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A process model for crossflow CO₂ absorption

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Abstract

Crossflow absorbers have some advantages over conventional vertical counterflow absorbers for post-combustion CO₂ capture and could reduce capital cost, particularly for applications which have height constraints and/or low vapour phase CO₂ concentration. However, predicting performance of crossflow absorbers is challenging because commercially available process simulation software only supports mass/energy transfer calculations based on counterflow contactors. We have developed and validated a rate-based crossflow CO₂ absorber model which can be integrated into Aspen Plus to support techno-economic assessments of this promising process option.

Contactors for CO₂ absorption are typically designed as vertical counterflow columns with packing or trays to promote mass transfer between the liquid solvent flowing down from the top of the column and the vapour flowing up from the bottom. A vertical counterflow orientation in the contactor maximizes the overall mass transfer driving force between the vapour CO₂ partial pressure and liquid solvent equilibrium CO₂ fugacity and minimizes the area footprint for most contactor designs. However, opposing flow directions increase the vapour pressure drop through the contactor and also limit the operable flow range due to backup of liquid in the contactor (flooding) at high flow rates and poor liquid distribution (channeling) at low liquid flux. These restrictions can be particularly pertinent in post-combustion CO₂ capture applications with low vapour phase CO₂ concentration, such as combined cycle gas turbines (CCGTs) and direct air capture (DAC). There are also some applications where physical layout constraints limit the allowable height for contactors, such as power generation for offshore oil and gas production facilities¹ and small-scale applications, particularly in the transport sector.

Crossflow contactors, where liquid flowing vertically down through a packing material contacts vapour moving horizontally through the packing, are widely used in industrial cooling and air pollution abatement applications and can also be applied to absorption processes². Crossflow contactor design is more flexible than counterflow because the gas and liquid cross-sectional flow areas are independent. Historically, crossflow absorbers have been assessed as a means of process intensification because they can accommodate liquid and gas fluxes substantially higher than the flooding limit of a counterflow absorber with the same dimensions³. More recently, crossflow absorbers have been used for solvent-based DAC, such as the Carbon Engineering hydroxide-based system^{2,4}, because the orthogonal flow directions reduce pressure drop and energy consumption for the large volume of air movement required for DAC, resulting in lower CO₂ capture cost compared to vertical absorption towers^{4,5}. Crossflow absorption can also accommodate much lower liquid-to-gas ratios than vertical counterflow absorption while

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maintaining wetted packing for efficient mass transfer². Similar crossflow absorbers have been demonstrated for DAC using aqueous amino acid⁶ and amine solvents⁷. Elliff et al.⁸ found that a crossflow contactor would reduce the capital cost of an industrial-scale post-combustion CO₂ absorber by 15% compared to a vertical counterflow design because the crossflow absorber is more amenable to modular construction and has reduced foundation requirements. A crossflow absorber could also be physically integrated into the horizontal flue gas flow path of a typical natural gas or coal-fired power plant without requiring the flue gas to change direction which would reduce the complexity and cost of the gas manifolding⁸.

The primary disadvantage of crossflow absorption is the loss of mass transfer driving force between the vapour and liquid phases in the portion of the absorber near the rich solvent and CO₂-depleted vapour outlets. This is less of an issue for hydroxide-based CO₂ absorption because the solvent has extremely low equilibrium CO₂ fugacity, but for amine-based absorption it can be mitigated by having multiple crossflow absorber beds in series with the solvent and vapour progressing through the beds in opposite directions³.

Process modelling of crossflow CO₂ absorption is challenged because the mass transfer unit operations built into commercially available process simulation software are based on conventional vertical counterflow contactors. Recent efforts to develop process models of amine-based crossflow CO₂ absorption have been approximations based on discretising the crossflow absorber into a two-dimensional matrix of counterflow contactors using the built-in mass transfer unit operations in Aspen Plus^{9,10}. While this approach provides a rough approximation of crossflow absorber performance, accuracy is impaired by limited discretisation and the use of conventional counterflow unit operations for the mass and heat transfer calculations.

A purpose-built, rate-based crossflow absorber model has been developed for detailed mass and energy balance calculations of CO₂ absorption from a vapour. The model was implemented in Visual Basic using Aspen Properties¹¹ for thermodynamic property calculations and can be incorporated in larger process models as a user unit operation in Aspen Plus¹². The crossflow absorber is discretised into a two-dimensional matrix of cell elements representing a slice of the absorber with mass/energy balance and interphase transfer calculations performed on each cell which can readily be configured to incorporate fine resolution in the matrix discretisation. The model includes rigorous modelling of the electrolyte chemistry and kinetic reactions involving CO₂ species. Interphase mass and energy transfer in each cell is calculated based on crossflow orientation using mass transfer coefficients, effective interfacial area, and liquid holdup correlations developed at the University of Texas using a wide range of random and structured packing materials in counterflow orientation that have been used to successfully model amine-based CO₂ absorption¹³⁻¹⁵. These specific counterflow correlations are believed to be a reasonable proxy for crossflow absorption with low to moderate vapour flow rate because the effective interfacial area and liquid holdup do not depend on vapour flow rate and, unlike the standard correlations available in Aspen Plus, the vapour mass transfer coefficient is not affected by the liquid flow rate (and vice versa).

Although the model is intended to model amine solvent-based CO₂ absorption, it also works for hydroxide-based solvent. The model was validated against experimental data collected at the University of Edinburgh for CO₂ absorption from air using a 0.5 M sodium hydroxide solvent with two beds (0.5 m wide x 0.5 m high x 0.8 m long) of Sulzer Mellapak 250Y polypropylene structured packing¹⁶. Modelled CO₂ capture rate showed good agreement with the experimental measurements across a wide range of liquid flux (4.5-22.6 m³/m²-h) and vapour rates (F-factor 0.8-3.6 Pa^{0.5}) – e.g., 1.9% root-mean square deviation at 18.1 m³/m²-h in Figure 1. We are currently collecting experimental data using MEA solvent (30% wt.) and engine exhaust with a crossflow absorber (0.8 m wide x 0.8 m high x 1.2 m long) to compare with model predictions. The validated model provides a useful tool to more confidently design and optimize larger scale equipment for pilot testing and techno-economic analyses and a framework to develop more refined models as additional data becomes available.

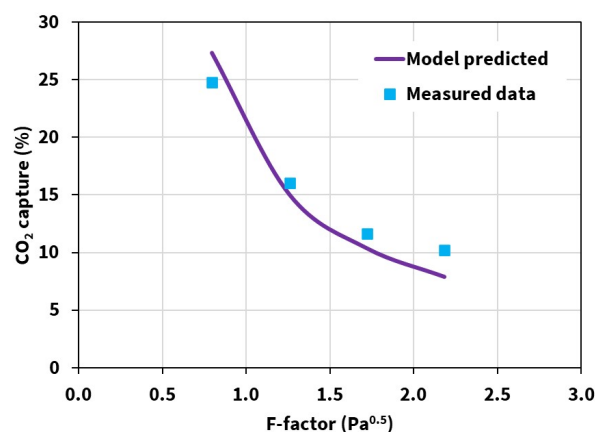


Figure 1. Comparison of model predicted CO₂ capture rate (purple line) and experimental data (blue squares) for crossflow CO₂ absorption from air with 0.5 M sodium hydroxide solvent using two absorber beds (each 0.5 m wide x 0.5 m high x 0.8 m long) in series with Sulzer Mellapak 250Y polypropylene structured packing and liquid flux 18 m³/m²-h. Experimental data from ref. ¹⁶.

Keywords: Carbon capture; absorption; crossflow; process modelling.

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