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Optimal Design of Intensified Towers for CO_2 Capture with Internal, Printed Heat Exchangers

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ABSTRACT

Solvent-based carbon capture processes typically suffer from the temperature rise of the solvent due to the heat of absorption of CO_2. This increased temperature is not thermodynamically favorable and results in a significant reduction in performance in the absorber column. As opposed to interstage coolers, which only remove, cool, and return the solvent at discrete locations in the column, internal coolers that are integrated with the packing can cool the process inline, which can result in improved efficiency. This work presents the modeling of these internal coolers within an existing generic, equation-oriented absorber column model that can cool the process while allowing for simultaneous mass transfer. Optimization of this model is also performed, which is capable of optimally choosing the best locations to place these devices, such that heat removal and mass transfer area are balanced. Results of the optimization have shown that optimally placed cooling elements result in a significant increase in the capture efficiency of the process, compared to a similar column with no internal cooling, with a common trend being the cooling of the column in the temperature bulge region. It is observed that by optimally placing an internal cooler, the solvent flow rate can be decreased, and the CO_2 lean loading can be increased while still maintaining the same efficiency. These process changes can lead to a substantial reduction in costs due to lower reboiler duty.

Keywords: CO2 capture, optimization, monoethanolamine, process intensification

INTRODUCTION

Post-combustion CO_2 capture is a critical approach for achieving net-zero emissions. Among many potential technologies for post-combustion CO_2 capture, solvent-based capture technologies are at the forefront due to their maturity, ease of operation, availability of efficient contactors for solvent-based capture systems, and many other advantages.

A common family of aqueous solvents utilized in this process is one that contains amine functional groups. The solvent absorption process excels compared to other capture methods, such as solid sorbents, due to its ability to react with carbon dioxide even at low partial pressure, as well as having a high capacity of absorption [1]. There are, however, two key disadvantages associated with this process. The first disadvantage is that the high concentration of water in these solvents requires a significant amount of steam for the use of the reboiler in the regeneration process [2]. This steam utilization results in the energy intensity of the process being up to 4 MJ per ton of CO_2 captured, which accounts for the majority of the operating cost. The second disadvantage is the high exothermic heat of absorption for CO_2. In the case of a 30 wt% MEA solvent, the heat of absorption can range from 84 to 100 kJ/mol CO_2 [3, 4]. This heat can cause temperatures within the absorber tower to rise substantially, thus increasing the equilibrium pressure of CO_2 by several orders of magnitude, resulting in a reduced mass transfer rate [3].

To keep the solvent capture process operating as efficiently as possible, heat needs to be removed from the absorber column to operate the tower under more favorable conditions. Therefore, it is common to use interstage coolers [5, 6], which withdraw a portion of the solvent or the entire solvent from discrete locations within the column, and cool and return it in the next stage below. Karimi et al. showed that optimal placement of an intercooler within the absorber column can result in energy savings as high as 7.27% by reducing the amount of solvent flow required for operation [7]. However, while cooling is achieved at discrete locations, intercoolers fail to achieve continuous heat removal along the height of the tower to reach an optimal temperature profile to maximize performance and/or economics. Therefore, the use of a cooling method that is integrated with the packing of the tower is needed.

Additive manufacturing has become a rapidly growing method of developing innovative technologies, with advancements in 3D printing leading to products that would be impossible to create using traditional manufacturing methods. A recent novel application of 3D printing is structured metal packing that can be utilized for a packed column. The intensified packing device created at Oak Ridge National Laboratory is a 3D printed structured packing element with a double-walled design, which creates two disjoint flow channels in which the process fluid can be kept in a flow channel separate from the cooling fluid in the other [8]. Tests utilizing this device have shown that carbon capture can be increased between 3 to 15% compared to columns with no cooling [9, 10].

A disadvantage of such a device is that the cooling water channels reduce the total amount of available process volume, which reduces the mass transfer area. In addition, there are several other aspects that need to be evaluated for the optimal configuration of absorbers in the presence of such intensified towers. This work seeks to answer the following questions. Where are the optimal locations at which to implement internal cooling such that performance is maximized? What is the best configuration of cooling water flow within the devices? How does variance in operating set points affect the optimal design and performance of the internal coolers?

MODELING

Column Model

The absorber column model employed in this work was developed by Akula et al. [11]. This model is a rate-based solvent absorber model that can be applied to many processes by using the built-in physical and chemical property models. For convenience, this work uses 30 wt% MEA as the solvent.

For the reactive absorption process, rate-based models are significantly more accurate than equilibrium-based models, but are more computationally expensive due to the consideration of transport through the films, especially the liquid film. To reduce this computation expense through removing the need to model film interactions, an enhancement factor method is used that accounts for the increase in mass transfer due to chemical reactions by using a set of algebraic equations [12]. Equations 1 and 2 are used to describe the enhancement factor, E, in terms of dimensionless concentration of MEA, Υ^i_{MEA}.

$$E = 1 + (E^*_\infty - 1)\frac{(1-\Upsilon^i_{MEA})}{(1-\Upsilon^b_{CO_2})} \tag{1}$$

$$E = Ha\sqrt{\Upsilon^i_{MEA}\frac{(1-\Upsilon^*_{CO_2})}{(1-\Upsilon^b_{CO_2})}} \tag{2}$$

The phase energy balance of the model considers three heat transfer mechanisms/sources/sinks: convective heat transfer between phases, heat of absorption of CO_2, and heat of vaporization of water. The interphase heat transfer is calculated by using a corrected heat transfer coefficient, which is required for high mass fluxes (Equation 3). Both heat of absorption and vaporization are accounted for in the liquid phase energy balance (Equation 4). The heat of absorption is fixed at -85 kJ/mol CO_2 for simplicity [13], while the heat of vaporization being used was derived from [14].

$$Q_V = h'_V a_e (T_L - T_V) \tag{3}$$

$$Q_L = Q_V + N_{CO_2,V}\Delta H_{abs} - N_{H_2O,V}\Delta H_{vap} \tag{4}$$

Internal Heat Exchanger Model

The modeling for the internal heat exchanger packing was modified from Moore et al., which considered a single bed absorber column. This model was modified to account for individual discretized elements of the column in which an internal heat exchanger can be independently placed from other elements. In this model, it is assumed that all heat removal from the process to the cooling water occurs through the liquid phase due to the higher wetted area and conductivity in the liquid phase leading to a negligible amount of heat being directly transferred to the gaseous phase. It was also assumed to be smooth transition between standard and intensified packing, if flooding velocity is not surpassed. The two decision variables are a binary variable, y, for each column element to indicate placement of an internal heat exchanger and a voidage term, ε^{cw}, that accounts for the volume occupied by the intensified cooler. Equation 5 calculates this heat transfer in each column element where the overall heat transfer coefficient, assumed to be constant, is from [9], which is a conservative estimate using the packing area, rather than wetted area, and is based on experimental data. In this equation, i is length index of the column, U is the overall heat transfer coefficient, a is the specific geometric area of the packing, T^{cw} and T^L are the temperatures of the cooling water and solvent phase, respectively, and Q^{cw} is the heat transfer rate through the boundary. Equation 6 calculates the updated voidage by accounting for the volume occupied by the intensified coolers where ε° is the standard voidage of the packing being used, and ε is the resulting voidage of the process.

$$Q_i^{cw} = y_i U a_i \left(T_i^{cw} - T_i^L\right) \tag{5}$$

$$\varepsilon_i = \varepsilon^{\circ}{}_i - \varepsilon_i^{cw} \tag{6}$$

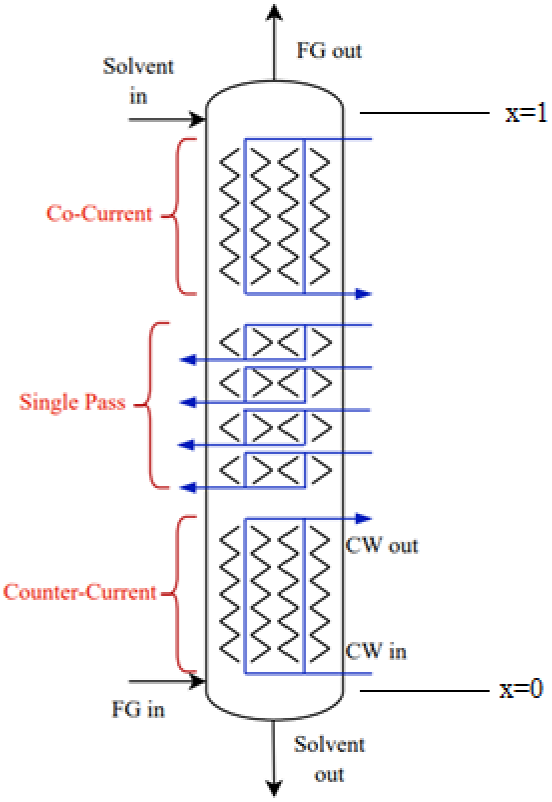

Figure 1. Configuration of cooling water flow through absorber tower

Modeling the different possible flow configurations of the cooling water through the intensified packing is necessary since different flow configurations can result in a significant difference in heat removal efficiency. Three different flow directions were included in this model: co- and counter-current flow, relative to liquid phase flow, and single pass flow. In this case, the single pass flow is a limiting case for the model since the cooling water enters and exits the column in the same finite element of the model resulting in the best case for heat removal rate. Figure 1 shows how each of these configurations moves cooling water through the column. These flow directions are selected through an integer variable, d^{cw}, which can take the values of -1, 0, or 1 corresponding to co-current, single pass, and counter-current configurations, respectively. The energy balance of the cooling water is performed by creating lower bound inequality constraints, which will be active depending on the value of d^{cw}.

To determine the best placement locations for the internal heat exchanger, an objective function is required. Three objective functions are utilized in this work, the first of these is minimization of CO₂ emissions, $F_{CO_2}^{V,out}$ (Equation 7). The next two are minimization of column height, H (Equation 8) and minimization of liquid to gas ratio (Equation 9) in which $F^{L,in}$ and $F^{V,in}$ is the molar inlet flow rate of the solvent and gaseous phases, respectively. An additional performance constraint is included with the last two objectives, which sets a lower bound on the allowed capture efficiency. These last two objectives were chosen as each can be used to estimate potential reductions in costs of the system since the absorber column is a large factor in the total capital cost and solvent flow rate is directly proportional to reboiler duty, which is greatest factor in operational costs.

$$\min_{\varepsilon^{cw},y,,d} F_{CO_2}^{V,out} \tag{7}$$

$$\min_{\varepsilon^{cw},y,,d} H \tag{8}$$

$$\min_{\varepsilon^{cw},y,,d} \frac{F^{L,in}}{F^{V,in}} \tag{9}$$

The absorber and internal cooler models were implemented using the IDAES platform, which is built on top of the Pyomo optimization suite [15].

RESULTS

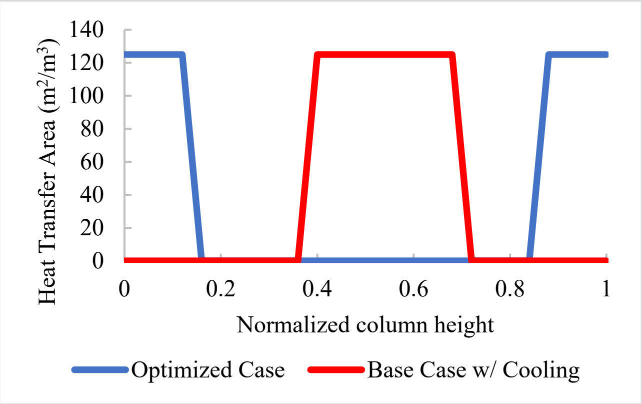

Figure 2. Optimal and suboptimal internal heat exchanger placement for minimizing CO₂ emissions.

The base configurations for the column design and process specifications are shown in Table 1. This first set of results utilizes the pilot scale configuration. Using this configuration, the placement of the internal heat exchangers was optimized by minimizing CO₂ emissions. Figure 2 shows the placement and area of the heat exchangers in the optimal solution, which were in the top and bottom 15% of the column. A separate case was also

Table 1. Column and process configurations for pilot-scale and process-scale models.

	Height (m)	Diameter (m)	Gas Inlet (mol/s)	CO_2 Gas Conc.	Liquid-Gas Ratio	CO_2 Lean Loading
Case 1 (Pilot Scale)	15	0.65	22	0.12	1.77	0.15
Case 2 (Process Scale)	20	12	12,000	0.042	1.83	0.22

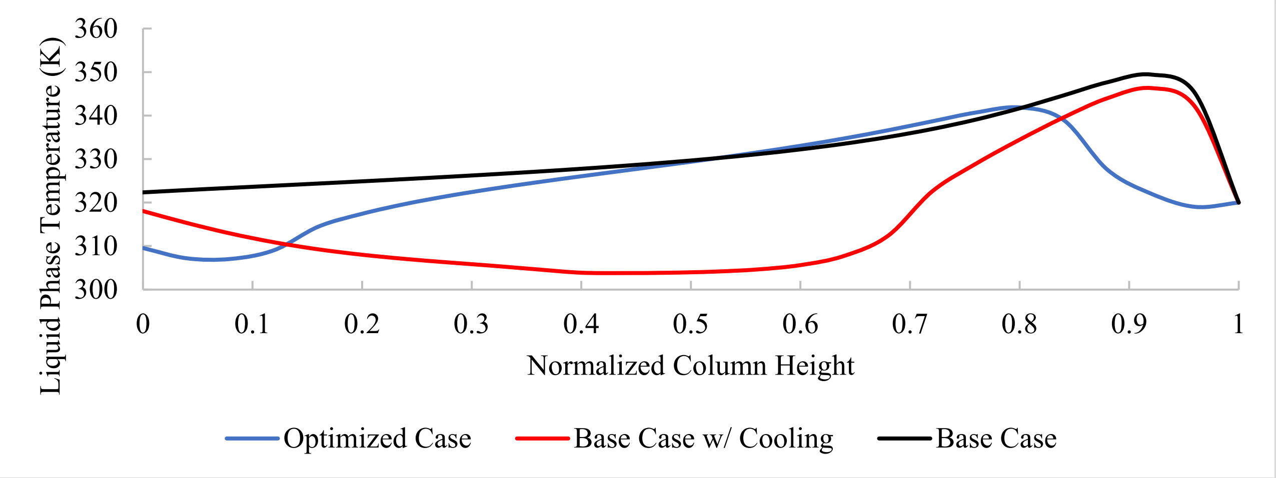

Figure 3. Solvent temperature profile for base case, base case with suboptimal cooling, and optimized case.

simulated in which the internal heat exchangers were placed in the center of the column with a similar amount of total available heat transfer area.

The liquid phase temperature profile for the base case without cooling, base case with suboptimal cooling, and optimized cooling are shown in Figure 3, with the capture efficiencies for each case shown in Table 2. The profile base case with out cooling exhibits a much higher average temperature across the length of the column. This higher temperature results in larger thermodynamic limitations to the mass transfer of CO_2, especially in the upper end of the column where a significant portion of mass transfer is occurring (indicated by the peak in temperature).The base case with suboptimal cooling appears to have a greater extent of heat removal when compared to the optimal case but results in lower capture efficiency. This is due to the given operating conditions. As previously mentioned, the majority of CO_2 mass occurs at the top 10–20% of the column, which is where the peak of the temperature bulge caused by the absorption energy is located in the base case. This causes this section of the column to be heavily thermodynamically limited, as opposed to physical limitations, such as mass transfer area, which explain the optimality of placing an internal cooling element in this region. The temperature profile in the bottom 15% of the column is comparatively level indicating very little mass transfer of CO_2. In this region, mass transfer is being solely limited by the reduced driving force due to higher loading in the solvent, which results in placement of the intensified packing as a means to further increase the total amount of heat removed from the system.

Table 2. Comparison of capture efficiency for cases with and without internal cooling

Case	Capture Efficiency
Base Case	72.46%
Base Case w/ Cooling	75.63%
Optimized Case	76.94%

As opposed to the top region of the column, the middle region is mass-transfer limited; therefore, maximizing mass transfer area to obtain high performance is desired. Finally, the bottom section of the column becomes thermodynamically limited; therefore, the optimal configuration is to include the cooling section there.

Using these optimized placements, a study was then conducted that investigated the variance of capture performance in different cooling water flow directions and flow rates. Both counter-current and co-current configurations were simulated using cooling water flow rates between 1.5 and 50 mol/s. The capture efficiencies for each of these configurations are shown in Figure 4. Below 20 mol/s of cooling water, the co-current configuration shows a slight advantage, which is due to how the solvent temperature changes in the upper internal cooler section. Analyzing these temperature profiles shows that the solvent enters the upper heat exchanger at the inlet temperature of 320 K and exits it at 338 K at a column height of 0.85. So, if cooling water is flowing in the counter-current direction, there is a pinch at the top of the column limiting the heat removal rate. However, once flow rates increase above 20 mol/s, the difference in performance between the two configurations is nearly the same.

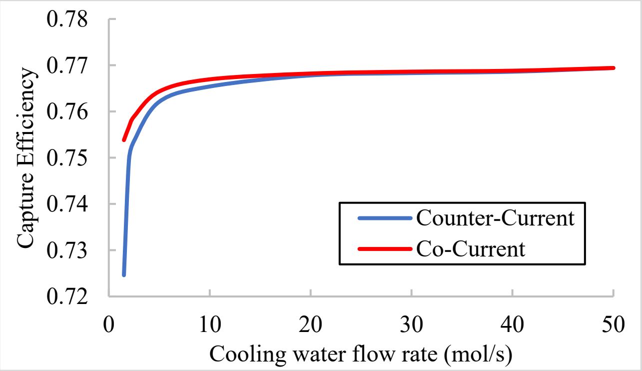

Figure 4. Capture efficiency at varying cooling water flow rates and flow direction.

The next results shown are from using the objective functions for minimizing the height and liquid-gas ratios (Equations 8 and 9). These objective functions are useful since each of these values correlates with major costs within the process, in which the column height accounts for an investment cost and the solvent flow correlates with the steam requirement in the reboiler. For these studies, the process scale model configuration was utilized, and each aspect was optimized at different CO_2 lean loadings and at different minimum capture efficiencies. The minimization of the column height in Figure 5 shows a maximum reduction of 6 meters when operating at a lean loading of 0.25 with a capture efficiency of 94%. The minimization of the liquid gas ratio in Figure 6 shows that the solvent rate can be reduced by upwards of 20% at the same lean loading of 0.25 operating at a capture efficiency of 91%.

Results for the column height minimization are shown in Figure 5. This data can be interpreted in two ways. When picking any point along the dotted line representing a column without internal cooling, moving down from this point to the corresponding solid line shows how much the column height can be reduced at a fixed capture efficiency. Alternatively, the horizontal shift shows how the capture efficiency can be increased with optimally placed internal coolers at a fixed column height. The improvement of reduced column height when using internal cooling units increases at higher capture rates and higher lean loading of the solvent. This same trend can be seen in the results for minimizing the liquid-gas ratio (Figure 6).

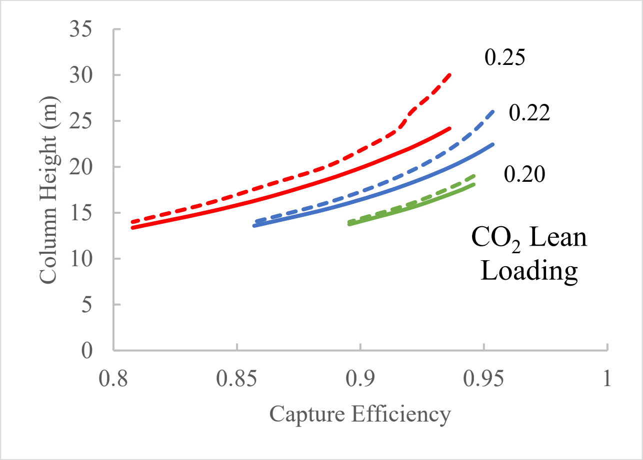

Figure 5. Variation is column height subject to capture efficiency, and lean loading without internal cooling (dashed line) and with internal cooling (solid line).

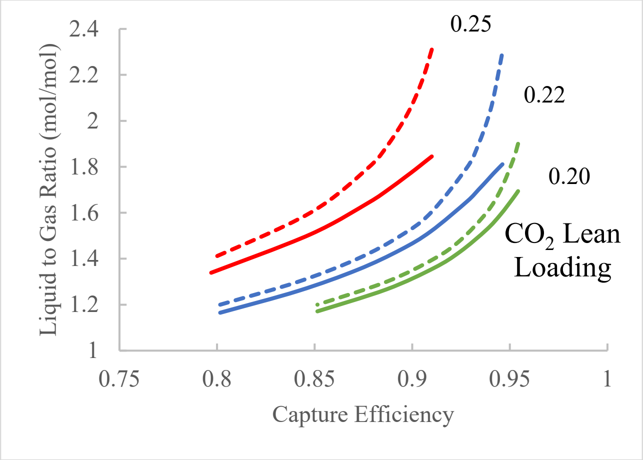

Figure 6. Variation is column height subject to capture efficiency, and lean loading without internal cooling (dashed line) and with internal cooling (solid line).

The reduction in both column height and solvent flow rate with the use of intensified packing has substantial implications on the cost saving for the process. The ability to reduce the height absorber bed, by up to 6 meters, while still retaining equal capture performance suggests that capital costs can be significantly reduced. Similarly, being able to reduce solvent flow rate by up to 20% corresponds with a similar reduction in the operating costs of the steam reboiler in the solvent capture process, which is a major factor in the determination of the levelized cost of capture of CO_2. Due to the infancy of this technology, an accurate cost model for the intensified packing is still required to allow for a robust economic optimization of the process to determine the trade off of these potential cost savings for the cost of the packing.

There are still other factors of this technology yet that need to be further investigated. The first of which is overall heat transfer coefficient of the packing. In a dynamic process, this coefficient is likely to vary based on the variance of the loading of flue gas to the column which can influence the heat removal rate. In terms of performance rating, due to the significant reductions found in process conditions through placement optimization and the use of a conservative value for heat transfer coefficient, the intensified packing is likely to retain an advantage over conventional structured packing even with a wide margin of uncertainty in heat transfer performance. Another factor to further consider is the comparison in performance to and absorbent system utilizing intercoolers, which again raises the need for an accurate cost for the intensified packing to determine if the cost of the complexity of the design outweighs the benefit of providing in-line, continuous cooling to the process.

CONCLUSION

An existing model for solvent-based carbon capture is modified to implement a model for an internal cooling element that is integrated within the structured packing of the column. The internal cooling model was designed so that the placement of these elements can be used as a decision variable in an MINLP problem. The flow direction of the cooling water is also taken into consideration with the possibility of co- and counter-current flows, along with a single pass option.

The results of this work have shown the feasibility of utilizing internal heat exchangers to boost the capture performance of a solvent capture system. Depending on the configuration, the capture efficiency is shown to increase as much as 5%, which can account for a significant reduction in CO_2 emissions being released into the atmosphere. As shown, the optimization for the placement of internal heat exchangers is necessary since a deviation from optimum placement leads to a reduction in capture efficiency. The results also show that when comparing co- and counter-current flows of cooling water through these elements, it is optimal to use the co-current configuration. In the counter-current configuration, a pinch point is created that significantly affects the effectiveness of cooling.

The case studies on optimizing the height and solvent flow rates have shown that significant reductions of up to 20% in each can be made. This reduction is even greater when operating at higher capture efficiencies, which can make this technology ideal for that area of implementation. Although a reduction in the absorber column height can result in significant cost reduction, the real benefit of implementing internal coolers is the reduction in steam costs from reboiler operation. This cost is heavily affected by the solvent flow rate and CO_2 loading of the lean solvent. Thus, the placement of an intensified absorber not only improves the performance of the absorber but the economics of the overall process due to the resulting effect on the stripper operation.

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DISCLAIMER

This project was funded by the U.S. Department of Energy, National Energy Technology Laboratory an agency of the United States Government, through a support contract. Neither the United States Government nor any agency thereof, nor any of its employees, nor the support contractor, nor any of their employees, makes any warranty, expressor implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

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