore lifetime - of the sorbent material.

In the DAC process concept under development, the sorbent is physically circulated between the adsorption and regeneration section. This enables separate optimisation of both the adsorption and regeneration section for their respective function. In this work the feasibility of a new regeneration strategy is explored by modelling and validated in a new pilot scale regeneration unit (capacity: ca. 10 kgCO₂/day). In this new regeneration concept, the sorbent is sequentially regenerated in four regeneration units. Two main objectives for the regeneration strategy include the prevention of thermal-oxidative degradation of the sorbent to increase its lifetime and secondly to avoid the use of inert purge gasses, enabling the production of pure CO₂. In short, the proposed four regeneration units aim respectively for (i) partial oxygen elimination by air evacuation, (ii) bulk H₂O desorption and complete oxygen elimination, (iii) CO₂ (and residual H₂O) desorption and (iv) sorbent cooling, before recirculation to the adsorber section. Overall, the objective is to find the optimal balance between productivity (kgCO₂/day) and specific energy consumption (kJ/kgCO₂).

The core of this new regeneration technology are the desorption units in step (ii) and (iii), for which a new type of contactor is used. In these regenerators, heated plates with vertically oriented fins are used to realise a high specific area for heat transfer and minimise the required heat penetration distance. The volume between the vertical fins is filled with sorbent by gravitational flow. The units are operated batchwise and under reduced pressure, avoiding the need for purge gasses to reduce the CO₂ partial pressure (and therefore increasing the driving force for desorption),
enabling the desorption of CO₂/H₂O gas mixture. The co-desorbed H₂O is condensed, yielding a pure CO₂ product.

In this work, first a modelling study is performed to assess the feasibility of the proposed regeneration strategy. Each unit is described by a two-dimensional numerical model (time and space), in which mass, species and energy balances are solved, incorporating empirical correlations for sorbate equilibria, reaction kinetics and heat transfer. The modelling study reveals that for the sorbent, loaded under realistic European weather conditions, H₂O desorption is favourable over CO₂ desorption between 40 and 70°C. This is found to be in agreement with preliminary experimental validation. Since H₂O desorption can take place at higher partial pressures and lower temperature compared to CO₂ desorption, it is unavoidable to co-desorb H₂O from the sorbent. However, it is favourable to separate the H₂O and CO₂ desorption steps in two different units operating at different pressure and temperature levels to minimize the energy consumption for gas evacuation under reduced pressures.

Following the modelling study, the separate regenerator units are constructed and experimental validation of the model results for the individual regenerator stages is started and ongoing. Initial experiments show that the four-step regeneration concept works and that H₂O and CO₂ recovery in separate desorption units is feasible. Further quantification of the performance and model validation is scheduled for the first quarter of 2022 and will be reported in the full paper. Next, experimentally validated models for the individual unit operations, as well as the overall pilot facility, will be used to guide optimisation of the pilot unit with respect to productivity and specific energy consumption.

The modelling study and preliminary experiments for the different regeneration stages show that it is possible to obtain CO₂ in high purity, enabling the possibility to use the product stream immediately in CO₂ utilisation applications requiring high-purity CO₂, such as electrochemical CO₂ conversion. The entire, mobile DAC pilot facility, targeted to produce 10 kgCO₂/day from ambient air, is currently under construction at the University of Twente and start up is scheduled for spring 2022. A first evaluation of the performance of the pilot facility is planned to be part of the full paper.

*Keywords: Direct Air Capture, Pilot Plant, Dynamic Modelling, Solid Amine Sorbent, CCU*