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Absorber Intercooling Configurations using Aqueous Piperazine for Capture from Sources with 4 to 27% CO_2

Darshan Sachde[a], Gary T. Rochelle[a]*

[a]*McKetta Department of Chemical Engineering, The University of Texas at Austin, 200 E. Dean Keeton St., C0400, Austin, TX 78712-1589 USA.*

Abstract

A systematic evaluation of the solvent capacity and mass transfer benefits of absorber intercooling was conducted for CO_2 capture with 8 m piperazine (PZ) for three different flue gas sources: NGCC (4.1% CO_2), coal-fired boiler (14.7% CO_2), and steel blast furnace (27.4% CO_2). The study identified the best intercooling strategy as a function of operating conditions (lean loading, liquid to gas ratio (L/G)). For all applications, operation at low lean loadings did not require intercooling to avoid temperature-related capacity or mass transfer penalties. In a broad intermediate loading range, simple in-and-out intercooling provided large solvent capacity benefits (measured as minimum solvent rate, L_{MIN}) compared to a column without intercooling. L_{MIN} was reduced by 71%, 53%, and 42% for NGCC, coal, and steel, respectively. Finally, the coal and steel applications had a large L/G range at higher loadings where intercooling was once again unnecessary. An enhanced intercooling design for NGCC could yield up to 62% reduction in packing and 46% reduction in solvent capacity at specific conditions (benefits for coal and steel were much lower). A recycle intercooling design for NGCC was introduced to reduce overall column temperatures and enhance mass transfer. For the case evaluated with recycle intercooling, the new design achieved significant packing reductions (>50%) compared to a simple intercooling design and approximated the performance of an isothermal column.

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Keywords: absorber intercooling, minimum solvent rate, piperazine, temperature bulge, mass transfer

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* Corresponding author. Tel.: +1-512-471-7230; fax: +1-512-471-7060.
E-mail address: gtr@che.utexas.edu

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1. Introduction

CO_2 capture with amine solvents results in the generation of a temperature maximum (or bulge) within the column due to heat released by absorption and reaction of CO_2. The higher solvent temperature limits the maximum capacity of the solvent (as measured by the change in solvent loading for a given CO_2 removal) and reduces average driving forces through the column. The reduced solvent capacity results in deterioration of the energy performance of the stripping system while the reduced driving forces can result in increased packing requirements in the absorber.

Intercooling the solvent to remove heat generated by CO_2 absorption can mitigate the capacity and driving force limitations, but the conditions of absorber operation (CO_2 concentration in flue gas, lean loading, L/G, etc.) affect the location and magnitude of the temperature bulge and overall temperature profile throughout the column. Therefore, the benefits of intercooling and the method used can vary with the operating conditions of the absorber.

Solvent intercooling or similar methods (e.g., flue gas cooling) have been proposed and evaluated for amine systems by several previous researchers [1-4]. However, many of these studies were focused on specific applications, configurations, solvent systems, or operating conditions and the results are difficult to generalize or appear to contradict one another on certain topics (e.g., the importance of the location of intercooling). The literature also contains minimal discussion of the limitations of intercooling methods or the development of new methods.

This work will present a systematic evaluation of intercooling benefits (solvent capacity and mass transfer benefits) across a range of CO_2 flue gas sources and operating conditions. The results will allow recommendations regarding the conditions for the application of intercooling and for development of improved intercooling design.

2. Modeling Framework

The absorber model used for the intercooling evaluation was implemented in Aspen Plus® in the RateSep™ module. The key components of the model are a solvent thermodynamic and kinetic model and a packing mass transfer model, which for allow rigorous rate-based modeling of the absorber.

All of the subsequent analysis will utilize 8 m aqueous piperazine (PZ) as the solvent. The thermodynamic model for the PZ-H_2O-CO_2 system was developed from experimental amine pKa, CO_2 solubility, heat capacity, speciation, and amine volatility data by regression of Gibbs free energy, enthalpy, heat capacity, and activity coefficient parameters within the electrolyte non-random two liquid (e-NRTL) framework.

The reaction set for the PZ model is as follows:

$$PZCOO^- + PZH^+ \leftrightarrow H^+PZCOO^- + PZ \quad (1)$$

$$PZCOO^- + H_2O + CO_2 \rightarrow H^+PZCOO^- + HCO_3{}^- \quad (2)$$

$$2PZ + CO_2 \rightarrow PZH^+ + PZCOO^- \quad (3)$$

$$2PZCOO^- + CO_2 \rightarrow H^+PZCOO^- + PZ(COO)_2{}^{2-} \quad (4)$$

Arrhenius rate expressions represent the rate constants for the kinetic reactions (last three reactions, including forward and reverse rates) where the pre-exponential and activation energy parameters were regressed from wetted wall column data collected over a range of temperatures, solvent concentrations, and loadings relevant for capture applications considered in this work. Finally, physical property models for binary diffusion coefficients, viscosity, and density were regressed as a function of amine concentration, loading, and temperature. For a detailed description of the “Independence” PZ model, see Frailie [5].

Mass transfer and area models were developed by Wang via regression of experimental data from a pilot scale column with a variety of random and structured packings [6]. Additional discussion of implementation of mass transfer models can be found in Sachde [7]. The area model developed by Wang is a modification of a model developed by Tsai (see Tsai for full theoretical and experimental details of the area model) [8].

3. Analysis and Methodology

The goal of this work is to systematically evaluate the potential benefits of intercooling over a range of representative operating conditions and to identify an intercooling method which will maximize benefits for each condition. Table 1 summarizes the conditions used for the evaluation. The entire analysis was conducted with 90% removal of CO_2 utilizing 8 m piperazine (PZ) as the solvent. The proposed lean loading ranges in Table 1 include low lean loading conditions that would lead to precipitation in practice with the PZ solvent. However, the wide range is included as an extrapolation of the PZ model in an attempt to allow generalization of the results.

Table 1. Summary of intercooling analysis design conditions

Flue Gas Source	**Inlet CO_2** (mol%)	**Lean Loading Range** mols CO_2/mols alkalinity
Natural Gas Combined Cycle Turbine	4.1	0.10 - 0.30
Coal-Fired Boiler	13.5	0.10 - 0.36
Steel Blast Furnace	27.4	0.10 - 0.39

The following process is used to evaluate intercooling benefits for the conditions identified in Table 1:

- Evaluate the solvent capacity of an adiabatic (no intercooling) absorber and an isothermal (“perfect” intercooling) absorber operated at 40 °C. These designs represent limiting cases of absorber performance. The adiabatic absorber (limited by temperature-induced equilibrium constraints) represents minimum solvent capacity and the isothermal absorber represents the maximum solvent capacity for a given operating condition.
- Quantify the packing required to approach maximum solvent capacity for adiabatic, isothermal, and intercooled absorber designs for each flue gas source. The results of this evaluation quantify benefits of each intercooling configuration in terms of solvent capacity (energy performance) and packing requirement (capital cost).
- Test process modifications (e.g., addition of intercooling loops) at the designated conditions to develop designs that approach maximum capacity (isothermal) with the minimum packing requirement.

Figure 1 illustrates the approach used in the analysis.

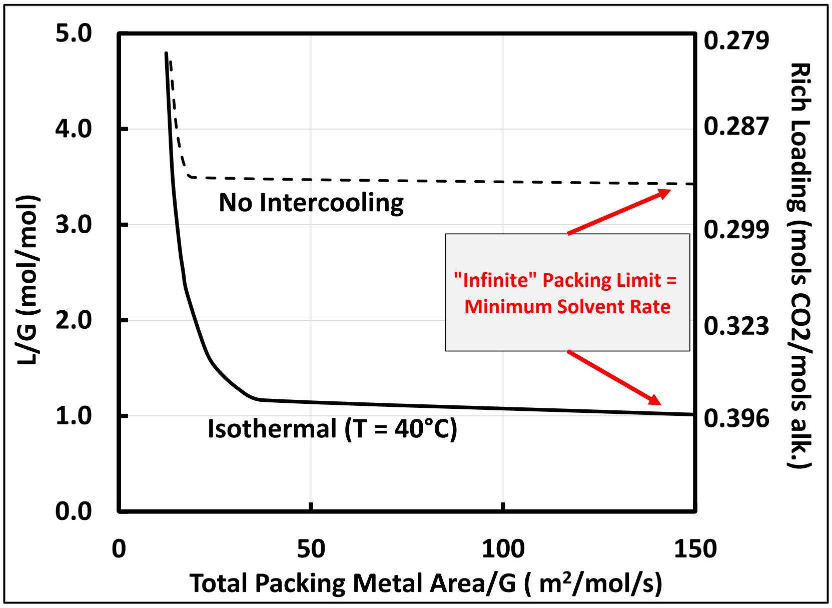

Figure 1: Design Curves for In-and-Out Intercooling (Blue), Recycle Intercooling at $L_{Recycle}/G = 3$ (Red) and Isothermal operation (Black). Each curve represents the packing requirement to achieve 90% CO_2 removal with a lean loading of 0.25 mols CO_2/mols alkalinity for a given liquid to gas ratio (L/G). For each point on the curve, the lean loading, removal, and solvent rates are fixed, so a unique rich loading exists as well (secondary y-axis). The rich loading can serve as a proxy for the energy performance of a given stripper configuration. The dashed line represents the minimum solvent rate required to achieve 90% CO_2 removal without intercooling.

Each curve represents the packing required for a given liquid to gas ratio (L/G) for each absorber design moving from one asymptotic limit (vertical asymptote of infinite solvent rate and minimum packing) to another (horizontal asymptote of infinite packing and minimum solvent rate). The minimum solvent rate (L_{MIN}) asymptote will be used to evaluate solvent capacity in this work. The L_{MIN} for any absorption process can be defined as the solvent rate required to achieve a specific solute removal (or specific gas inlet and outlet compositions) for a given inlet solvent composition (loading) with infinite mass transfer area available. Lower values of L_{MIN} are indicative of larger solvent capacity and are typically associated with better overall energy performance of the system. The ratio of the adiabatic L_{MIN} to the isothermal L_{MIN} at each condition can serve as a screening tool for the conditions where intercooling will be beneficial for energy performance. A high ratio indicates potentially significant benefits of intercooling. A ratio equal to (or approaching) unity indicates that the capacity benefits of intercooling are negligible or non-existent.

The second portion of the analysis will consider mass transfer constraints or packing requirements to approach maximum solvent capacity. The "infinite" packing limit is not a practical operating condition due to potentially prohibitive packing costs and operational instability near pinched conditions. Therefore, to properly evaluate absorber designs, the trade-off between solvent rate and packing requirement must be explicitly quantified. The curves in Figure 1 are representative of the packing-solvent circulation rate trade-off, and will be developed over the range of conditions (Table 1).

4. Results and Discussion

4.1. Minimum Solvent Rate Analysis: Adiabatic Absorber

Figures 2 to 4 include the L_{MIN} ratios for an adiabatic (no intercooling) absorber for all three flue gas sources in Table 1.

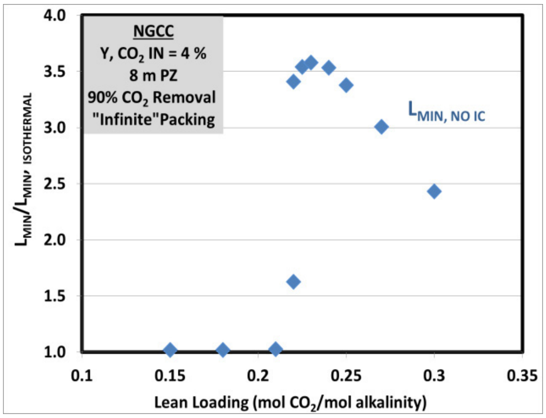

Figure 2: Ratio of the minimum solvent rate (“infinite” packing) achieved for an adiabatic absorber (no intercooling) to an isothermal absorber (T = 40 °C) for 90% CO_2 capture from a NGCC power plant (4.1% CO_2) utilizing 8 m PZ as the solvent.

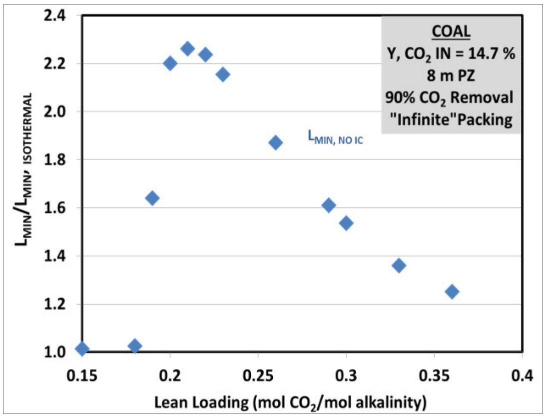

Figure 3: Ratio of the minimum solvent rate (“infinite” packing) achieved for an adiabatic absorber (no intercooling) to an isothermal absorber (T = 40 °C) for 90% CO_2 capture from a coal-fired boiler (14.7% CO_2) utilizing 8 m PZ as the solvent.

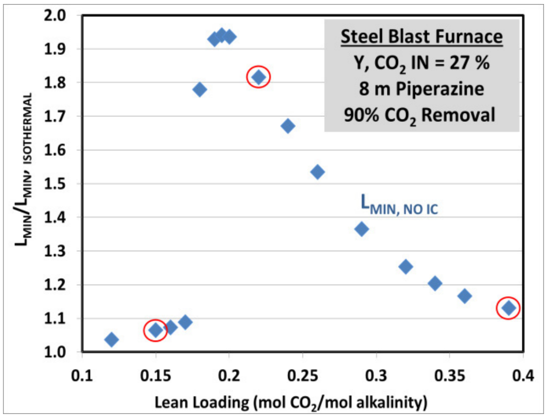

Figure 4: Ratio of the minimum solvent rate (“infinite” packing) achieved for an adiabatic absorber (no intercooling) to an isothermal absorber (T = 40 °C) for 90% CO_2 capture from a steel blast furnace (27.4% CO_2) utilizing 8 m PZ as the solvent. Points denoted by a red circle have corresponding equilibrium-operating line charts in Figures 5–7.

In all three cases, an intermediate range of loadings results in the largest ratios (largest deviation from an ideal isothermal absorber) and identifies conditions where the greatest energy benefits may be attained via intercooling. At extreme loadings (low and high) the ratios approach unity, indicating intercooling will provide limited benefits.

To understand the trends with loading, equilibrium-operating line constructions for cases without intercooling can provide insight into the effect of a temperature bulge in the column on the maximum capacity attainable (L_{MIN} achieved). Figures 5 through 7 include representative equilibrium-operating line charts in the three loading ranges (low, mid, and high) for the steel blast furnace application. The loadings selected for detailed analysis are highlighted in Figure 4 (red circles) for reference.

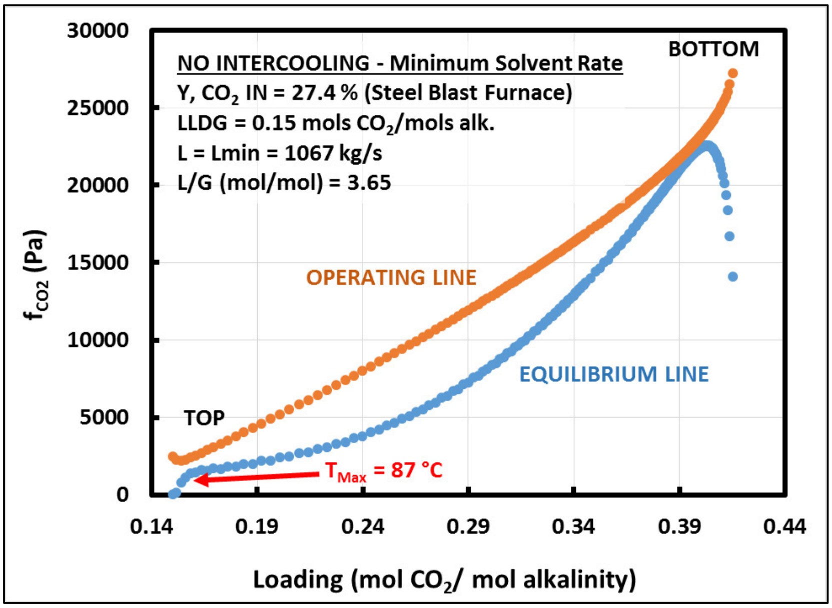

Figure 5: Operating and equilibrium curves for an adiabatic absorber (no intercooling) operated at the minimum solvent rate (“infinite” packing) to achieve 90% CO_2 removal with a lean loading of 0.15 mol CO_2/mol alkalinity (low loading region). The mass transfer pinch, represented by contact of the equilibrium and operating lines, occurs at the rich end of the column (bottom, T = 63 °C) and is unrelated to the maximum temperature (87 °C) near the top of the column.

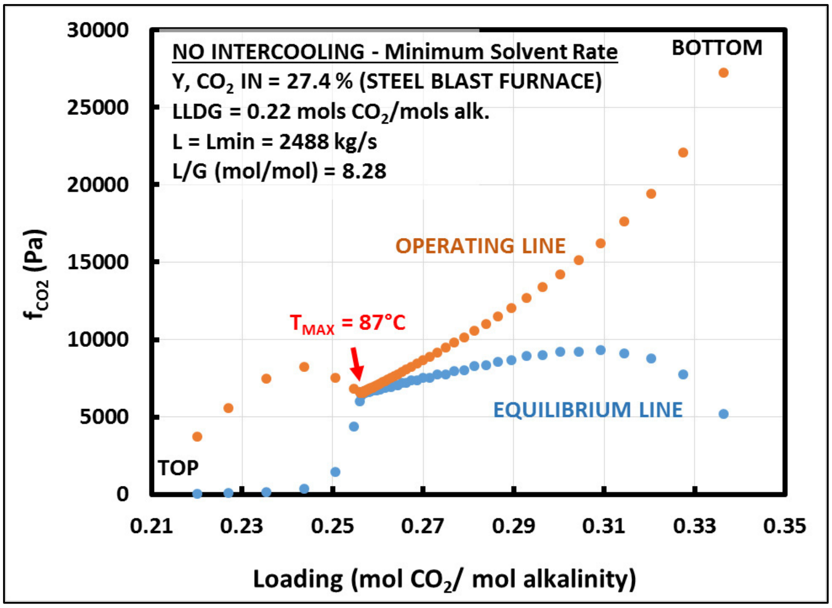

Figure 6: Operating and equilibrium curves for an adiabatic absorber (no intercooling) operated at the minimum solvent rate (“infinite” packing) to achieve 90% CO_2 removal with a lean loading of 0.22 mol CO_2/mol alkalinity (mid-loading region). The mass transfer pinch, represented by contact of the equilibrium and operating lines, occurs in the interior of the column and coincides with the maximum temperature (87 °C) realized in the absorber.

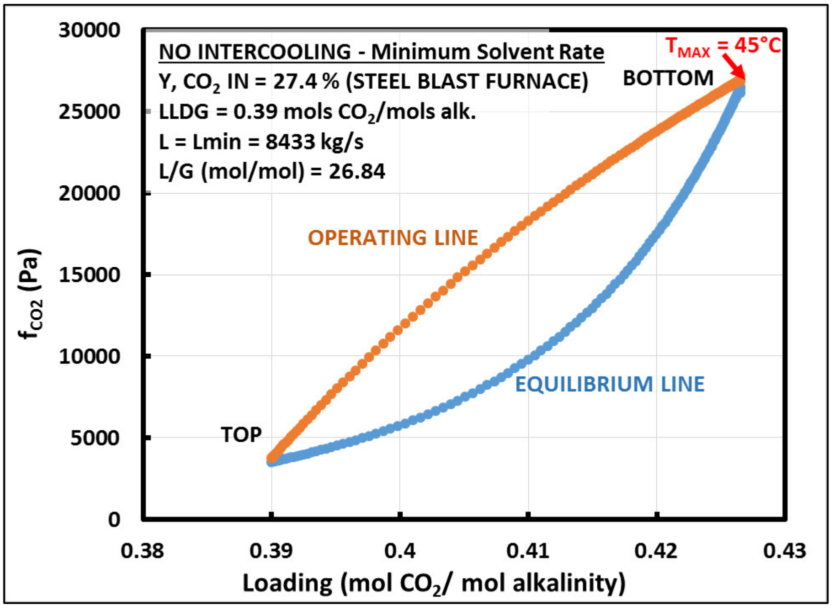

Figure 7: Operating and equilibrium curves for an adiabatic absorber operated at the minimum solvent rate (“infinite” packing) to achieve 90% CO_2 removal with a lean loading of 0.39 mols CO_2/mols alkalinity (high loading region). The mass transfer pinch, represented by contact of the equilibrium and operating lines, occurs near the rich end (bottom) of the column and coincides with the maximum temperature (45 °C) realized in the absorber.

The slope of the operating line (upper line in orange) in the preceding figures represents the liquid to gas ratio (L/G). The curvature present in the operating lines in the preceding diagrams is due to the concurrent transfer of water and an L/G that may vary significantly in different parts of the column. This differs from the typical binary diagram with only a single transferring component. As the liquid rate is reduced, the operating line becomes flatter (reduced slope) until it comes in contact with the equilibrium line; at this point in the column, the driving force is 0 (the column is “pinched”), and the solvent rate cannot be reduced any further while meeting the 90% removal specification. Therefore, the slope of the operating line when the pinch occurs represents the minimum solvent rate to achieve 90% removal for the given operating conditions and column configuration. The best performance (in terms of L_{MIN}) achievable for a given condition is an isothermal absorber with a mass transfer pinch at the rich end (bottom) of the column at a temperature of 40 °C. Equilibrium-operating line constructions provide insight into the effect of a temperature bulge in the column on the maximum capacity attainable (L_{MIN} achieved).

In Figure 5, the temperature bulge occurs near the top of the column due to the relatively low L/G at the lean operating conditions. At the low L/G condition, gas heat capacity dominates and carries heat generated by CO_2 absorption to the top of the column. The low lean loading also imposes large driving forces at the top of the column, so the temperature bulge in this region does not result in a mass transfer pinch. The pinch occurs near the rich end of the column at a temperature of 63 °C. Therefore, while some benefit can be derived by reducing the temperature at the rich end of the column, the temperature maxima in the column do not limit the solvent capacity, thus limiting potential benefits of intercooling.

In Figure 6, the mid-loading condition results in a temperature bulge towards the interior of the column. In this loading range, the solvent carries an increasing portion of the heat generated by CO_2 absorption and the heat is “trapped” in the column. The temperature bulge corresponds to a mass transfer pinch which limits the solvent capacity (as noted by the steep slope of the operating line). This condition corresponds to the largest potential benefits from intercooling via removal of the mass transfer limiting temperature bulge.

Finally, in Figure 7, the high lean loading results in a temperature bulge at the rich end of the column. The high L/G (liquid heat capacity dominates) required in this loading region drives the temperature bulge to the bottom of the column and results in a relatively low temperature at the bulge (45 °C). The mass transfer pinch occurs at this bulge, but the low temperature and proximity to the rich end of the column results in a performance very close to an isothermal column. Therefore, intercooling will provide limited benefits in this loading region.

The trends in the L_{MIN} ratios are generalizable from the steel case as discussed to the NGCC and coal cases. However, the figures do show some notable differences with CO_2 concentration (flue gas source). The maximum deviation from isothermal behavior decreases with increasing CO_2 concentration. The NGCC case reaches a maximum ratio of approximately 3.5, coal reaches a maximum of 2.2, and steel reaches a maximum of 2. This seems counter-intuitive since the higher CO_2 cases yield the highest absolute temperatures in the column and thus would appear to deviate from an isothermal column to a greater degree.

However, the deviation from an isothermal column is driven by the liquid to gas (L/G) ratio and the driving force for mass transfer in each case. In the NGCC, coal, and steel applications, the L/G ranges are 0.6–3.8, 2.3–10.6, and 3.7–26.8, respectively. While the absolute solvent temperatures increase with increasing CO_2 concentration, the corresponding increase in L/G dampens the relative temperature rise. Furthermore, the driving forces for mass transfer increase with CO_2 concentration. Figure 8 re-plots the preceding L_{MIN} ratio results as a function of the driving force at the top of the column.

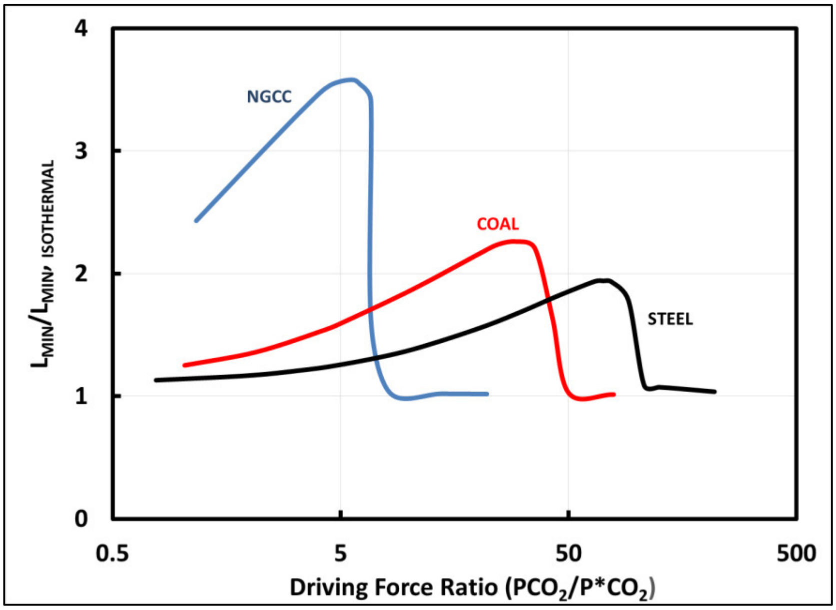

Figure 8: Ratio of the minimum solvent rate ("infinite" packing) achieved for an adiabatic absorber (no intercooling) to an isothermal absorber (T = 40 °C) for 90% CO_2 capture from a natural gas combined cycle power plant (4.1% CO_2), coal-fired boiler (14.7% CO_2), and a steel blast furnace (27.4% CO_2) utilizing 8 m PZ as the solvent. The ratios are plotted as a function of the relative driving force at the lean end (top) of the column, where PCO_2 represents the gas-side CO_2 partial pressure in the exiting flue gas and P^*CO_2 represents the equilibrium partial pressure of CO_2 corresponding to the lean solvent feed.

The NGCC case experiences the smallest driving force coupled with the smallest temperature buffering effect of the solvent rate, leading to the largest deviation from isothermal behavior. The driving forces and solvent buffering effect increase with CO_2 concentration, mitigating the capacity penalty of a temperature-related pinch in the absorber.

4.2. Minimum Solvent Rate Analysis: In-and-Out Intercooling

The preceding analysis on adiabatic absorbers identified specific conditions where intercooling may provide significant solvent capacity benefits (intermediate loading range). Figure 9 depicts a simple intercooling design, in-and-out intercooling, where the solvent is drawn off the column and cooled to 40°C at a single point in the column.

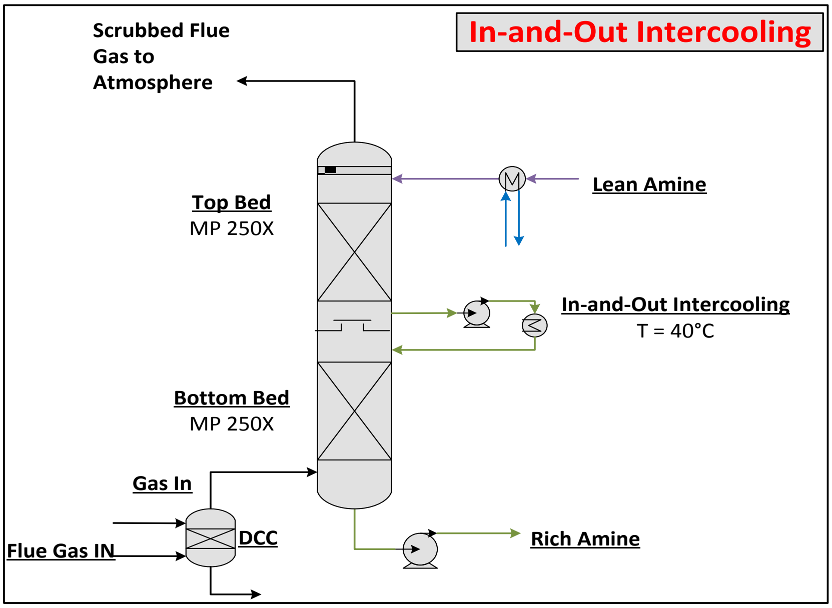

Figure 9: Absorber PFD for In-and-Out Intercooling. Two sections of packing (MP 250X) are used with liquid draw-off, cooling, and return between the packed sections. The solvent is cooled to 40 °C before returning to the column.

Figures 10-12 provide the L_{MIN} ratio results for the intercooled column.

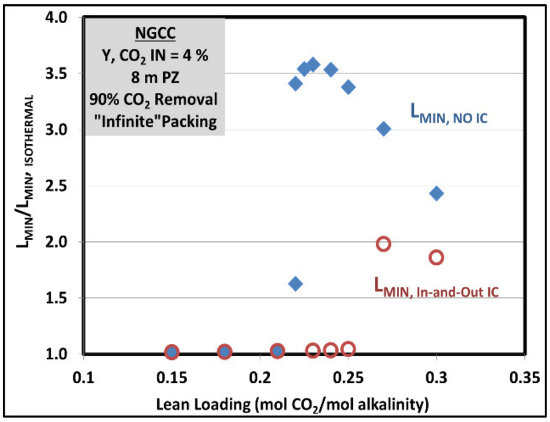

Figure 10: Ratio of the minimum solvent rate ("infinite" packing) for an adiabatic absorber (no intercooling) and intercooled absorber (in-and-out intercooling) to an isothermal absorber (40 °C) for 90% CO_2 capture from a natural gas combined cycle power plant (4.1% CO_2) using 8 m PZ.

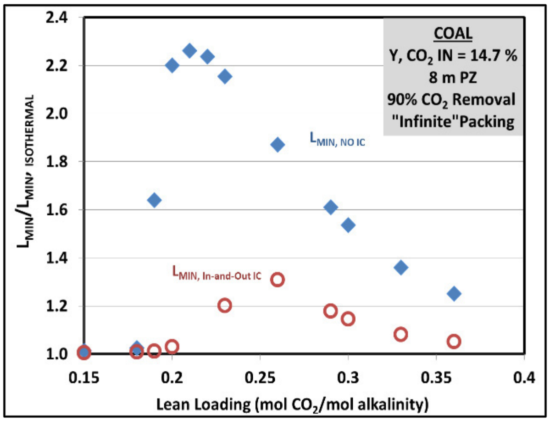

Figure 11: Ratio of the minimum solvent rate ("infinite" packing) for an adiabatic absorber (no intercooling) and intercooled absorber (in-and-out intercooling) to an isothermal absorber (40 °C) for 90% CO_2 capture from a coal-fired boiler (14.7% CO_2) using 8 m PZ.

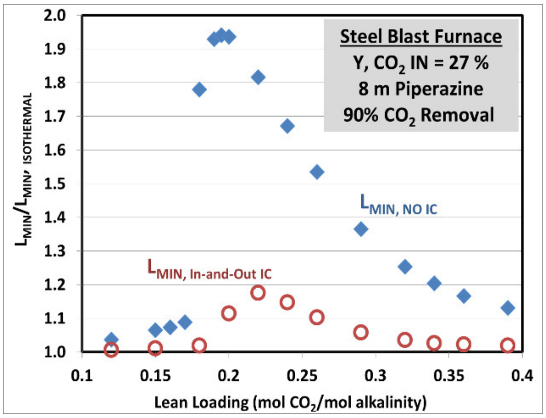

Figure 12: Ratio of the minimum solvent rate ("infinite" packing) for an adiabatic absorber (no intercooling) and intercooled absorber (in-and-out intercooling) to an isothermal absorber (40 °C) for 90% CO_2 capture from a steel blast furnace (27.4% CO_2) using 8 m PZ.

The intercooled absorber provides capacity benefits (denoted by lower ratios) when compared to the adiabatic absorber for all cases across the entire loading range evaluated. However, as with the adiabatic absorber, the intermediate loading range results in the highest ratios (greatest deviation from an ideal isothermal absorber) indicating that the in-and-out intercooling configuration does not completely address temperature limitations. The largest deviations from maximum solvent capacity are 100% for NGCC, 30% for coal, and 20% for the steel blast furnace application with in-and-out intercooling. In-and-out intercooling has expanded the loading range where near-isothermal capacity is achieved, but an operating range where novel intercooling configurations may provide solvent capacity benefits still exists for all cases.

4.3. Approach to Maximum Capacity: Mass Transfer Effects and Packing Requirement

To evaluate the packing requirements necessary to achieve the capacity benefits, representative loadings were selected for further evaluation (see Table 2). These loadings can be broadly classified from the L_{MIN} ratio analysis (Figures 10-12) as follows:

- "Over-stripped" Loading Range: Low lean loadings (large driving forces) and low L/G (lean end temperature bulge) prevent a mass transfer pinch at the temperature maxima. Intercooling is not required to provide capacity benefits in this region.
- Simple Intercooling Range: Simple in-and-out intercooling addresses temperature-related mass transfer pinch and provides near-maximum solvent capacity.
- Advanced Intercooling Range: Simple intercooling does not prevent a temperature-related pinch in this range and novel designs may achieve additional capacity benefits.
- Large L/G Range: Capacity benefits of intercooling diminish at high loadings due to high solvent rate limiting temperature effects in the absorber. This region only exists for the coal-fired boiler and steel blast furnace.

Table 2. Summary of loading ranges used in packing evaluation

Loading Range	**Loading Range** **mol CO_2/mol alkalinity**		
	Natural Gas Combined Cycle (NGCC)	**Coal-Fired Boiler**	**Steel Blast Furnace**
"Over-Stripped"	0.15–0.21	0.15–0.18	0.12-0.17
Simple Intercooling	0.22–0.26	0.19–0.21	0.18 – 0.19
Advanced Intercooling	> 0.26	0.22–0.30	0.20 – 0.29
Large Solvent Rate	N/A	> 0.30	> 0.30

For each loading range and application (11 total cases), a design curve, as in Figure 1, was developed to quantify the relationship between the solvent rate and packing requirement over a large range of operating conditions for an isothermal, adiabatic, and simple in-and-out intercooled absorber. The design curves for these cases can be found in Appendix A. The results of the analysis have been summarized in Table 3 as the maximum packing reduction that would be achieved if the best design for each case (either adiabatic or in-and-out intercooling) was replaced by an isothermal absorber (i.e., if perfect intercooling could be achieved). Alternatively, the slope of the adiabatic or intercooled design curve can be compared to the slope of the isothermal design curve. The slope of the design curve represents the packing requirement to approach maximum solvent capacity, where a steep slope represents a smaller packing requirement. In addition, the solvent capacity benefits from the L_{MIN} ratio analysis are summarized alongside the packing results in Table 3.

Table 3. Packing and capacity improvements possible with isothermal operation in place of best case absorber design (adiabatic or simple intercooling)

	NGCC		**Coal**		**Steel**	
Intercooling Region	**Maximum Packing Reduction**	**Capacity Improvement @L_{MIN}**	**Maximum Packing Reduction**	**Capacity Improvement @L_{MIN}**	**Maximum Packing Reduction**	**Capacity Improvement @L_{MIN}**
Over-stripped*	25%	2.1%	30%	1.4%	30%	3.5%
Simple Intercooling	**62%**	4.5%	29%	3%	15%	1.9%
Advanced IC	25%	**46%**	6.6%	24%	0%	15%
Large Solvent Rate*	N/A	N/A	15%	4.9%	17%	1.9%

* Over-stripped and Large Solvent Rate results are generated by comparimg an adiabatic absorber to an isothermal absorber. The remaining two regions compare in-and-out intercooling to an isothermal absorber.

The results are discussed subsequently by loading region.

4.3.1. "Over-Stripped" Loading Range

For all applications in the "over-stripped" loading range, the adiabatic and isothermal design curves nearly overlap (Figure 16-18 in Appendix A). This indicates that there is minimal opportunity for packing or capacity benefits via intercooling, as confirmed by the maximum benefits summarized in Table 3. The large driving forces at the over-stripped condition combined with a relatively low L/G ratio (gas carrying heat out of the column) mitigate temperature-related equilibrium effects in the absorber. In each case, the limited packing reduction without accompanying capacity benefits is unlikely to justify implementation of intercooling at the over-stripped condition.

4.3.2. "Simple Intercooling" Loading Range

In this region, intercooling is necessary due to the large capacity benefits achieved over the adiabatic absorber. However, the intercooling design should also minimize the packing requirement to achieve the capacity benefit. For the NGCC case, simple in-and-out intercooling can achieve the maximum solvent capacity, but the slope of this curve in Figure 19 is relatively flat in its approach to L_{MIN}. The slope of the curve reflects the packing requirement for an incremental improvement in capacity (reduction in solvent rate); a flat curve indicates a large packing requirement to achieve capacity improvement. The maximum packing reduction possible with isothermal operation in place of in-and-out intercooling is 62% for the NGCC application. Despite achieving maximum capacity, the simple intercooling design is not effective in addressing mass transfer limitations at the low L/G conditions, and a novel intercooling design should be developed.

In the coal-fired boiler and steel blast furnace cases (Figure 20 and Figure 21, respectively), in-and-out intercooling tracks closely to the isothermal curve over the entire range of conditions. The relatively large L/G ratios (solvent carries more of the heat) for these applications make in-and-out intercooling more effective for addressing temperature-related constraints. The maximum packing reduction achieved with isothermal operation in place of in-and-out intercooling is 29% for coal and 15% for steel, but is non-existent over most operating conditions. Therefore, simple intercooling is likely to be adequate to address mass transfer and capacity limitations for the higher CO_2 flue gas applications.

4.3.3. "Advanced Intercooling" Loading Range

A new intercooling design is expected for all applications in this region based on the inadequacy of in-and-out intercooling to achieve maximum solvent capacity (> 15% capacity improvement possible for all applications with improved intercooling). For the NGCC case, the design curve exhibits a relatively flat slope in the approach to maximum capacity (Figure 22). Therefore, in addition to capacity benefits, significant packing reduction (up to 25%) may also be possible with a novel intercooling design.

The higher CO_2 applications (coal and steel) do not provide extensive packing benefits (< 10% for both cases) to supplement the capacity benefits (Figure 23 and Figure 24). Once again, the relatively large solvent rates make in-and-out intercooling more effective since the solvent carries a greater portion of the heat in the column. Therefore, a novel intercooling design for the coal and steel applications must include minimal additional capital expenditure and operating complexity while achieving maximum solvent capacity to justify implementation over in-and-out intercooling in this loading region.

4.3.4. "Large Solvent Rate" Loading Range

For the high CO_2 applications (coal and steel), an additional loading range exists where the solvent rate is sufficiently high to mitigate temperature-related equilibrium constraints and the system can approach maximum capacity without intercooling. Figure 25 and 26 provide absorber design curves for this operating range.

In both cases, the relatively large solvent rate dampens temperature effects and pushes the temperature-related pinch towards the rich end of the column allowing the adiabatic absorber to perform similarly to an isothermal absorber. The maximum packing reduction of 15% for both applications with the limited capacity improvements are unlikely to justify implementation of any intercooling method at this condition.

4.4. Advanced Intercooling Development

The packing and capacity analysis highlighted the potentially significant benefits of novel intercooling development for the NGCC application across a large range of operating conditions. A new intercooling design was applied to the "simple intercooling" loading range for the NGCC application. The new recycle intercooling design is depicted in Figure 13 and the design curves for the 4 absorber designs (adiabatic, isothermal, simple intercooling, advanced intercooling) are shown in Figure 14.

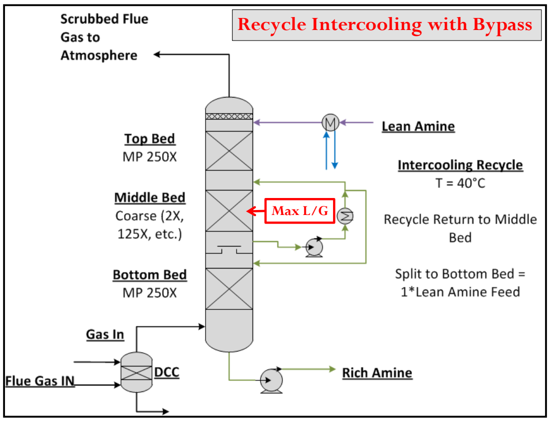

Figure 13: Absorber PFD for Recycle Intercooling with Bypass. Three packing sections are used, with the packing height of each section optimized for each design case to minimize total packing area. MP-250X is used in the top and bottom section and various coarse structured packing is used in the middle (recycle section) to maintain 70% max approach to flood. Solvent is drawn off the bottom of the middle section and cooled to 40 °C. A portion of the solvent is sent directly to the bottom section of the column (equal to the nominal liquid feed rate of the column) while the remaining liquid is recycled to the top of the middle section.

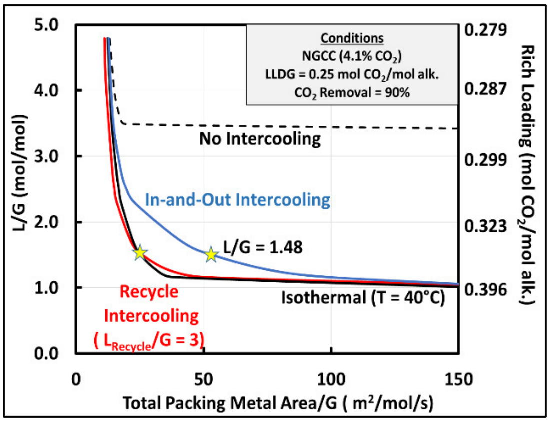

Figure 14: Design curves for adiabatic (dashed), in-and-out intercooling (Blue), and isothermal (solid) operation for NGCC application (4.1% CO_2) in the simple intercooling loading range (0.25 mol CO_2/mol alkalinity). Each curve represents the packing requirement to achieve 90% CO_2 removal for a given liquid to gas ratio (L/G). For each point on the curve, the lean loading, removal, and solvent rates are fixed, so a unique rich loading exists (secondary y-axis). The point highlighted in the chart with a star represent a constant L/G = 1.48.

The recycle intercooling design modifies the simple intercooling method by recycling the cooled solvent around a bed of packing in the middle of the column. The recycle rate in this section is now a new degree of freedom, constrained by flooding, pressure drop, and cost of pumping the solvent. The packing type in the middle section can also be optimized to reduce pressure drop with the large solvent rate (use of coarse packing). The recycle design was expected to be effective in the NGCC case due to the low L/G inherent to the application. Since the gas carries a greater portion of the heat generated in the column in low L/G applications, using a large amount of solvent to cool the gas will effectively reduce temperatures throughout the column. In addition, the new degrees of freedom regarding the solvent recycle and packing type create an opportunity to generate turbulence in the recycle section (large solvent rate over smaller surface or perimeter) which is associated with enhanced liquid side mass transfer and generation of additional wetted area. Equation 1 shows the dependence of the mass transfer and area models used in this work on the solvent rate per unit perimeter of packing [6,8].

$$k_L \sim \left(\frac{U_{SL}}{a_p}\right)^{0.63} \quad a_f \sim \left(\frac{U_{SL}}{a_p}\right)^{0.16} \tag{1}$$

Where:

k_L is the liquid side mass transfer coefficient (m/s),
a_f is the fractional packing area available for mass transfer (m^2/ m^2),
U_{SL} is the superficial liquid velocity (m/s),
a_p is the specific area of the packing (m^2/m^3).

As seen in Figure 14, the recycle design shows significant improvement over the simple in-and-out intercooling design and nearly tracks the isothermal column. Though both intercooling designs ultimately achieve similar maximum solvent capacities (similar L_{MIN}), the recycle intercooling design can approach the maximum capacity with significantly less mass transfer area (steeper slope in Figure 14). For example, the two intercooling designs can be compared at a common solvent rate (equivalent solvent capacity/energy performance). The point identified in Figure 14 corresponds to an L/G of 1.48 mol/mol. At this condition, the recycle design requires 51% less packing than the in-and-out intercooled design and essentially identical packing to an isothermal column. Figure 15 illustrates the reduced column temperatures compared to in-and-out intercooling at the selected operating condition.

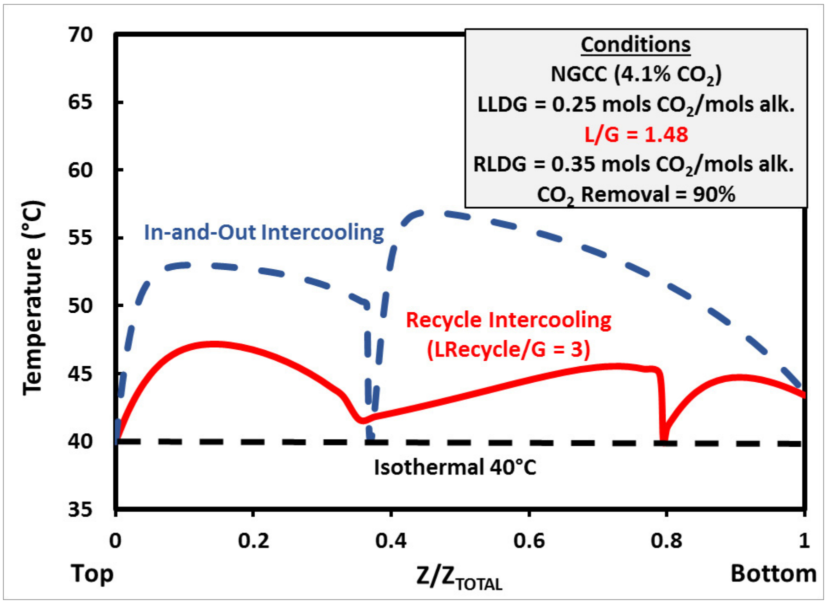

Figure 15: Temperature profiles for In-and-Out Intercooling (Blue), Recycle Intercooling at $L_{Recycle}/G = 3$ (Red) and Isothermal operation (**Black**). Each curve represents the profile 90% CO_2 removal with a lean loading of 0.25 mols CO_2/mols alkalinity for the given liquid to gas ratio (L/G = 1.48).

The recycle design effectively reduces temperatures throughout the column by cooling the gas in the recycle section and sending cold solvent to the bottom bed of the column. However, the column is not operating isothermally, so all benefits are not explained by solvent cooling and enhanced driving forces. The remaining mass transfer enhancements that allow performance comparable to an isothermal column arise from the turbulence generated in the column and quantified via the mass transfer models in Equation 1.

5. Conclusions

A systematic evaluation of potential solvent capacity and mass transfer benefits of intercooling was conducted for 90% CO_2 capture utilizing 8 m PZ for three flue gas sources with CO_2 concentrations from 4–27%. For each flue gas source, a wide range of operating conditions were evaluated (lean loading, L/G) to identify trends in intercooling benefits, generalize benefits as a function of operating conditions, and identify conditions where new intercooling designs were needed to improve performance.

Minimum solvent analysis was used as a screening tool for solvent capacity benefits of intercooling.

- The largest intercooling benefits for each of the three flue gas sources occurred in the intermediate or mid-loading range. At extreme loadings (low and high), the L_{MIN} ratio approached unity, indicating minimal potential benefits from intercooling.
    - Low Loading Range: The temperature bulge is driven to the lean end of the column (low L/G) where large driving forces exist, thereby preventing a mass transfer pinch corresponding to the bulge..
    - Mid-loading Range: A lean end pinch forms as the solvent rate is increased (moderate L/G), severely limiting solvent capacity. Large solvent capacity benefits can be realized with intercooling by removing the temperature bulge and associated pinch.
    - High loading Range: The temperature bulge tends toward the rich end of the column (high L/G) and coincides with a limiting mass transfer pinch. However, the temperatures are lower due to the large liquid rate, and the pinch approximates a rich end pinch in an isothermal absorber ("perfect intercooling").
- The largest potential solvent capacity benefits of intercooling occur for the NGCC application. The maximum deviation from isothermal L_{MIN} for the NGCC case was ~3.5, for coal ~2.2, and for steel ~2. For higher CO_2 content, the relatively high L/G moderates the temperature increase and the impact of the temperature increase is limited by larger driving forces in the column.
- Simple in-and-out intercooling was introduced as a baseline intercooling method. The method provided significant capacity benefits over the adiabatic absorber, but did not achieve maximum solvent capacity in all cases. The maximum deviation from the isothermal L_{MIN} dropped to ~2 for NGCC, ~1.3 for coal, and ~1.18 for steel. The range of lean loadings where the column exhibited near isothermal solvent capacity also expanded.

The mass transfer requirements to approach maximum solvent capacity were evaluated by developing design curves that described the trade-off between solvent rate and packing requirement for representative loading ranges. By considering packing requirements alongside the solvent capacity results from the minimum solvent analysis, the following recommendations were developed regarding intercooling:

- "Over-stripped" Loading Range
    - NGCC (0.15 – 0.21 mol CO_2/mol alkalinity): No Intercooling
    - Coal (0.15 – 0.18 mol CO_2/mol alkalinity): No Intercooling
    - Steel (0.12 – 0.17 mol CO_2/mol alkalinity): No Intercooling
- "Simple Intercooling" Loading Range
    - NGCC (0.22 – 0.26 mol CO_2/mol alkalinity): Advanced Intercooling – Large packing benefits
    - Coal (0.19 – 0.21 mol CO_2/mol alkalinity): Simple In-and-Out Intercooling
    - Steel (0.18 – 0.19 mol CO_2/mol alkalinity): Simple In-and-Out Intercooling
- "Advanced Intercooling" Loading Range
    - NGCC (> 0.26 mol CO_2/mol alkalinity): Advanced Intercooling – Large packing and capacity benefits
    - Coal (0.22 – 0.30 mol CO_2/mol alkalinity): Advanced Intercooling – Only capacity benefits
    - Steel (0.20 – 0.29 mol CO_2/mol alkalinity): Advanced Intercooling – Only capacity benefits
- "Large Solvent Rate" Loading Range
    - Coal (>0.30 mol CO_2/mol alkalinity): No Intercooling
    - Steel (>0.30 mol CO_2/mol alkalinity): No Intercooling

Finally, recycle intercooling was introduced for the NGCC application to take advantage of the inherently low L/G in this case. Recycling a large amount of solvent in the intercooling loop effectively cooled the gas in addition to the liquid and reduced temperatures throughout the column. The recycle loop also generates mass turbulence in the middle section of the column by introducing the large solvent flow to a coarse packing. The turbulence enhances the wetted area available as well as the liquid-side mass transfer coefficient. When evaluated at a constant solvent rate (equivalent solvent capacity), the recycle intercooling design required 51% less packing than the simple intercooling design and approximated the mass transfer performance of an isothermal column. The potential for novel designs was identified for each of the three applications with the recycle design showing promise for several conditions in the NGCC application.

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The authors declare the following competing financial interest(s): One author of this publication consults for Southern Company and for Neumann Systems Group on the development of amine scrubbing technology. The terms of this arrangement have been reviewed and approved by the University of Texas at Austin in accordance with its policy on objectivity in research. The authors have financial interests in intellectual property owned by the University of Texas that includes ideas reported in this paper.

Appendix A. Design Curves for 4 Loading Regions

A.1. "Over-Stripped" Loading Range

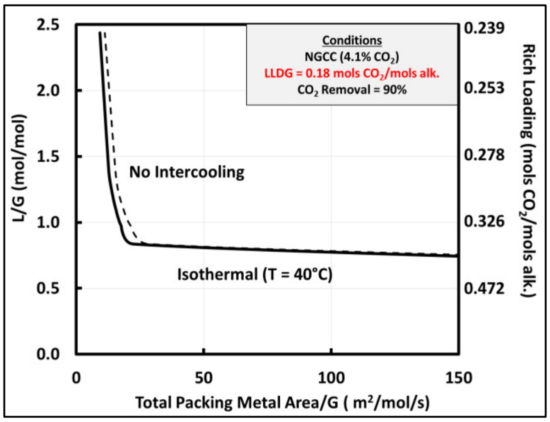

Figure 16: Design curves for adiabatic (**dashed**) and isothermal (**solid**) operation for NGCC application (4.1% CO_2) in the "over-stripped" loading range (0.18 mol CO_2/mol alkalinity). Each curve represents the packing requirement to achieve 90% CO_2 removal for a given liquid to gas ratio (L/G). For each point on the curve, the lean loading, removal, and solvent rates are fixed, so a unique rich loading exists (secondary y-axis).

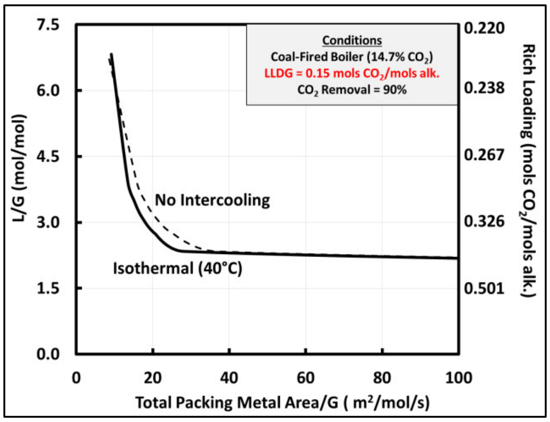

Figure 17: Design curves for adiabatic (**dashed**) and isothermal (**solid**) operation for coal-fired boiler application (14.7% CO_2) in the "over-stripped" loading range (0.15 mol CO_2/mol alkalinity). Each curve represents the packing requirement to achieve 90% CO_2 removal for a given liquid to gas ratio (L/G). For each point on the curve, the lean loading, removal, and solvent rates are fixed, so a unique rich loading exists (secondary y-axis).

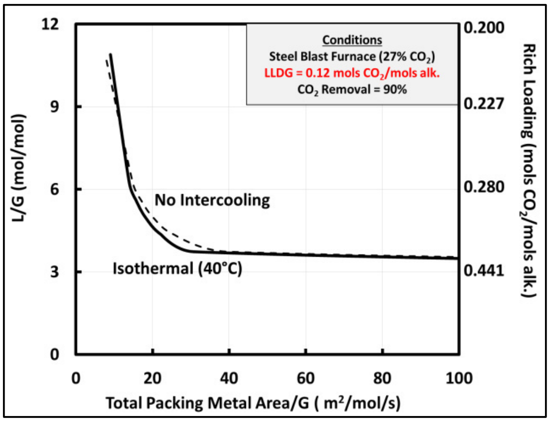

Figure 18: Design curves for adiabatic (**dashed**) and isothermal (**solid**) operation for steel blast furnace application (27% CO_2) in the "over-stripped" loading range (0.12 mol CO_2/mol alkalinity). Each curve represents the packing requirement to achieve 90% CO_2 removal for a given liquid to gas ratio (L/G). For each point on the curve, the lean loading, removal, and solvent rates are fixed, so a unique rich loading exists (secondary y-axis).

A.2. "Simple Intercooling" Loading Range

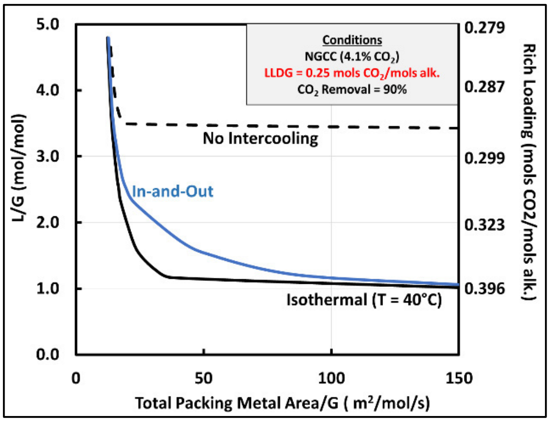

Figure 19: Design curves for adiabatic (**dashed**), in-and-out intercooling (**blue**), and isothermal (**solid**) operation for NGCC application (4.1% CO_2) in the simple intercooling loading range (0.25 mol CO_2/mol alkalinity). Each curve represents the packing requirement to achieve 90% CO_2 removal for a given liquid to gas ratio (L/G). For each point on the curve, the lean loading, removal, and solvent rates are fixed, so a unique rich loading exists (secondary y-axis).

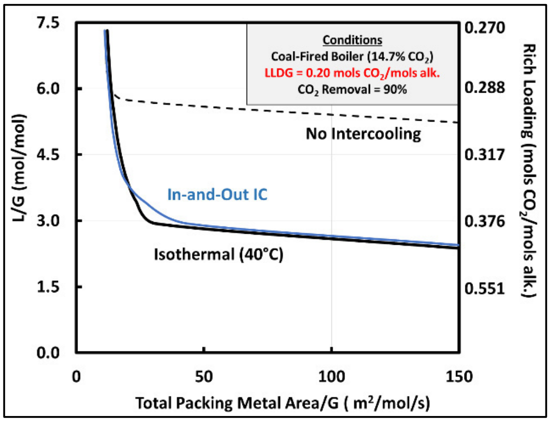

Figure 20: Design curves for adiabatic (**dashed**), in-and-out intercooling (**blue**), and isothermal (**solid**) operation for coal-fired boiler application (14.7% CO_2) in the simple intercooling loading range (0.20 mol CO_2/mol alkalinity). Each curve represents the packing requirement to achieve 90% CO_2 removal for a given liquid to gas ratio (L/G). For each point on the curve, the lean loading, removal, and solvent rates are fixed, so a unique rich loading exists (secondary y-axis).

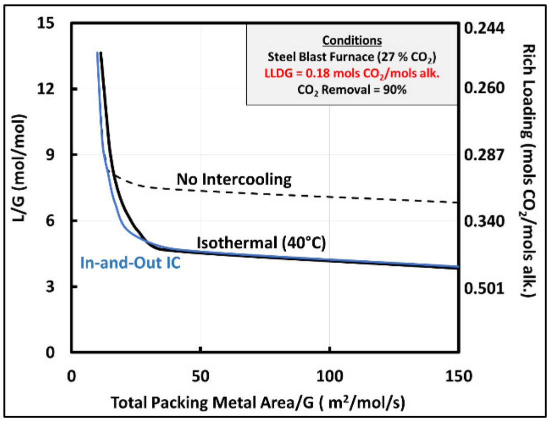

Figure 21: Design curves for adiabatic (**dashed**), in-and-out intercooling (**blue**), and isothermal (**solid**) operation for steel blast furnace application (27% CO_2) in the simple intercooling loading range (0.18 mol CO_2/mol alkalinity). Each curve represents the packing requirement to achieve 90% CO_2 removal for a given liquid to gas ratio (L/G). For each point on the curve, the lean loading, removal, and solvent rates are fixed, so a unique rich loading exists (secondary y-axis).

A.3. "Advanced Intercooling" Loading Range

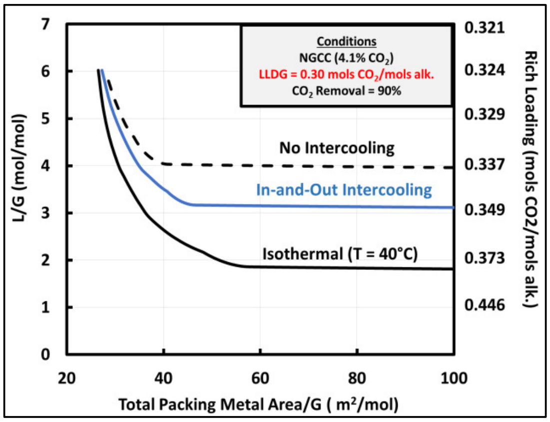

Figure 22: Design curves for adiabatic (**dashed**), in-and-out intercooling (**blue**), and isothermal (**solid**) operation for NGCC application (4.1% CO_2) in the novel intercooling loading range (0.30 mol CO_2/mol alkalinity). Each curve represents the packing requirement to achieve 90% CO_2 removal for a given liquid to gas ratio (L/G). For each point on the curve, the lean loading, removal, and solvent rates are fixed, so a unique rich loading exists (secondary y-axis).

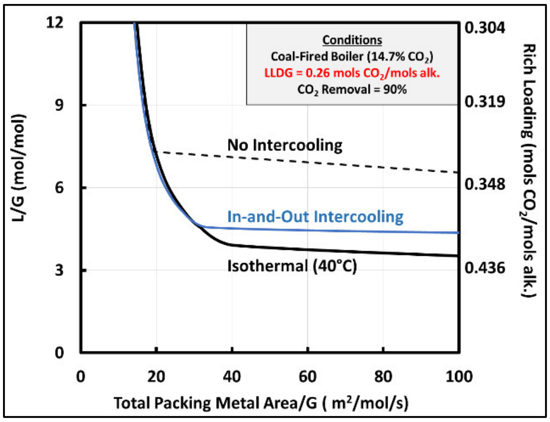

Figure 23: Design curves for adiabatic (**dashed**), in-and-out intercooling (**blue**), and isothermal (**solid**) operation for coal-fired boiler application (14.7% CO_2) in the novel intercooling loading range (0.26 mol CO_2/mol alkalinity). Each curve represents the packing requirement to achieve 90% CO_2 removal for a given liquid to gas ratio (L/G). For each point on the curve, the lean loading, removal, and solvent rates are fixed, so a unique rich loading exists (secondary y-axis).

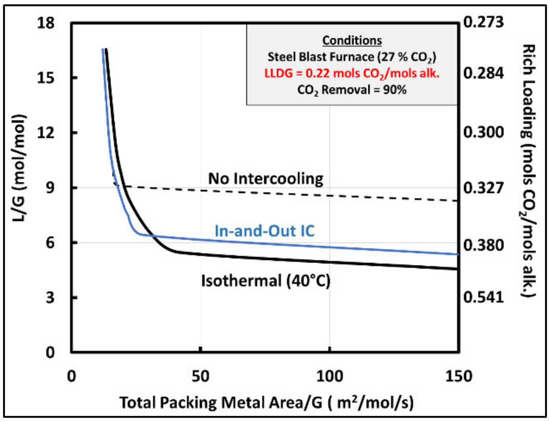

Figure 24: Design curves for adiabatic (**dashed**), in-and-out intercooling (**blue**), and isothermal (**solid**) operation for steel blast furnace application (27% CO_2) in the novel intercooling loading range (0.22 mol CO_2/mol alkalinity). Each curve represents the packing requirement to achieve 90% CO_2 removal for a given liquid to gas ratio (L/G). For each point on the curve, the lean loading, removal, and solvent rates are fixed, so a unique rich loading exists (secondary y-axis).

A.4. "Large Solvent Rate" Loading Range

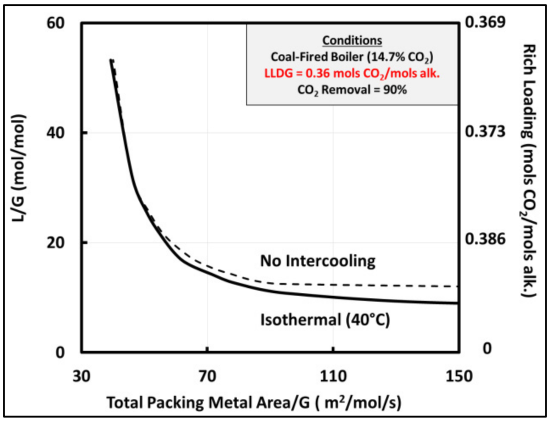

Figure 25: Design curves for adiabatic (dashed) and isothermal (solid) operation for coal-fired boiler application (14.7% CO_2) in the large solvent rate loading range (0.36 mol CO_2/mol alkalinity). Each curve represents the packing requirement to achieve 90% CO_2 removal for a given liquid to gas ratio (L/G). For each point on the curve, the lean loading, removal, and solvent rates are fixed, so a unique rich loading exists (secondary y-axis).

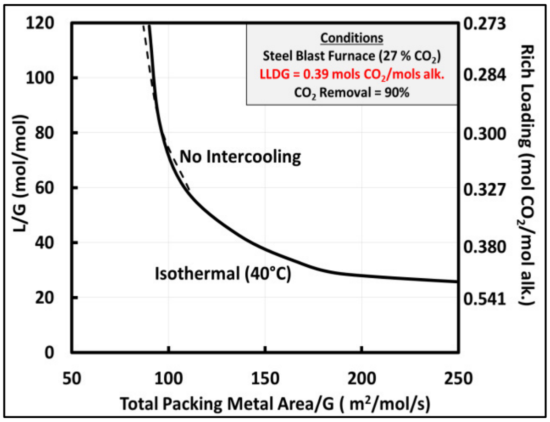

Figure 26: Design curves for adiabatic (dashed) and isothermal (solid) operation for steel blast furnace application (27% CO_2) in the large solvent rate loading range (0.39 mol CO_2/mol alkalinity). Each curve represents the packing requirement to achieve 90% CO_2 removal for a given liquid to gas ratio (L/G). For each point on the curve, the lean loading, removal, and solvent rates are fixed, so a unique rich loading exists (secondary y-axis).

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