Simulation of the Energy Consumption of CO_2 Capture by Aqueous Monoethanolamine in Pilot Plant

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Abstract

Monoethanolamine is a conventional solvent used for capturing carbon dioxide from coal fired power plants. Its stable performance makes it a good benchmark to explore other absorbent or technological processes. In this paper, process simulations based on Aspen Plus are conducted to identify how operating parameters influence energy consumption and efficiency for the carbon dioxide capture. Experimental results carried out in a pilot plant will be reported along with the comparisons to the simulated results. Comparative discussion against existing publications in the same area regarding the simulation validity is also carried out.

Keywords: Carbon dioxide, Simulation, Monoethanolamine, Carbon capture, Pilot plant

1. Introduction

Global warming has become an unequivocal issue and its multitude of negative impacts on human lives and a sustainable environment are looming more urgently than ever before (Safire, 2007). The 2011 United Nations Climate Change Conference is an ongoing conference, held in Durban, South Africa, from November 28th to December 11th, 2011, which is another landmark to combat global warming since the Kyoto Protocol is expiring in 2013 (UNFCCC website, 2011). In the United States, the White House announced on Nov. 25th, 2009 that President Obama offered a U.S. target for reducing greenhouse gas (GHG) emissions in the range of 17% below 2005 levels by 2020 (Whitehouse website, 2009). World energy-related carbon dioxide emissions will grow from 29.0 billion metric tons in 2006 to 33.1 billion metric tons in 2015 and 40.4 billion metric tons in 2030 (EIA website, 2006). In the United States, energy–related carbon dioxide (CO_2) emissions alone constitute 83% of emissions from all sources on a CO_2 equivalent basis, and the process of generating electricity is the single largest source of CO_2 emissions from fossil fuel combustion, approximately 41 % in 2006 (EPA website, 2011).

There has been a vast array of technologies proposed to capture CO_2 from existing coal-fired power plants. Presently, processes such as chemical solvent scrubbing, physical solvent scrubbing, adsorption, membranes, cryogenics, and oxy-fuel technology are all capable of removing CO_2 from gas streams. Among these technologies, chemical solvent scrubbing is the most developed and commonly used method, and is the only commercialized technology to date. Using aqueous amine solvents to capture acidic gases has a long history, and it can be traced to the first commercially available gas – treating plants in 1930. But until the late 1970s, this technology was only utilized for enhanced oil recovery operations, not in response to the concern about the greenhouse effect (Kohl & Nielsen, 1997).

Several of the large commercial plants using flue gas as a CO_2 feed source have been summarized by Global CCS Institute (CCS website, 2011). Currently, nearly all large commercial plants use the monoethanolamine (MEA) based solvent to capture CO_2. Chapel has summarized the main properties of MEA – based solvents as absorbents for food grade CO_2 production from flue gas in commercial power plants (Chapel, Mariz, & Ernest, 1999). There are several benefits to using MEA to capture CO_2 at low partial pressures. Three key advantages to using this approach include (1) The low molecular weight of MEA results in a high solution capacity at high concentrations on a weight basis; (2) In the MEA molecule, the nitrogen functional group produces a high alkalinity for this primary amine; (3) MEA has a high reaction rate with CO_2, which is especially important when the CO_2 concentration is low as is the case in power plant post-combustion flue gas. There are two major disadvantages with using MEA however. The first is that MEA has a high heat of consumption for regeneration compared with other amines, and the second is that MEA solutions are more corrosive than most other amine

solutions, particularly if the amine concentration exceeds 20% and the solutions are highly loaded with acidic gas (Kohl & Nielsen, 1997).

Despite the high energy consumption, MEA is an ideal solvent to use as a benchmark to explore other solvents in traditional CO_2 absorber/stripper systems due to its stable performance. Several pilot plants have been established, and a series of tests have been carried out for design variables and process parameter evaluation (Wilson *et al*., 2004; Yokoyama, 2004; Dugas, 2006; Mangalapally *et al*., 2009; Kvamsdal *et al*., 2011). Process simulations of MEA-based solvent CO_2 capture through an absorption/stripping operation offer a valuable alternative method to help identify the key process variables and design parameters (Desideri & Paolucci, 1999; Abu-Zahra, Schneiders, Niederer, Feron, & Versteeg, 2007; Khan, Simioni, Van Zijll de Long, & Stevens, 2007; Kvamsdal *et al*., 2011). The ratio of solvent circulation rate to flue gas rate (L/G), identified as an important operation parameter for solvent based CO_2 capture process, has yet to be thoroughly scrutinized (Freguia & Rochelle, 2003; Hassan, 2005; Abu-Zahra, Schneiders, Niederer, Feron, & Versteeg, 2007). Developing a correlation between L/G and the CO_2 capture efficiency through simulations can determine the optimal combination of economics and efficiency at the set conditions. Furthermore, simulations provide an easy way to explore the effect of other variables on energy consumption, which could aid in the development of new solvents in the future.

The Center for Applied Energy Research (CAER) at the University of Kentucky has constructed a CO_2 absorber/stripper combined pilot plant, as shown in Figure 1, to explore and test for cost effective technologies for CO_2 capture under coal-derived flue gas conditions. The scale of the pilot plant is considered to be appropriate to obtain engineering data required for pre-demonstration.

In this paper, an electrolyte phase equilibrium model was built in the Aspen Plus environment to identify the effects that different operating parameters have on CO_2 capture efficiency and energy consumption. Then a series of tentative experiments was conducted and an explanation for the experimental results based on the simulation results is given. In addition, some constructive suggestions were derived from the simulation results for further experimental design and development.

The equilibrium model is not only "economic" and "simple", but may be more suitable for the energy-related research in the pilot plant system (Abu-Zahra, Schneiders, Niederer, Feron, & Versteeg, 2007; Kvamsdal *et al*., 2011). The comparison of energy consumption for different solvents could only have meaningful results at each other's optimal operating conditions (Mangalapally *et al*., 2009). The equilibrium is one of the optimal conditions which could be approached or approximately approached by reasonable design, such as absorber column size or packing selection. This is especially valid for MEA, which has high reaction rate with CO_2. Rate-based models are more dependent on specific pilot plant configuration and may produce different results even for the same solvent and similar operating conditions. These models are more suitable for other parameter research such as temperature or concentration distribution in the absorber rather than energy-related parameters (Lawal, Wang, Stephenson, & Yeung, 2009).

2. Simulation Process

The simulation model is constructed using Aspen Plus software, which is the most commonly used tool in chemical engineering. Figure 2 shows the flowsheet of the absorber/stripper combined system as displayed in Aspen Plus.

The simulated flue gas entering the absorber column from the bottom stage flows up through the column, countercurrent to the lean aqueous solvent. CO_2 in the flue gas chemically reacts with the amine solvent, while the purified gas is vented to the atmosphere from the top of the absorber, and the solvent enriched by CO_2 is pumped to a rich/lean solvent cross heat exchanger for energy recovery. The rich solvent enters the top of the stripper where it flows down through the column to the reboiler and returns to the absorber from the bottom to finish one circulation.

The selected method in Aspen Plus to describe the interaction between CO_2 and the amine solvent is electrolyte NRTL, and the absorber and stripper are chosen to be RADFRAC columns. The absorber is specified as a four-stage column with the flue gas introduced from the fourth stage and the amine solvent from the first stage (counted from the top). The stripper is specified as a three-stage column with rich solvent introduced from the first stage, and lean solvent discharged from the reboiler located at the bottom of the stripper. The considered reactions that occur between amine aqueous solvent and CO_2 are (Austgen, Rochelle, Peng, & Chen, 1989; White, 2002):

$$MEACOO^- + H_2O \leftrightarrow MEA + HCO_3^- \tag{1}$$

$$CO_2 + H_2O \leftrightarrow HCO_3^- + H^+ \tag{2}$$

$$HCO_3^- \leftrightarrow CO_3^{2-} + H^+ \tag{3}$$

$$MEAH^+ \leftrightarrow MEA + H^+ \tag{4}$$

$$H_2O \leftrightarrow H^+ + OH^- \tag{5}$$
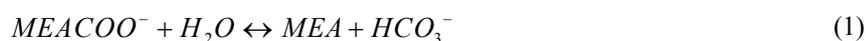
When using this model, several general assumptions must be made which are listed below (Lawal, Wang, Stephenson, & Yeung, 2009):

1)   Thermodynamic equilibrium is kept throughout the process.

2)   Henry's law-type mass transfer for the gas to liquid phase for CO_2, O_2 and N_2.

3)   An adiabatic absorber process.

To make convergence easier and accelerate calculations, several more steps are taken as follows:

1)   The rich/lean heat exchanger is set as two separate heaters. The outlet temperature of the rich solvent from this heater is specified based on the experimental measurement. According to the actual run data, illustrated in Figure 3, the rich loading solvent outlet temperature exiting from the heater B4 is set to be 75 ºC. This step substantially improves the calculation efficiency because of avoiding the coupling calculation of the lean/rich streams, which often leads to highly unstable results or failure to converge.

2)   An open circle flowsheet is first adopted for the system to obtain a convergent solution under the specified conditions. A closed circle system is finally introduced to fulfill other parameter demands (Alie, Backham, Croiset, & Douglas, 2005).

3)   Direct substitution method was selected as the convergence method. It is more possible to get to a convergent result than other methods even if it may be a more time consuming option.

Based on the actual pilot plant operating conditions, both absorber and stripper are simulated at a pressure of 1 atm. The absorber temperature is simulated at 40 ºC, and the stripper temperature is decided by reboiler heat input, CO_2 capture efficiency and solvent flow rate.

3. Results and Discussion

3.1 Energy consumption and relevant variables

A flue gas rate of 4.72×10^{-3} m³/s is introduced in the simulation to explore the correlation between energy consumption and CO_2 capture efficiency. The premixed gas composition is 14 vol% carbon dioxide, 6 vol% oxygen and 80 vol% nitrogen, which is the most common content in coal-based power plant flue gas. The simulated flue gas temperature is set to 25 ºC. A 30 wt% MEA concentration is used in the solvent.

Figure 4 illustrates the correlation between energy consumption and CO_2 capture efficiency. The circulation solvent is chosen to meet the request of L/G to be 8.02m³/1000m³. The energy consumptions at different efficiencies show only a slight increase and could be considered stable over a wide efficiency range. The ramp energy change only happens when the efficiency is greater than 93%. Before this critical point, the specific energy consumptions are approximately 4300 kJ/kg CO_2 captured.

The component parts for the energy used for every unit CO_2 capture can be expressed as follows (Chakma, 1997):

Total Energy=Heat of Reaction + Sensible Heat + Latent Heat of Vaporization of Water + Latent Heat of Vaporization of MEA

Under all the varied run conditions, only aqueous MEA solvent is used to react for CO_2 regeneration, so the heat of reaction can be treated as a constant. Also, the high boiling point of MEA compared to that of water makes the amount of vaporized MEA negligible when compared with the amount of vaporized CO_2 and water. With these assumptions, only two terms in the formula need to be considered for their contribution to the total energy change: Sensible heat and latent heat of vaporization of water.

The sensible heat can be determined by the solvent's mass, the specific heat of the solvent and the temperature difference. Specific heat can be treated as a constant in all processes, so the specific heat will not be a factor in the change in sensible heat calculation. Also, the gas flow rates and L/G is held constant, therefore the solvent mass also can be treated as a constant and not affect the change in sensible heat. The rich solvent input temperature is measured and set to be 75ºC, so the temperature at the top and bottom stages of the stripper can be

taken as the sole indicator of the change in sensible heat in the simulation.

The latent heat of vaporization of water is dependent on the specific latent heat and the amount of water. In this process, the specific latent heat can be treated as a constant, so the amount of water vaporized becomes the only effective term. Under the consideration that total energy is based on every unit CO_2 captured, the mole ratio of water to CO_2 from stripper exhaust is proper to be used as the indicator for latent heat of vaporization of water. Figure 5 shows the relationship between top/bottom stages temperature with CO_2 capture efficiency, and Figure 6 shows the relationship between mole ratio of H_2O to CO_2 and CO_2 capture efficiency.

The curves in Figures 5 and 6 display a pattern similar to that in Figure 4. In Figure 5, the temperatures in both the top and bottom stages of the stripper slowly increase as the CO_2 capture efficiency increases. These temperature increases in the stripper contribute to the sensible energy increase; however, it should be noted that the CO_2 capture efficiency increases as well. This means the impact on sensible heat caused by the temperature increases in the stripper may be offset by the increased amount of CO_2 captured. In Figure 6, the mole ratio of H_2O to CO_2, which represents the latent heat of vaporization of water, shows a gradual increase and may represent the most significant contribution to the increase in total energy.

Since there is only a slight increase in energy consumption when compared at different efficiencies (shown previously in Figure 4), it seems highly possible to achieve a higher CO_2 capture efficiency at little or no additional energy costs. On the other hand, this process feature can provide a realistic estimate for energy consumption at high efficiencies using the data obtained at the same run conditions but at a lower efficiency. Running the process at a lower efficiency is often easier, and in some situations is the only option because higher efficiencies cannot be reached due to the capacity limits on the equipment.

3.2 Effect of L/G on energy consumption

To investigate the impact of L/G on the energy consumption of the process, some parameters need to be set. During these experiments, the flue gas flow rate was held at 4.72×10^{-3} m^3/s, so the L/G variable was manipulated solely by changing the solvent flow rate. Figure 7 illustrates the relationship between energy consumption and CO_2 capture efficiency at different L/G values while maintaining the same gas flow rate.

Without considering the ramp energy changes at high efficiencies (>93%), the general trend of the curves shows that at greater L/G values there is a reduction in energy consumption when compared at the same capture efficiency. In other words, processes run at lower L/G values cannot reach ideal CO_2 capture efficiency if the heat output of the reboiler is kept constant due to an increased energy requirement at the lower L/G. It can be noted that the energy difference between the adjacent L/G curves tends to decrease at the same efficiency as L/G increases. This strongly suggests the existence of an optimal L/G for specific run conditions.

Figure 8 illustrates the relationship between the temperature at the top and bottom stages of the stripper and efficiency. Figure 9 shows the relationship between the mole ratio of H_2O to CO_2 and the efficiency. These findings show that both stage temperature and the mole ratio of H_2O to CO_2 decrease along with the L/G increases in the simulated range. However, the difference between the stage temperature and the mole ratio of H_2O/CO_2 also decreases with the L/G increases.

Although the lower stage temperatures would have a positive effect on the total energy consumption by reducing the sensible heat, the increase of L/G by adding more solvent to the system would more than likely offset any savings.

The smaller mole ratio of H_2O to CO_2 means that for each unit of CO_2 vaporization, there is less water being vaporized with it in the stripper. This is a favorable result because it would indicate that less energy would be consumed during the process for the vaporization of water. Although an escalation of the ratio of H_2O to CO_2 with greater L/G values is not observed in the simulated L/G range, it is a reasonable presumption. For a given gas flow rate, the maximum CO_2 vaporization would equal the feed amount, and the stripper temperature would have to be kept above some limit to maintain the absorber/stripper circulation. This temperature limit would support increasing water vaporization with rising L/G values at the given gas flow rate. Due to the constraint of the CO_2 vaporization maximum by the given gas flow, the increasing water vaporization would cause the ratio of H_2O to CO_2 to increase after a certain L/G point. At that L/G, both sensible and latent heat would increase, which means specific energy consumption would increase more quickly afterward, and the optimal L/G should exist at or before that point.

3.3 Experimental and simulated results comparison

The pilot plant at the Center for Applied Energy Research (CAER) of the University of Kentucky is designed to explore and test cost effective solvent-based technology for post-combustion CO_2 capture. The main components

of the pilot plant are the absorber and the stripper. The flue gas is generated by mixing three individual lines of air, nitrogen, and carbon dioxide before it is supplied to the absorber column. The gas composition is set to 14 vol% CO_2, 6 vol% O_2 and 80 vol% N_2 by separate mass flow controllers and can be directly read out from the LabView program on the computer.

The absorber is composed of a cylinder-shaped column (5.5 m high and 0.1 m ID) made of polyvinyl chloride (PVC) and packed with 1/2" (1.27 cm) ceramic Berl saddles. The column consists of four equal sections with a total packing height of 4.8 m. The solvent enters the column from the top, and the flue gas enters the column from the bottom allowing for the maximum amount of time for interaction. The CO_2 reacts with the solvent and is separated from the rest of the flue gas as it gets carried to the stripper.

The stripper is made of stainless steel and built of three sections, which are each equipped with four elements of the structured packing Mellapak 250Y. The total packing height in the stripper is 2.52 m, and the column has a diameter of 0.10m. The reboiler circulates a hot oil to heat up the solvent in the stripper. The rich solvent goes through vaporization process and becomes lean solvent. The latter will flow through the heat exchanger and back to the absorber to begin a new circulation.

A series of primary experiments have been carried out in the pilot plant at UK's CAER using MEA aqueous solvent as an absorbent to determine the influence of operating parameters on CO_2 capture efficiency. In the experiments, the reboiler at the bottom of the stripper provides the stable heat flux at 9 kW, so that the total energy input can be treated as a constant. With a set gas flow rate and total energy input, a particular CO_2 capture efficiency and specific energy consumption can be determined. Figure 10 presents the experimental results showing that CO_2 capture efficiency increases noticeably between L/G values of 4.01 and 5.35 $m^3/1000m^3$. It is also noticed for the L/G higher than 5.35 $m^3/1000$ m^3, the efficiency doesn't change much with the L/G variation in the researched range, which means the operating range of L/G to keep a suitable CO_2 capture efficiency is wide.

Regarding the different gas flow rates at the same L/G, from an equilibrium standpoint, differing flow rates should not impact the specific energy consumption. If the total energy inputs are the same, CO_2 capture efficiency would decrease with the increasing gas flow rates due to an unchanging specific energy requirement. Figure 10 shows this general trend when comparing the efficiencies at the same L/G condition between different gas flow rates; however, the efficiency differences between the different gas flow rates at the same L/G vary, which suggests that the specific energy consumption at different gas flow rates may not be the same. Figure 11 presents the specific energy consumptions for these experiments, which is consistent with the presumption above.

Since many variables may affect the actual specific energy consumption, here a tentative effort is performed to introduce a manipulated coefficient that will reconcile the calculated theoretical energy and the actual measured data. Assuming the coefficient is specific to gas flow rate, Figure 12 shows the theoretical and modified experimental specific energy consumption at different L/Gs for three gas flow rates. At each gas flow rate, the modified experimental specific energy is correlated to the simulated specific energy. These adopted ratios are applied as the correction factor to the experimental data. The coefficients are 1:1.8, 1:1.6, and 1:1.4 for gas flow rates of 4.72×10^{-3}, 5.66×10^{-3} and 7.08×10^{-3} m^3/s, respectively. For L/G values ranging from 5.35 to 8.02 $m^3/1000m^3$, the modified experimental data generally match the simulation results. The absolute deviations are less than 2.5%. However, it is noticeable when L/G is 4.01 $m^3/1000m^3$, the mismatch of modified experimental data and simulation results for energy consumed increases for the system.

Though the mismatch between simulation and experimental data is not uncommon (Freguia & Rochelle, 2003), comparative discussion against the existing publications in the same area is necessary. Compared with other simulation and experimental results, the simulated energy consumption locates in the same range (Abu-Zahra, Schneiders, Niederer, Feron, & Versteeg, 2007; Kvamsdal *et al*., 2011). The experimental results in the literature are closer to the simulated results than to results given here from pilot plants with similar configuration and operating conditions. Especially, a dramatic specific energy increase is also observed which suggests the validity of this simulation (Mangalapally *et al*., 2009). Moreover, for the mismatch in Freguia's results (Freguia & Rochelle, 2003), the author's simulated results are actually 10 to 30% higher than the experimental ones, contrary to the results here. Considering the experiment here as only a preliminary practice, it suggests some aspects of the experimental design need to improve to increase the CO_2 capture efficiency or save specific energy.

Figure 13 presents the relationship between L/G and solvent CO_2 rich/lean loading using the simulated results. It could be found that the rich loading is constant at about 0.52, and the lean loading increases from around 0.31 to

0.39 with the increasing L/G for all the three gas flow rates. It should be noted that the decrease of the difference between rich and lean solvent is a result of the increase of L/G necessary to keep the same CO_2 capture efficiency. The constant rich loading suggests a "saturated" solvent in the researched conditions and means that the lean loading could be in direct correlation to L/G along with the CO_2 capture efficiency. However, the measurement facility of L/G makes it more suitable to be selected to correlate with other operating parameters.

Overall, the modified experimental data seems to fit to the simulated curves to some extent. In the future, more work needs to be done to determine the reason for higher energy consumption in the pilot plant compared to the simulated results. On this basis, additional experiments could proceed for other promising solvents. The simulation research could be used in this process to determine the most suitable operating conditions.

4. Conclusions

Several conclusions can be drawn from the simulated results along with experimental data:

1) Regarding a given L/G, specific energy consumption will slightly increase as the efficiency increases over a wide efficiency range, which makes it possible to pursue higher CO_2 capture efficiencies for a similar cost.

2) L/G is a crucial parameter in this process. In the researched range, at a defined energy input, an increase in L/G can improve the CO_2 capture efficiency. The extent of benefit for the efficiency increase reduces as the L/G increases, which suggests that an optimal L/G would exist. At this point, the process would obtain the most economic CO_2 capture.

3) The change in the mole ratio of H_2O to CO_2 is an important contributor to the change of energy consumption. Use of cost effective methods to gain a lower ratio of H_2O to CO_2 may be a key point for an energy saving campaign.

4) The primary experiments conducted in the pilot plant support the main points drawn from the simulation. More detailed experiments need to be developed in order to expand the data matrix and provide a more comprehensive comparison with the simulation results.

Acknowledgments

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Figure 1. CO_2 absorber/stripper combined pilot plant at UK CAER
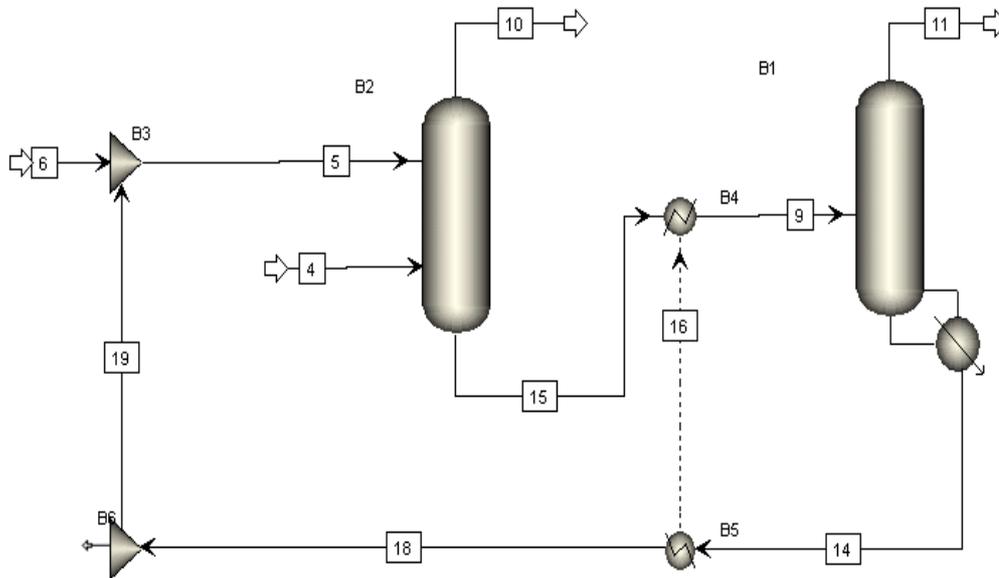
Figure 2. Schematic diagram of CO_2 absorber/ stripper system in Aspen Plus
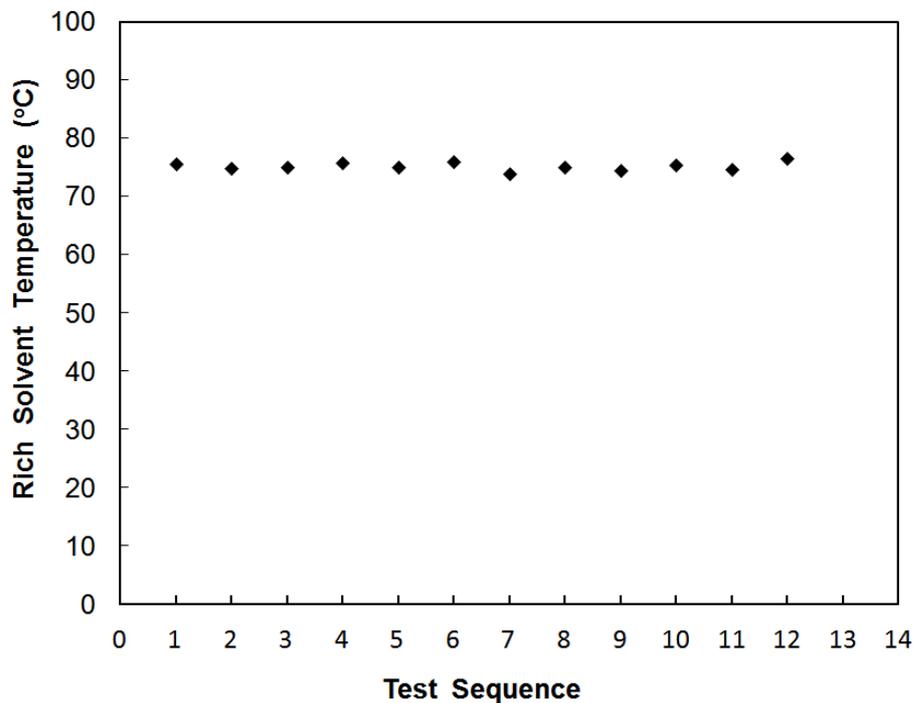
Figure 3. Temperature profile of rich CO_2 loading solvent at different run cases
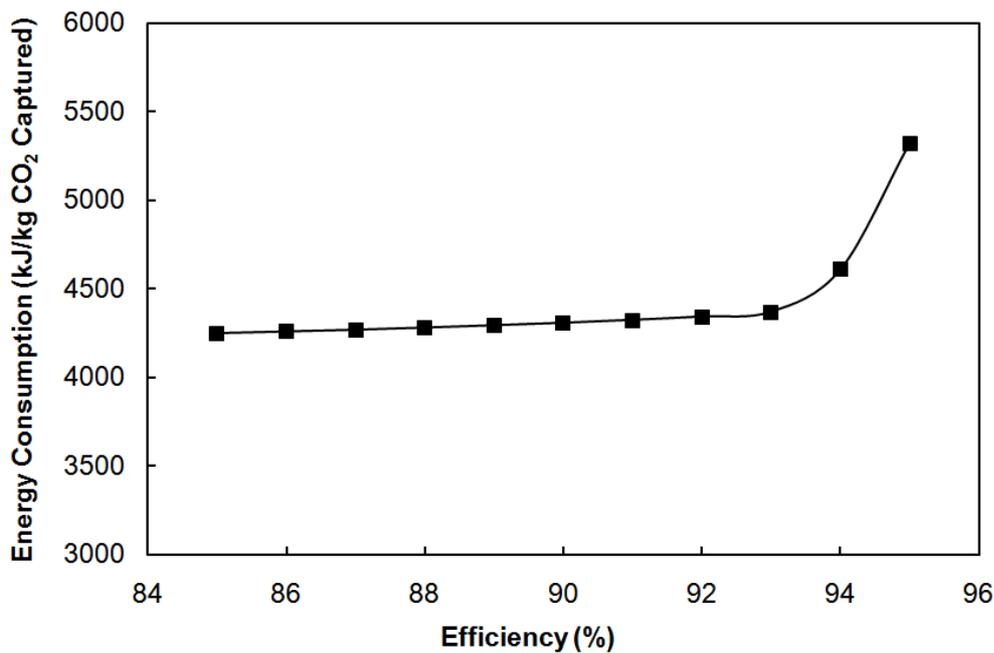
Figure 4. The relationship between specific energy consumption and
CO_2 capture efficiency at a gas flow rate of 4.72×10^{-3} m^3/s with L/G=8.02 $m^3/1000m^3$
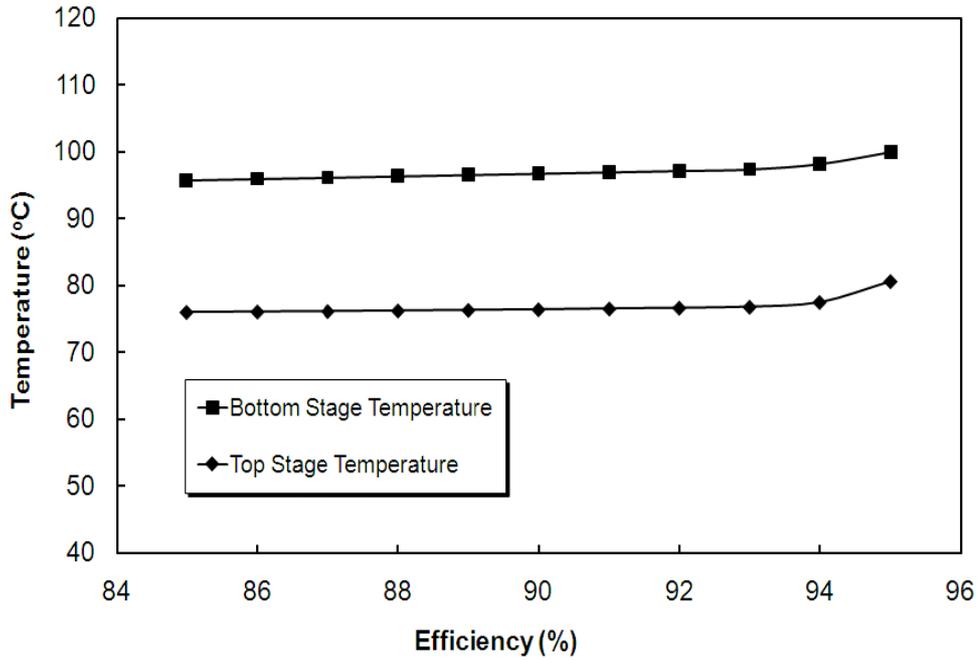
Figure 5. The relationship between top/bottom temperature and
CO_2 capture efficiency at the gas flow rate of 4.72×10^{-3} m^3/s with L/G=8.02 m^3/1000m^3
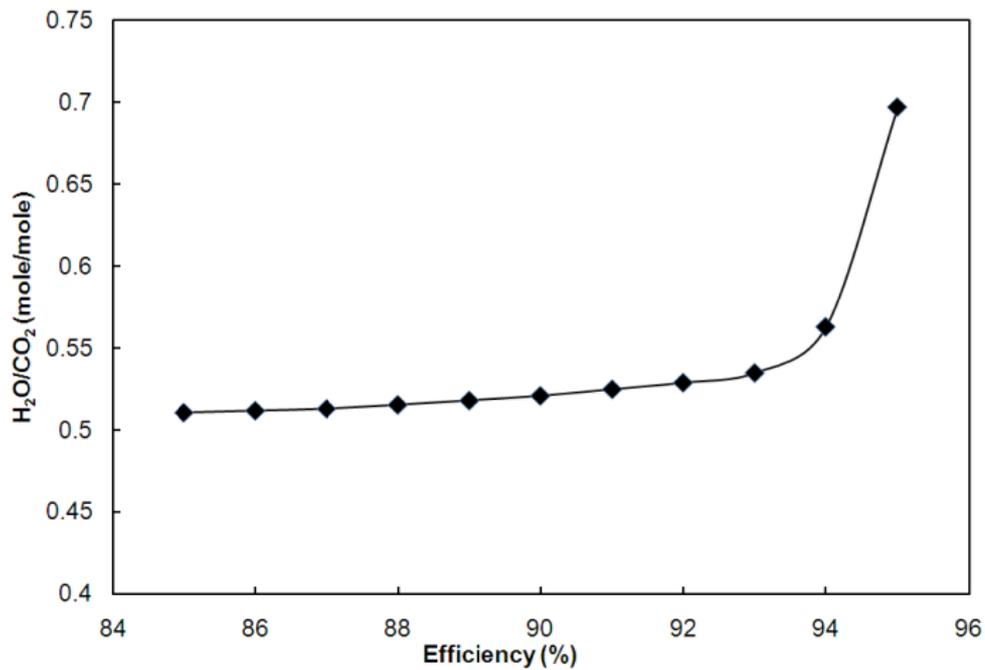
Figure 6. The relationship between H_2O/CO_2 mole ratio and the
CO_2 capture efficiency at the gas flow rate of 4.72×10^{-3} m^3/s with L/G=8.02 m^3/1000m^3
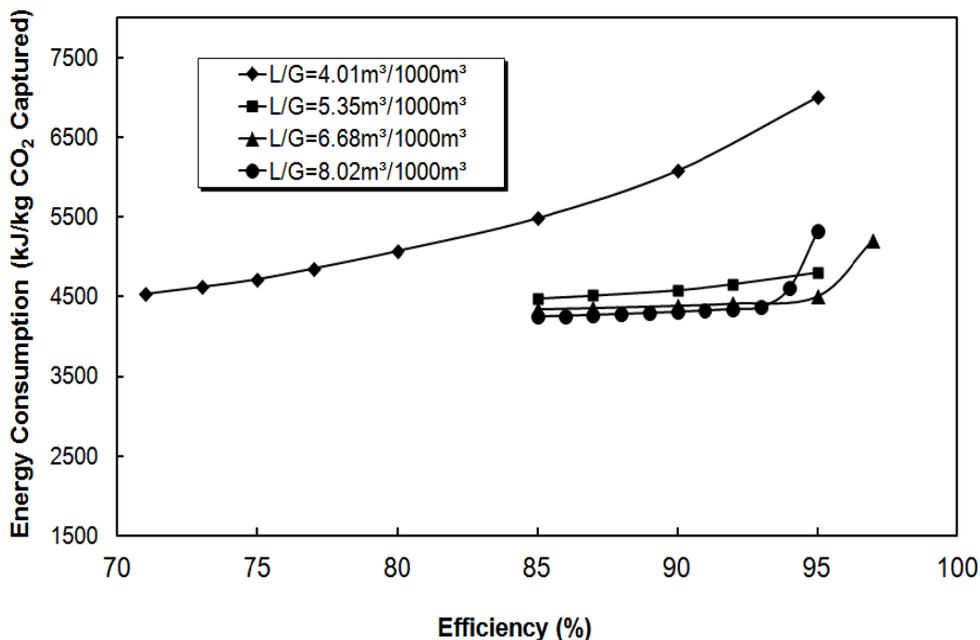
Figure 7. The relationship between specific energy consumption and CO_2 capture efficiency at different L/G for the gas flow rate of 4.72×10^{-3} m^3/s
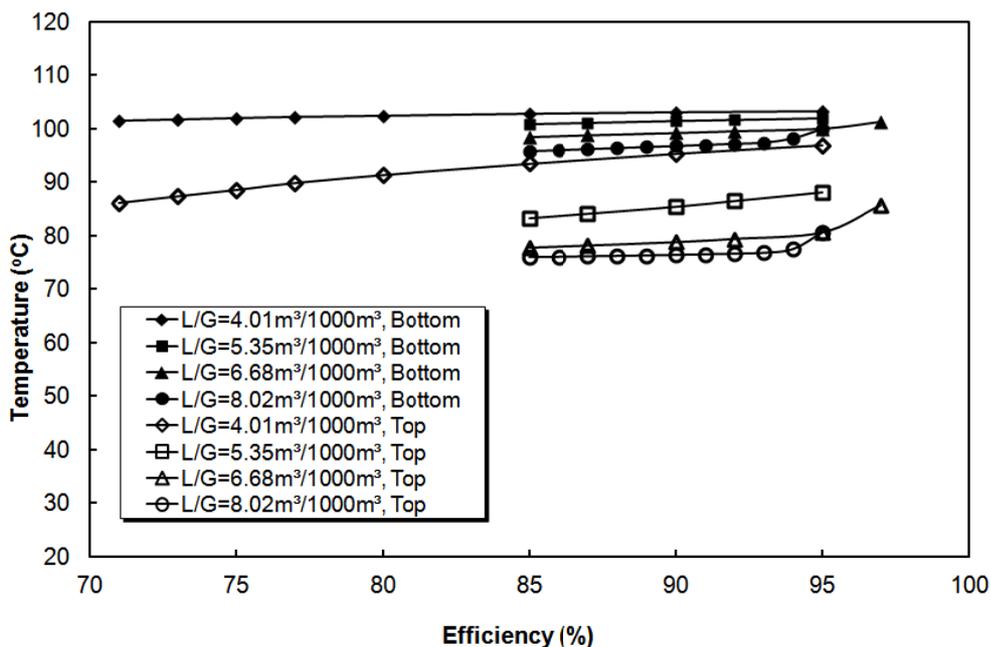
Figure 8. The relationship between top/bottom temperature with CO_2 capture efficiency at different L/G for a gas flow rate of 4.72×10^{-3} m^3/s
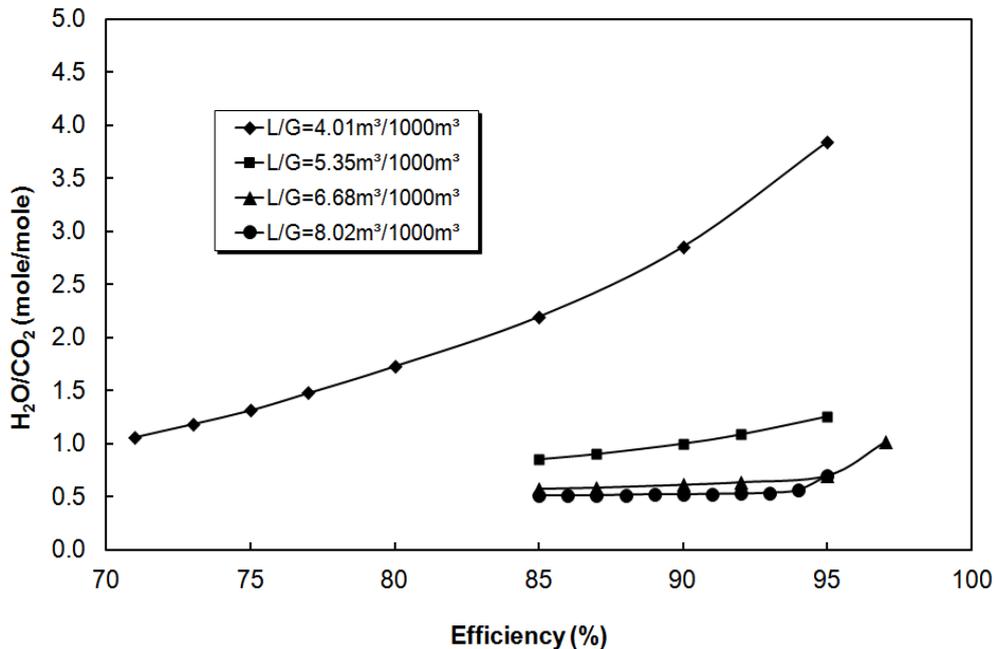
Figure 9. The relationship between H_2O/CO_2 mole ratio and

the CO_2 capture efficiency at different L/G values for a gas flow rate of 4.72×10^{-3} m^3/s
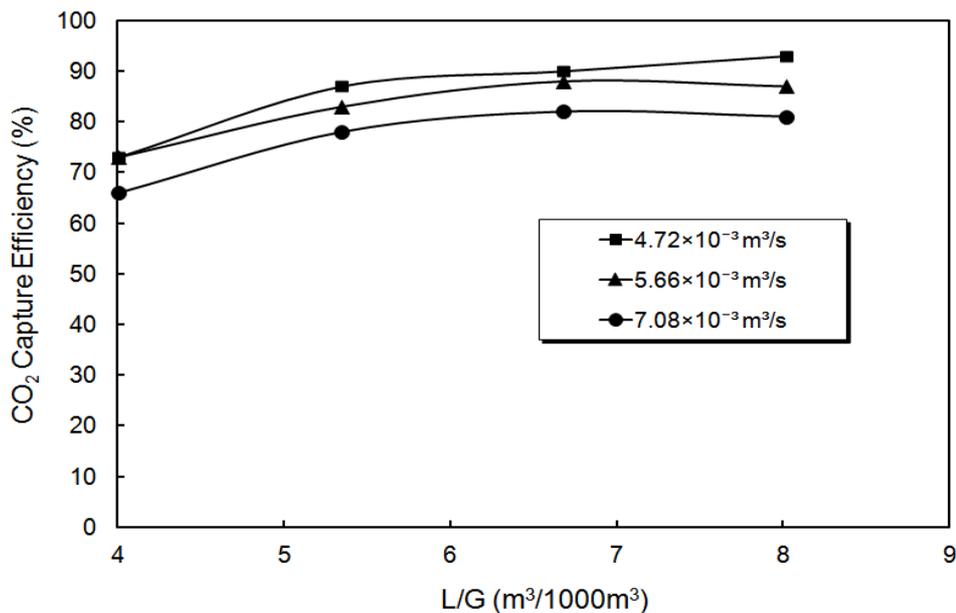
Figure 10. The relationship between CO_2 capture efficiency and L/G at various gas flow rates
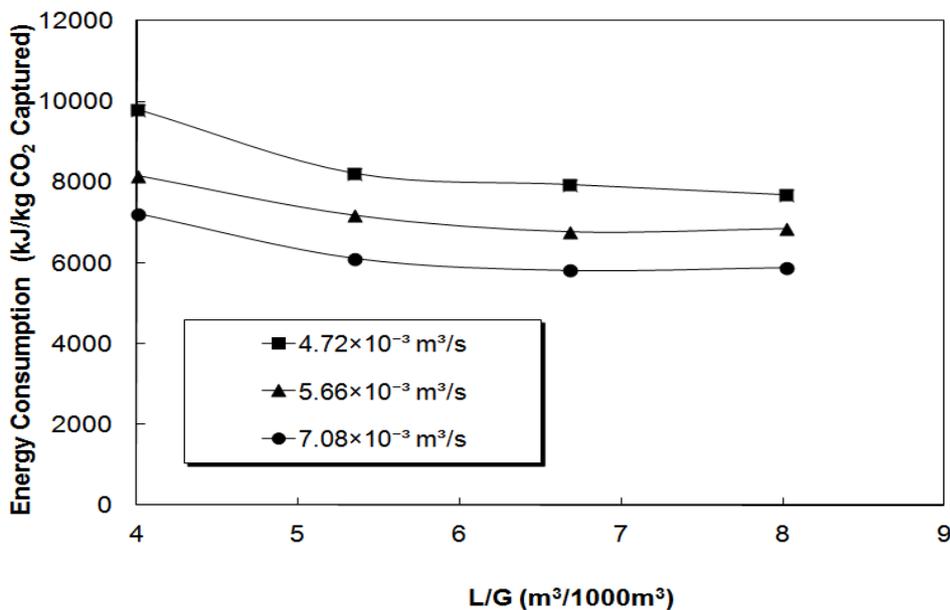
Figure 11. The relationship between the specific energy consumption and L/G at different gas flow rates
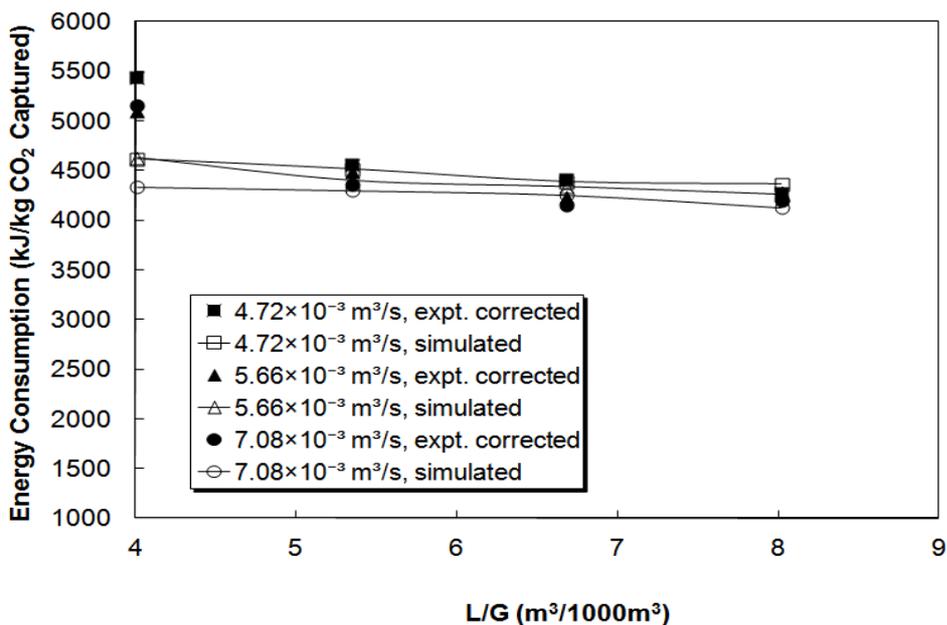
Figure 12. The relationship between specific energy consumption and L/G,
with the modified experimental and simulated data
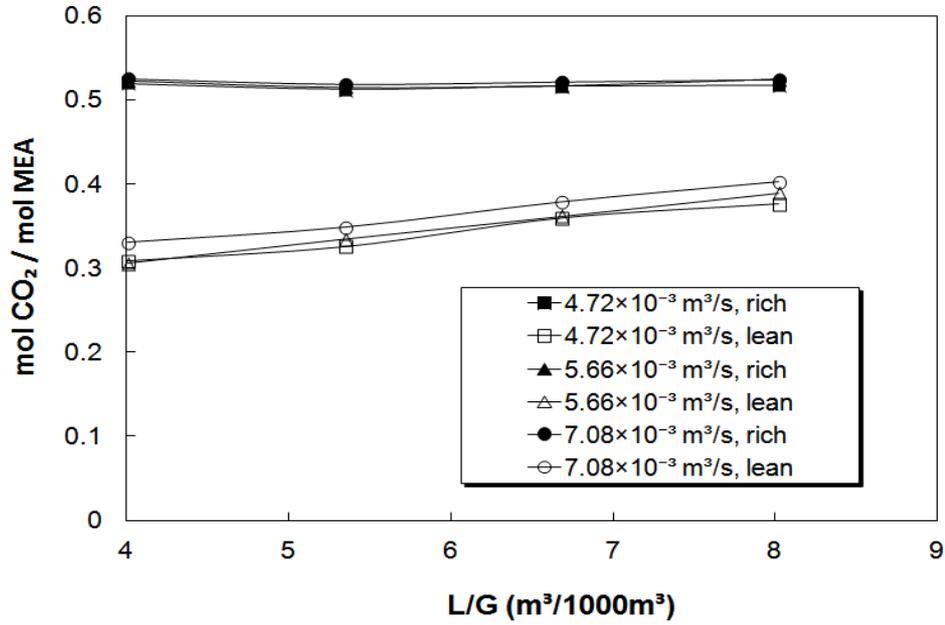
Figure 13. The relationship between L/G and solvent CO_2 rich/lean loading