

Investigating CO₂ Absorption in Rotating Packed Beds: Development, Validation, and Operational Analysis of a Rate-Based Model

Babis Kantouros^a, Panagiotis Kazepidis^b, Athanasios I. Papadopoulos^b, Panos Seferlis^{a,*}

^aDepartment of Mechanical Engineering, Aristotle University of Thessaloniki, 54124 Thessaloniki, Greece

^bChemical Process and Energy Resources Institute, Centre for Research and Technology-Hellas (CERTH), Thessaloniki 57001, Greece
seferlis@auth.gr

Effective modelling of the Rotating Packed Bed (RPB) process is crucial for accurately simulating CO₂ absorption in aqueous amine solutions. Existing models often incorporate simplifying assumptions and rarely capture the simultaneous variations of critical phenomena inside the RPB apparatus, such as gas phase CO₂ mass flow rate, liquid and gas temperatures, and pressure drop. This work addresses these challenges by developing a rate-based, steady-state model designed for CO₂ capture, using monoethanolamine (MEA) as the solvent. Unlike previous works, this model effectively captures all these critical phenomena simultaneously. The model employs a first principles approach and solves material and energy balance equations using the shooting method. Two-film theory is used to represent mass transfer in conjunction with up-to-date correlations. The predicted results exhibit excellent agreement with the experimental data for a 30 wt. % MEA solution when comparing rich loading, with a maximum deviation of 1.1 %. The analysis of previously overlooked phenomena in RPB-based CO₂ absorption provides valuable insights regarding the variations of key parameters along the radius, such as the influence of pressure drop and temperature. The observed decrease in gas pressure and sharp drop near the inner radius demonstrate the significant impact of frictional losses. In contrast, the temperature profiles of the gas and liquid phase reveal the interplay between exothermic reactions and counter-current flow.

1. Introduction

Post-combustion CO₂ capture absorption systems are receiving increased attention to mitigate greenhouse gas emissions. Rotating Packed Bed (RPB) units and reactive solvents have demonstrated significant potential for improving CO₂ capture efficiency (Borhani et al., 2018). Process intensification using RPBs is a complex procedure due to the simultaneous phenomena that occur between the gas and liquid phases. Accurate modelling of this process is considered a significant challenge, as it can serve as a prediction and design tool for different operating conditions (Dimoliani et al., 2021). Research studies involving custom-made models for CO₂ capture have often employed simplifying assumptions concerning the absorption process. While these assumptions may be adequate for studying small RPB units, they could lead to significant discrepancies when applied to scaled-up operations (Hendry et al., 2020). For instance, Qian et al. (2009) developed a mathematical model to describe CO₂ absorption using an aqueous N-methyldiethanolamine (MDEA) solution. However, their model assumed isothermal operation, overlooked the pressure drop along the radius, and omitted the presence of water in the gas phase. Similarly, Yi et al. (2009) conducted an analysis using as a solvent the Benfield solution (hot potassium carbonate promoted by diethanolamine). Their model neglected the presence of water in the gas phase and the pressure drop while assuming isothermal absorption. Otitoju et al. (2023) reviewed existing models for MEA-based RPB absorbers that utilise the two-film theory for mass transfer. Commercial tools such as Aspen Plus® or gPROMS® were primarily used. The developed models focused on the effects of key operational parameters, such as rotational speed, lean MEA solution concentration and temperature and

CO₂ capture efficiency. However, these studies provided limited results concerning the depiction of phenomena occurring across the radial direction of the RPB. Joel et al. (2014) presented the liquid temperature profile along the radius in their model, while Borhani et al. (2018) focused on the CO₂ mole fraction profile in the gas phase. Luo et al. (2021) expanded the modelling parameters to include the CO₂ absorption profile along the radius, alongside liquid and gas temperatures, using the surface renewal theory. Their analysis did not include the pressure drop profile.

The objective of this work is to develop and validate a first principles model for CO₂ absorption in RPBs that employs an aqueous MEA solution. The model uses the two-film theory to represent the mass transfer process and integrates a broader range of phenomena occurring along the RPB radius than what is typically considered in existing models. Such phenomena include variations in gas phase pressure, temperature, and CO₂ mass flow rate. The model includes all the necessary compounds (CO₂, H₂O, MEA) in both the gas and liquid phases. The mass transfer coefficients are based on comparisons between a variety of mass transfer correlations and experimental data (Oko et al., 2019). Counter-current flow is implemented to augment mass transfer compared to co-current and cross-flow configurations (Kolawole et al., 2018). Figure 1 illustrates the gas flowing from the outer section towards the inner section while the liquid flows in the opposite direction. Water at the gas inlet is accounted for to mimic realistic operating conditions during the absorption process.

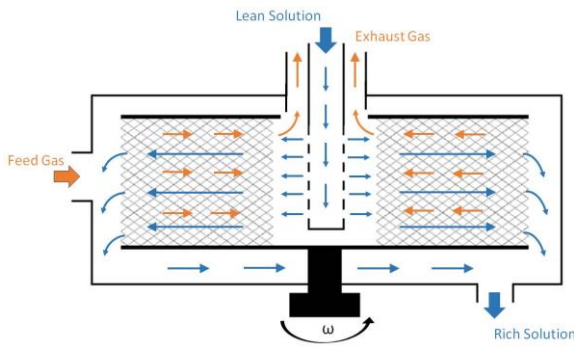


Figure 1: Counter-Current Flow in a Rotating Packed Bed absorption process

2. Methodology

A rate-based approach is essential to accurately describe the absorption process, primarily due to the increased rates of mass and heat transfer. Material and energy balance equations incorporate steady-state conditions and one-dimensional fluid flow in the radial direction. The gas phase is considered ideal and consists of CO₂, H₂O, N₂, and MEA. The mass transfer of N₂ can be ignored since it is an inert gas. Chemical reactions occur exclusively in the liquid film and are quantified by the enhancement factor.

2.1 Model development

In this work, the material and energy balance equations are derived based on the work of Kang et al. (2014), as presented in equations Eq(1) to Eq(4). The equation for pressure drop (Eq(5)) is incorporated, as its significant role in the absorption process has been highlighted by Hendry et al. (2020). These equations along with the corresponding boundary conditions are presented as follows:

Material Balance for liquid phase:

$$\frac{d(F_l x_i)}{dr} = \varepsilon a_{gl} A_c N_i, \quad BC: x_i = x_0 \text{ at } r = r_{inner}, \quad \frac{d(F_l x_i)}{dr} = 0 \text{ at } r = r_{outer} \quad (1)$$

Energy Balance for liquid phase:

$$\frac{d(F_l c_{pl} T_l)}{dr} = \varepsilon a_{gl} A_c (h_{gl} (T_l - T_g) - \Delta H_{rxn} N_{CO_2} - \Delta H_{vap} N_{H_2O}), \quad BC: T_l = T_{l0} \text{ at } r = r_{inner}, \quad \frac{d(F_l c_{pl} T_l)}{dr} = 0 \text{ at } r = r_{outer} \quad (2)$$

Material Balance for gas phase:

$$\frac{d(F_g y_i)}{dr} = \varepsilon a_{gl} A_c N_i, \quad BC: y_{inner} = y_0 \text{ at } r = r_{outer}, \quad \frac{d(F_g y_i)}{dr} = 0 \text{ at } r = r_{inner} \quad (3)$$

Energy Balance for gas phase:

$$\frac{d(F_g c_{pg} T_g)}{dr} = \varepsilon a_{gl} h_{gl} A_c (T_l - T_g), \quad BC: T_g = T_{g0} \text{ at } r = r_{outer}, \quad \frac{d(F_g c_{pg} T_g)}{dr} = 0 \text{ at } r = r_{inner} \quad (4)$$

Pressure Drop for gas phase:

$$\frac{dP}{dr} = \rho_g \omega^2 r + \rho_g u_g \frac{du}{dr} + \frac{150(1 - \varepsilon_g)^2 \mu_g}{d_p^2 \varepsilon_g^3} u_g + \frac{1.75(1 - \varepsilon_g) \rho_g}{d_p \varepsilon_g^3} u_g^2, \quad BC: P = P_0 \text{ at } r = r_{outer},$$

$$\frac{dP}{dr} = 0 \text{ at } r = r_{inner}$$
(5)

The equations encapsulate molar flow balances for each component i along the radius r (m), represented by the derivative terms. These derivative terms contain various variables specific to the liquid phase such as the molar flow rate F_l (mol/s), the mole fraction of each component x_i (-), the temperature T_l (°C) and the specific heat capacity C_{pl} (J/mol°C). For the gas phase the derivative terms include variables such as the molar flow F_g (mol/s), the mole fraction of each component y_i (-), the temperature T_g (°C), the specific heat capacity C_{pg} (J/mol°C) and the gas pressure P (kPa). In the boundary conditions the symbols x_0, T_{l0}, y_0, T_{g0} and P_0 represent the inlet conditions of each respective phase. The equations also include $A_c = 2\pi r z$ (m²) representing the tangential area, the void fraction of packing ε (-), the gas-liquid interfacial area a_{gl} (m²/m³), and the axial length z (m). Energy balance equations encompass interfacial heat transfer, the exothermic reaction of CO₂ in the liquid phase, and the process of water vaporization or condensation. Specifically, the value of heat transfer interfacial coefficient h_{gl} (W/m²°C) is determined based on thermophysical properties. The reaction enthalpy ΔH_{rxn} of CO₂ (J/mol) is calculated as a function of liquid temperature and loading (mol CO₂/mol MEA) using the correlation provided by Luo et al. (2021). The heat of vaporization ΔH_{vap} (J/mol) is computed as a function of temperature. The pressure drop equation includes the effects of centrifugal acceleration, the radial acceleration effect from Bernoulli's principle, and frictional losses inside the packing. Frictional losses are incorporated in Eq(5) using the Ergun equation (Hendry et al., 2020). Variables relevant to the pressure drop equation encompass gas thermophysical properties like density ρ_g (kg/m³) and viscosity μ_g (Pas), the angular speed ω (rad/s), the superficial gas velocity u_g (m/s), the gas hold-up ε_g (-), and d_p (m), denoting the spherical equivalent particle diameter.

2.2 Two-Film theory

The molar transfer flux, N_i (mol/m²s), is calculated for each molecular component (CO₂, H₂O, MEA) based on two-film theory as described by Kvamsdal et al. (2009) in Eq(6):

$$N_i = K_{g,i}(P_{g,i} - P_i^*)$$
(6)

where $P_{g,i}$ (Pa) is the partial pressure of bulk gas phase, P_i^* (Pa) is the equilibrium gas partial pressure and $K_{g,i}$ (mol/m² Pa s) is the overall mass transfer coefficient as follows:

$$\frac{1}{K_{g,i}} = \frac{R_g T_g}{k_{g,i}} + \frac{H e_{l,i}}{E_i k_{l,i}}$$
(7)

The right-hand side terms of Eq(7) represent the gas phase resistance and liquid phase resistance. The liquid phase resistance for MEA and H₂O is considered negligible. Henry's constant $H e_{l,i}$ (Pa m³/mol) for the solubility of CO₂ in aqueous MEA solution is determined using the expression described in Restrepo et al. (2015). The enhancement factor is employed in the mass transfer model to account for the reaction rate of CO₂ with MEA, as illustrated in Eq(8):

$$E_{CO_2} = \frac{\sqrt{k_{app} D_{l,CO_2}}}{k_{l,CO_2}}$$
(8)

where D_{l,CO_2} (m²/s) is the liquid-phase diffusivity of CO₂, k_{app} (1/s) is the apparent reaction rate constant. The latter is derived from the overall reaction between CO₂ and MEA, adhering to the termolecular mechanism (Aboudheir et al., 2003). The mass transfer model necessitates reliable correlations to accurately predict the effective interfacial area and the mass transfer coefficients for both liquid and gas films. The scarcity of experimental data for mass transfer in RPBs underscores the challenges of modelling this process. This study applies the correlation for the gas film mass transfer coefficient, $k_{g,i}$ (m/s), provided by Chen et al. (2011), which is based on experimental data for $K_c a$ (1/s), related to an RPB. The liquid film mass transfer coefficient, $k_{l,i}$ (m/s), as proposed by Tung and Mah (1985), offers superior accuracy in comparison to more complex correlations. The effective interfacial area (Billet and Schultes, 1999) is employed because of its ability to consider the impact of rotational speed accurately. Thermophysical properties, crucial for the calculation of mass transfer coefficients in both liquid and gas phases, are solely derived from literature in this study, specifically

employing correlations from the works of Borhani et al. (2018). The calculation of equilibrium component partial pressures in Vapor-Liquid Equilibrium (VLE) data critically affects the mass transfer rate, notably for CO₂. These pressures are estimated in line with the work of Damartzis et al. (2014) for a 30 wt. % MEA solution, adopting polynomial approximations in relation to loading and temperature. The shooting method (Yi et al., 2009) serves to solve the BVP as shown in Eq(1) to Eq(5). The BVP is converted into an Initial Value Problem (IVP) by assuming gas boundary conditions at the inner radius and progressing from the inner to the outer radius. By iteratively adjusting the initial boundary condition guess, the solution was fine-tuned until reaching the requisite level of precision.

3. Results and discussion

3.1 Model validation

The developed model is validated using steady-state experimental data for aqueous MEA solution from Jassim (2002). The pilot-plant absorber had an inner diameter of 0.156 (m), outer diameter of 0.396 (m) and axial length of 0.025 (m). The RPB contained an expanded stainless-steel mesh which had a void fraction of 0.76 and specific surface area of 2132 (m²/m³). The experimental conditions are listed in Table 1.

Table 1: Experimental Conditions (Jassim, 2002)- RPB Absorber

Case Run	w/w MEA (%)	Rotor speed (rpm)	Lean flow (kg/ s)	Lean temperature (°C)	Lean loading (-)	Vapor CO ₂ (vol %)	Vapor H ₂ O (vol %)	Vapor N ₂ (vol %)	Gas flow (kmol/h)
1	32	600	0.66	40.5	0.3349	0.0449	16.9	78.5	2.87
2	30	1,000	0.66	40.6	0.3374	0.0433	17.05	78.5	2.87
3	31	600	0.35	39.9	0.3254	0.0438	17.03	78.5	2.87
4	31	1,000	0.35	39.3	0.3282	0.0361	17.41	78.5	2.87

In the dataset of Jassim (2002) the CO₂ concentration at the inlet was 4 mol %, and the initial CO₂ loading in MEA was approximately 0.3 (mol CO₂/mol MEA). The validation of the intensified absorber model was performed by assessing rich loading.

Table 2: Key Variable Validation, RD stands for relative deviation

Case	Experimental Loading (mol CO ₂ / mol amine)	Predicted Loading (mol CO ₂ / mol amine)	RD (%)
1	0.3378	0.3416	1.1
2	0.3411	0.3443	0.9
3	0.3342	0.3372	0.9
4	0.3310	0.3382	0.2

In Table 2, the model demonstrates a significant level of agreement with published data for rich loading. Specifically, the maximum relative deviation for rich loading does not exceed 1.1 (%). These findings highlight the accuracy and reliability of the MEA model in simulating CO₂ absorption processes.

3.2 Operational analysis

The operational analysis is presented here that includes for the first time results for phenomena that were investigated separately in previous works, due to omission of the underlying representations. Figures 2 and 3 present the radial profiles for gas phase CO₂ mass flow rate, gas pressure drop, and liquid and gas temperatures. Realistic operating conditions for CO₂ capture applications required certain alterations from the conditions used for validation. The experimental setup of Jassim (2002) included a liquid-to-gas ratio, L/G (kg/kg), of up to 25, a level that could potentially lead to flooding. An L/G ratio of 3.4 was chosen for a closer approximation of a standard MEA absorption system, coupled with a rotational speed of 300 (rpm) (Qammar et al., 2018). The feed gas was based on Kazepidis et al. (2021). Details are shown in Table 3.

Table 3: RPB Simulation Conditions

Initial Conditions			
Gas flow in absorber (mol/s)	320	MEA weight percentage (w/w %)	30
CO ₂ composition in flue gas (v/v %)	14.11	Lean loading (mol CO ₂ / mol amine)	0.2
Gas inlet temperature (°C)	25	Liquid temperature (°C)	40
L/G (kg/kg)	3.4	N (rpm)	300

The presentation approach of the following figures allows for a clear examination of the behaviour of each variable within different segments of the RPB radius, which is the main focus of the analysis. The results are presented in relation to the normalized radius from the inner to the outer section of the RPB.

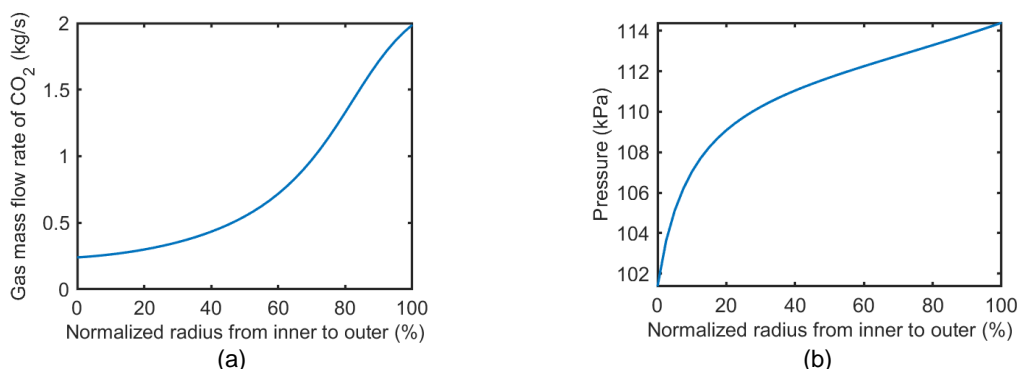


Figure 2: a) Gas mass flow rate of CO₂ profile, b) Gas pressure profile

Figure 2a reveals a steep gradient near the outer radius, the point that the feed gas is introduced into the RPB. This gradient is explained by the increased partial pressure of the CO₂ gas and the maximum tangential area. As the gas flows toward the inner radius, the gradient gradually stabilized until it reached a plateau. Figure 2b depicts a steady decrease in gas pressure across the majority of the radius, until a sharp drop occurs upon the inner radius. This phenomenon is primarily a consequence of frictional losses within the packing material. In particular, an increase in viscosity of the fluid is combined with a rise in superficial gas velocity. The latter is a result of the limited available tangential area. Together, these factors mainly contribute to an overall increase in frictional losses. Figure 3a reveals the temperature profile of the liquid, which reflects the trend of the CO₂ absorption profile shown in Figure 2a. The increase in liquid phase temperature, driven by the exothermic reaction between MEA and CO₂, leads to heat transfer and influences the variation in gas temperature, Figure 3b. It is worth noting that the liquid phase dominates the overall behaviour of the system. This interplay is a consequence of the counter-current flow, facilitating efficient heat exchange between the phases. Specifically, the heat transfer rate in the gas phase is significantly dependent on an accurate estimation of the interfacial heat transfer coefficient.

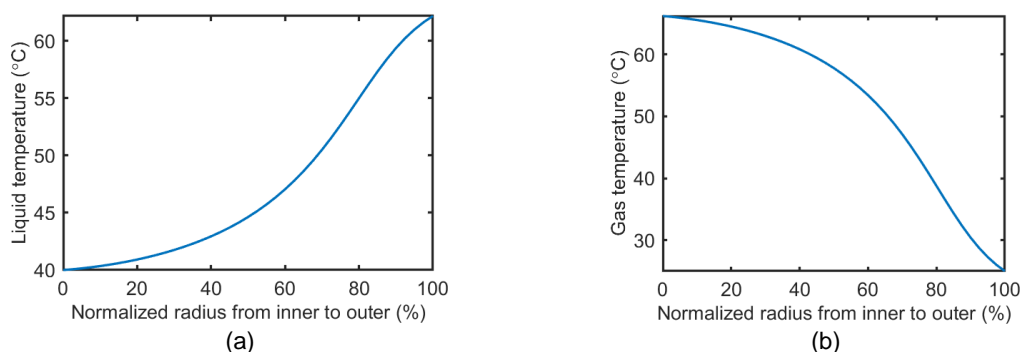


Figure 3: a) Liquid temperature profile, b) Gas temperature profile

4. Conclusions

A custom-made model was developed to simulate all critical phenomena during solvent-based CO₂ capture in a Rotating Packed Bed (RPB) unit. The model was validated against experimental data and found to exhibit excellent agreement, using monoethanolamine (MEA) as the solvent. Utilizing a rate-based approach under steady-state conditions, the model applied the two-film theory for the simulation of mass transfer. This work also presented comprehensive process diagrams that provide unique insights into the combined phenomena occurring along the RPB radius. The simulation revealed distinct patterns in gas pressure, temperature profiles, and CO₂ mass flow rate inside the RPB. Future work will involve applying this model to scale-up studies, while exploring its use with other solvents. This will provide a foundation for more reliable and comparative analysis

for intensified CO₂ absorption. In conclusion, precise process modelling in Rotating Packed Beds is crucial for further advancements in this field.

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