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Experimental Study of Chemical Absorption of CO2 in a Bench-Scale Spray Dryer Absorber

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Abstract: The removal of CO2 through chemical absorption in a bench-scale spray dryer absorber was investigated experimentally using the lime slurry as absorbent. The effect of important operating parameters on CO2 removal efficiency has been investigated; in selected ranges of operating parameters, increasing gas inlet temperature and absorbent concentration lead to permanent efficiency decline, increasing liquid to gas flow rate ratio and inlet gas humidity and lowering CO2 concentration have favored the removal efficiency. Adding Na (OH) solution to the absorbent increases its ability to absorb CO2 whiles decreases its tendency to produce final dried powder. It was found that adding 100 mL of Na (OH) solution with 1 mol/L concentration per each 1000 mL of lime slurry gives a removal efficiency of 70.5%. Other operating parameters were according to the following: Tg,in = 200 ○C, CA,in = 5 %(vol.), CB,in = 0.7 mol/L, (L/G)in = 0.025 mL/L, Hin=0.024 kg/kg and N=12600 rpm.

Keywords: CO2 Removal; Chemical Absorption; Spray Dryer Absorber; Lime Slurry

1. Introduction

Global warming resulted from the emission of greenhouse gases has received widespread attention. Among the greenhouse gases, CO2 contributes more than 60% to global warming because of its huge emission amount (Albo, Luis, & Irabien, 2010; Herzog, Drake, & Adams, 1997; Rubin, Davison, & Herzog, 2015).

Capture and storage of CO2 produced by fossil fuel combustion is one of the important strategies for reducing or sustaining atmospheric levels of greenhouse gas emissions (Davidson, Freund, & Smith, 2001; El Hadri, Quang, Goetheer, & Zahra, 2017; Oh, Binns, Cho, & Kim, 2016).

Today, there are a variety of technologies capable of capturing CO2 from industrial flue gas streams, like physical and chemical absorption, cryogenic separation, membrane separation, and adsorption. Among them chemical absorption into an aqueous alkaline solution is the most well-established technology available for CO2 capture. This technology has been proven for more than a half century to work successfully in the oil and gas as well as chemical industries (Rinker, Ashour, & Sandall, 2000; Singh et al., 2018; Yeh & Bai, 1999).

Different absorbents have been used in chemical absorption process like ionic liquids, alkanolamines and their blended aqueous solutions. However, the alkanolamine aqueous solutions possess some drawbacks such as high equipment corrosion rate, high energy consumption in regeneration, and a large absorber volume required (Yu, Huang, & Tan, 2012).

One of the most used equipment for chemical absorption of CO2 is packed bed absorber. Typically, the operation pressure in it is around 1.0 bar and the temperature in the absorber is generally in the ranges of 40–60°C. Spray dryer absorber is a gas pollution control device that has been used in power generating stations and waste incinerators to remove the acid gases, heavy metals, mercury, dioxin and other harmful components from the flue gas before this gas is released to the environment (Nguyen & Spink, 1993; Wang, Lawal, Stephenson, Sidders, & Ramshaw, 2011;

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Weiss, Ruhland, & Kind, 1990). The process is based on contacting the hot polluted gas to the fine absorbent droplets in a chamber. The main advantage of this process is that there is no need to additional wastewater treatment facilities since the final residue is solid that is suitable for land fill although other possibilities of reuse like filler and so on can be considered (Feldkamp, Neumann, & Fahlenkamp, 2003; Heebink et al., 2007; Jacobsen, Maimann, Rasmussen, & Fridberg, 2014). Other advantages include no need to use a demister inside the equipment, low level of corrosion occurrence, law capital and maintenance costs and low energy consumption.

In this research to remove CO_2 from a simulated flue gas stream a bench scale spray dryer absorber with lime slurry as absorbent has been used. It should be mentioned that the process of reactive absorption of CO_2 in spray dryer absorber mainly with NaOH and alkanolamines solutions as absorbent, has been investigated by few researchers (Chen, Fang, Tang, & Liu, 2005; Kavoshi, Hatamipour, & Rahimi, 2011, 2013; Kavoshi, Rahimi, & Hatamipour, 2015). In this study the effectiveness of absorbent lime slurry has been tested and the effect of different operating parameters on removal efficiency has been examined.

It should be mentioned that it is possible to use limestone ($CaCO_3$) instead of lime (CaO) but using lime offers better utilization of the reagent, also the operation is more flexible. The major disadvantage is the higher cost of lime compared to limestone.

2. Materials and Methods

The schematic diagram of the setup is illustrated in Figure 1. The main chamber was consisted of a cylindrical part with 97 cm diameter and 102 cm height attached to a conical part with 74 cm height. There were two fans in the setup; the first fan blows the air to the heat exchanger through a 15 cm diameter pipe, the second fan was placed after the cyclone and sent the gas to the environment. The latter fan made suction on whole of the process. CO_2 from the capsule was added to the inlet air, also the vapor from a steam generator part that was considered to increase the inlet gas humidity, was added to the air. Air, CO_2 and water vapor entered the heater that was a shell and tube heat exchanger and natural gas was burned in its shell. When the temperature, CO_2 concentration and humidity of inlet gas were fixed the lime slurry was pumped from the tank with agitator to the atomizer through a peristaltic pump.

The atomizer used in this study was rotary disc atomizer and was placed in the center of dryer roof and 12 cm below it. Disc diameter was 7 cm and there were 8 holes each 0.7 cm diameter each. Changing the rotation speed has changed the droplets diameter released from the edge of the disc.

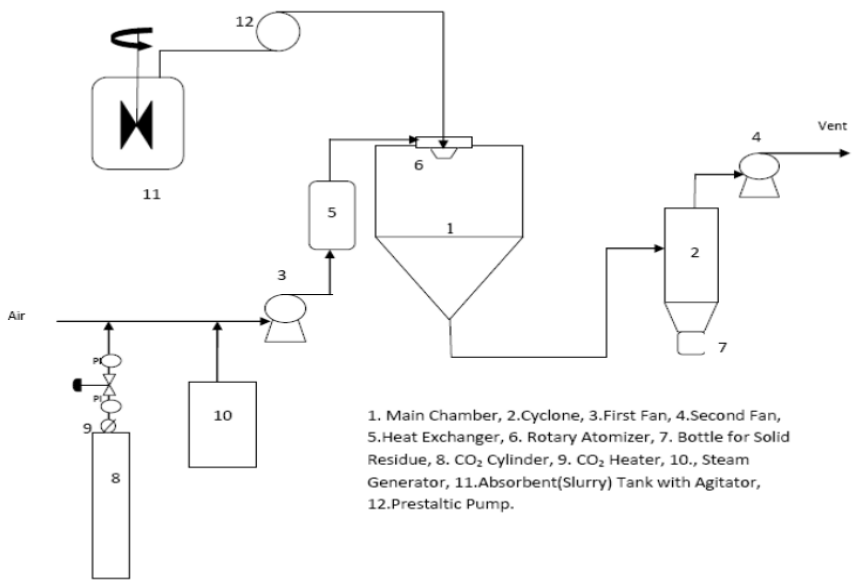

Figure 1. The schematic diagram of the setup

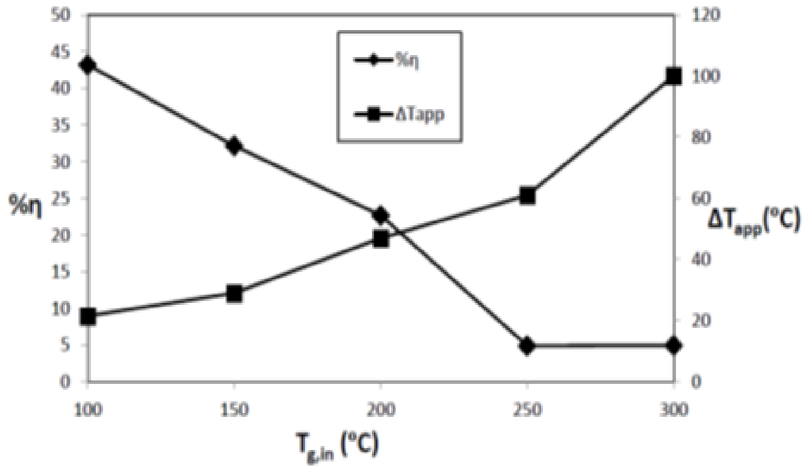

Figure 2. The effect of inlet gas temperature on removal efficiency and approach temperature ($C_{A,in}$=10% vol., $C_{B,in}$=0.7 mol/L, $(L/G)_{in}$=0.02 mL/L, N=21000 rpm, H_{in}=0.006 kg/kg), $S(\eta) = 0.52, S(\Delta T_{app}) = 1.11$

Inlet gas flowed axially through an annulus around the atomizer in the chamber. A 5 cm diameter pipe was attached to the conical part to conduct the gas/solid mixture to the cyclone which separated the solid from the gas. In each experiment inlet gas sampling was done before starting the pump. Humidity of the gas was determined through measuring the dry and wet bulb temperatures of the gas whenever needed. Before releasing the outlet gas to the environment sampling was done to determine the volume percent of CO2 in it. The absorbent, CaO (Merck, Germany), purchased from a chemical trading company in Iran. For preparation of the absorbent slurry with specific concentration, the mass of CaO was determined according to the following reaction and mixed with distilled water.

$$CaO + H_2O \rightarrow Ca(OH)_2 \tag{1}$$

The reaction occurs in this process is as follows:

$$CO_2 + Ca(OH)_2 \rightarrow CaCO_3 + H_2O \tag{2}$$

The operation began by introducing air and CO_2 mixture without any spray and allowing it to heat up to reach the predefined value of inlet gas temperature and CO_2 concentration. Attaining a steady state condition, the feed pump was turned on and the prepared absorbent was pumped and sprayed. A CO_2 analyzer (0–100% in volume, ±2% uncertainty, Guardian Plus- D600, USA) was used to measure the inlet and outlet CO_2 concentration in the gas stream. The uncertainty of the temperature was ±0.1 °C.

Each experiment took about 20-30 min from the injection startup. In order to improve the precision of the results each experiment was repeated two times. Since each experiment was repeated twice with nearly five readings, there were 10 data points among them the average (arithmetic mean) has been reported as the final number. But the standard deviation for each number in the graphs was calculated through the following formulas:

$$S = \sqrt{\frac{\sum_{i=1}^{n}(X_i - \bar{X})^2}{n}} \tag{3}$$

Where S is the standard deviation, $\bar{X}$ is the average, X_i is data points and n is the number of data.

The operating parameters that were changed during the experiments were inlet gas temperature: $T_{g,in}$(100-300°C), absorbent concentration: $C_{B,in}$(0.7-3.9 mol/L), CO_2 concentration: $C_{A,in}$(5-15% (vol.)), slurry flow rate: L_{in}(1-4 mL/s), gas flow rate: G_{in}(40-160 L/s), gas humidity: H_{in} (0.006-0.024 kg/kg) and the speed of disc atomizer rotation: N (4200-21000 rpm).

3. Results and Discussion

To evaluate the overall performance of spray dryer in CO2 removal, the molar based removal efficiency is defined as follows:

$$\%\eta = \frac{\dot{n}_{A,in} - \dot{n}_{A,out}}{\dot{n}_{A,in}} \times 100 \tag{4}$$

Where $\dot{n}_{A,in}$ and $\dot{n}_{A,out}$ are inlet and outlet CO_2 molar flow respectively. Also the approach temperature is defined as follows:

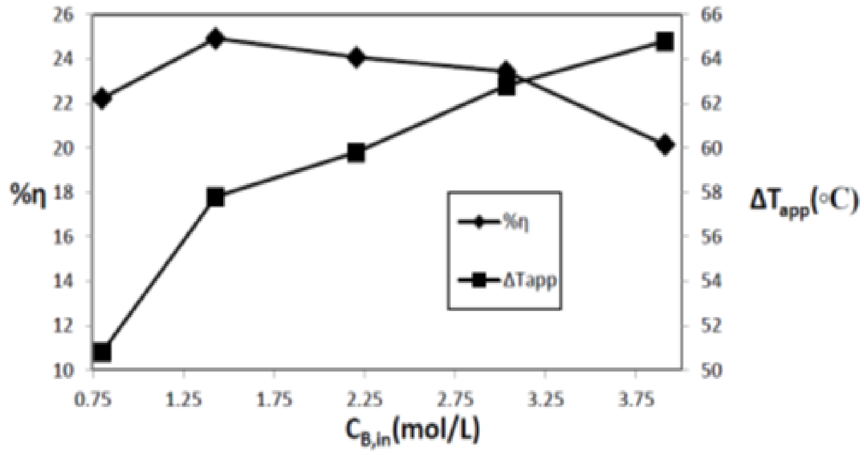

Figure 3. The effect of absorbent concentration on removal efficiency and approach temperature ($T_{g,in}$=200 °C, $C_{A,in}$=10% vol., $(L/G)_{in}$=0.025 mL/L, N=21000 rpm, H_{in}=0.008 kg/kg), $S(\eta) = 0.41, S(\Delta T_{app}) = 1.11$.

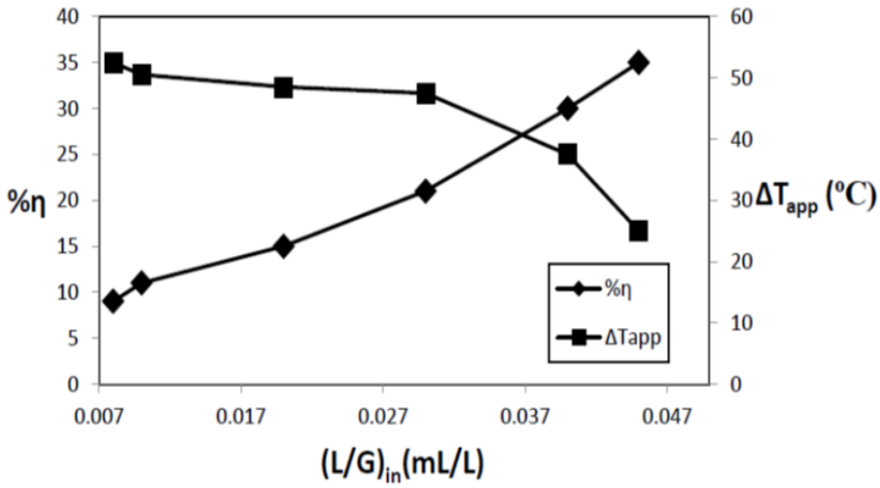

Figure 4. The effect of (L/G)in on the removal efficiency and approach temperature (Tg,in=200 C, CA,in=10% vol., CB,in=0.7 mol/ L, Hin=0.009 kg/kg, N=21000 rpm), S(η)=0.45,S(ΔT _app)=0.49.

$$\Delta T_{app} = T_{g,out} - T_{wb,in} \quad (5)$$

Where $T_{g,out}$ represents the outlet gas temperature and $T_{wb,in}$ is wet bulb temperature of inlet gas.

3.1. Effect of Inlet Gas Temperature

In Figure 2 the effect of increasing inlet gas temperature on overall removal efficiency and approach temperature is depicted. As can be seen increasing inlet gas temperature decreases the removal efficiency and increases the approach temperature continuously. The reason can be ascribed to domination the drying as the inlet gas temperature is increased. Since lime slurry has a high tendency to lose the water content, the droplets start to be dried and the solid crust formation occurs instantaneously when the droplets release to the chamber. Also it can be seen after 250°C the removal efficiency decrease is stopped. It can be said up to 250°Cincreasing inlet gas temperature accelerates the solid crust formation but after that, the temperature increase has no more effect since the droplets have been completely dried and the reaction doesn't proceed.

3.2. Effect of Absorbent Concentration

In Figure 3 the effect of absorbent concentration increase on removal efficiency and approach temperature is shown.

Although it is expected that increasing absorbent concentration leads to improve the removal efficiency but a different manner has been observed. This can be due to the domination of drying on the reaction; for lime slurry, increasing the absorbent concentration strengthen the tendency to drying. So as can be seen after a small increase in removal efficiency it begins to decrease gradually because of decrease in water content and tendency to drying.

Also approach temperature is increased with absorbent concentration increase, because decreasing water content of the droplets in high absorbent concentration cause to less heat transferred to the droplets and so outlet gas temperature is higher.

3.3. Effect of CO_2 Concentration

In all of the experiments done to investigate the effect of CO2 concentration on the removal efficiency it was observed that increasing CO2 concentration leads to decrease the removal efficiency. This effect can be interpreted as follows: decreasing the removal efficiency happens because of low flow rate of liquid to gas maintained to obtain the dry powder. The low $(L/G)_{in}$ leads to low molar ratio of absorbent to CO2 less than the stoichiometric ratio. By increasing CO2 concentration, the reaction accelerates but since there is not enough absorbent in the system to react, so the denominator in Eq.4 increases while the numerator is nearly constant and the removal efficiency decreases. Experimental results show that CO2 concentration doesn't have a considerable effect on the approach temperature.

3.4. Effect of Liquid to Gas Flow Rate Ratio

In several experiments the effect of changing $(L/G)_{in}$ on process performance was investigated. In some Lin was changed that reveals the high dependence of the removal efficiency of the system on water presence since increasing Lin drastically causes to increase the removal efficiency. In some of the experiments Gin was changed that reveals the higher efficiency in lower gas flow rates that can be because of increasing the gas and droplet residence time. Also it was observed that with increasing the gas flow rate, the approach temperature is decreased. This is due to saturation of gas with humidity and decreasing the evaporation rate that leads to higher outlet gas temperature. It should be mentioned that in lower gas flow rates the powder moisture is considerable so except in the experiments related to Figure 4 always the gas flow rate was set to maximum possible value. In this figure changes of removal efficiency and approach temperature with $(L/G)_{in}$ is shown.

3.5. Effect of Droplet Diameter

Since CO_2 removal occurs through reaction on droplet surface so it is expected that the process efficiency may depend on droplet surface area i.e. the higher surface area will provide the more place for reaction to take place. On the other hand, when a rotary atomizer is used to atomize the liquid, the droplets diameter and surface area, depend on disc rotation speed; the higher speeds produce finer droplets.

In the experiments it was observed that the removal efficiency is a function of disc rotation speed but there was a special speed (between the highest and the lowest speeds) that corresponded to the maximum removal efficiency (see Figure 5).

This can be interpreted with respect to available surface area and humidity in the process. In high rotation speeds that produce lower diameter and large surface area two opposite effects exist; large surface are causes the absorption rate to be increased, in the other hand water evaporation also be increased for finer droplets. In this process since the slurry droplets intensively tend to dry and also the reactivity of these droplets is highly depend on the presence of water in the system, so with increasing the evaporation rate, reaction progress considerably be decreased. It seems that in very low droplet diameter, water evaporation intensifies. Also the additional resistance of solid crust decreases the reaction rate. In low rotation speeds that produce coarser diameters interface area of gas and droplets and so the reaction rate is decreased.

3.6. Effect of Inlet Gas Humidity

The effect of inlet gas humidity changes on overall process performance is shown in Figure 6. Increasing gas humidity, improve the removal efficiency considerably. High gas humidity leads efficiency improvement in two ways; first solid crust formation postponement due to lower evaporation and second higher reaction rate due to higher gas temperature. It should be mentioned that in high gas humidity solid powder moisture is considerable. Since a typical flue gas has a humidity of about 0.14 kg/kg and maximum inlet gas humidity used here was 0.024 kg/kg, it can be supposed that in the case of using a flue gas the removal efficiency improved.

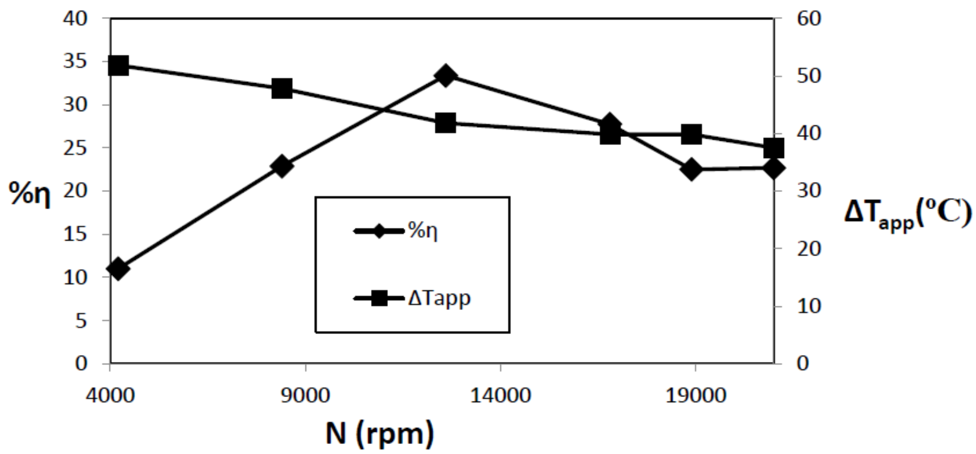

Figure 5. The effect of disc rotating speed on removal efficiency and approach temperature in bench-scale spray dryer, other operating conditions: $T_{g,in}$=200 °C, $C_{A,in}$=10% vol., $C_{B,in}$=0.7 mol/L, $(L/G)_{in}$=0.025 mL/L, H_{in}=0.008 kg/kg, $S(\eta) = 0.60, S(\Delta T_{app}) = 1.40$.

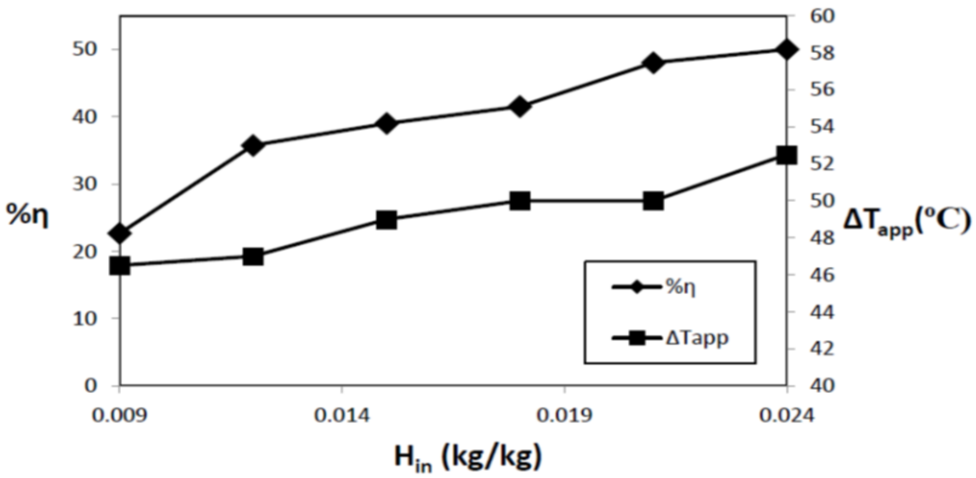

Figure 6. The effect of inlet gas humidity on removal efficiency and approach temperature, other operating conditions: $T_{g,in}$=200 °C, $C_{A,in}$=10% vol., $C_{B,in}$=0.7 mol/L, $(L/G)_{in}$=0.025 mL/L, N=21000 rpm, $S(\eta) = 0.36, S(\Delta T_{app}) = 1.63$.

3.7. Improving the process performance

With the aim of improving the process performance and abating the sever clogging of paths and intensive deposit sticking on the walls of the spray dryer that is very prevalent when using lime slurry, adding Na(OH) solution to the absorbent was examined. It should be mentioned that adding Na (OH) solution noticeably heighten the absorption capacity of the slurry but because of the higher price of Na (OH) than lime it should be used in little amount beside the fact that adding Na (OH) to lime slurry decreases the tendency of the droplets to drying so adding the alkalinity of slurry by Na (OH) so much leads to an undesirable wet final powder. Also the use of Na (OH) solution decreases clogging of paths.

In Figure 7 the results of one of the experiments has been shown; the changes of the removal efficiency against gas inlet temperature for three absorbents (Na (OH), $Ca(OH)_2$ and the mixture of them) has been depicted. It is clear that adding Na (OH) to the lime slurry improves the absorbent performance noticeably.

Through several experiments it was found that for this setup adding 100 mL of Na (OH) solution with 1 mol/L concentration per each 1000 mL of lime slurry gives a removal efficiency of 70.5%. Other operating parameters were according to the following: $T_{g,in}$ = 200 ○C, $C_{A,in}$ = 5 %(vol.), $C_{B,in}$ = 0.7 mol/L, $(L/G)_{in}$ = 0.025 mL/L, H_{in}=0.024 kg/kg and N=12600 rpm.

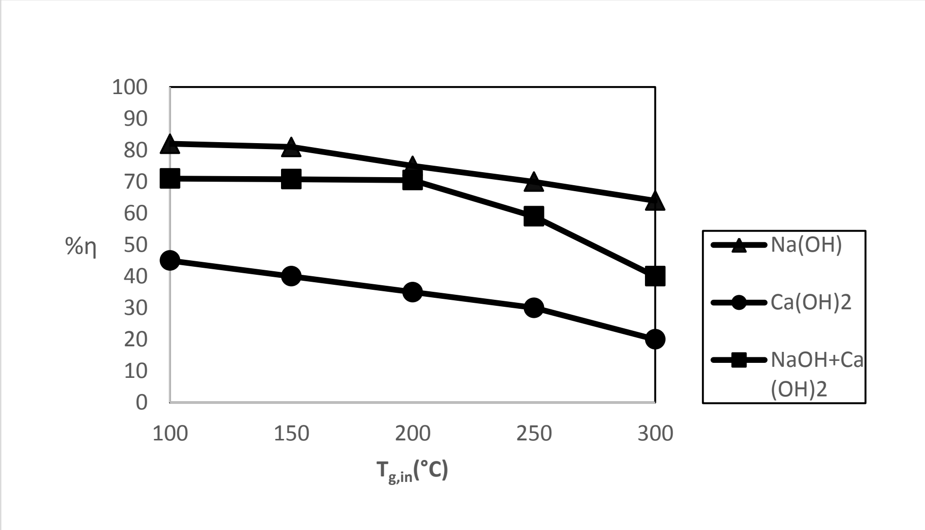

Figure 7. The effect of inlet gas temperature on removal efficiency for three absorbents ($C_{Na(OH)}$=1 mol/l, $C_{Ca(OH)2}$=0.7 mol/l), other operating conditions: $C_{A,in}$=5% vol., $(L/G)_{in}$=0.025 mL/L,H_{in}=0.024 kg/kg, N=12600 rpm, $S(\eta)$ = 0.29.

4. Conclusion

In this paper the effect of operating parameter on process performance was evaluated and through experimental observations it was found that generally two main phenomena happen in the process of chemical absorption of CO2 in spray dryer absorber; chemical reaction and drying. Accelerating the drying leads to decline the chemical reaction and postponing the drying causes the reaction proceeds to more extents.

To increase the removal efficiency, Na (OH) solution was added to the absorbent; an appropriate Na (OH) concentration has been determined. Also other operating parameters have been set such that maximum removal efficiency obtained in selected ranges.

Nomenclature

C_B, mol/L	Absorbent concentration
C_A, %vol	CO2 concentration gas stream
L, mL/s	Flow rate of slurry stream
G, L/s	Flow rate of gas stream
Tg,◦C	Gas temperature
H, kg/kg	Gas humidity
$\dot{n}_A$, mol/s	Molar flow of CO2
N, rpm	Speed of rotation for atomizer
$S(\eta)$	Mean Standard Deviation for Removal Efficiency
$S(\Delta T_{app})$	Mean Standard Deviation for Apparent Temperature
Greek letters	
η	Removal efficiency
Δ	Difference
Subscripts	
in	Inlet
out	Outlet
app	Apparent
Wb	Wet bulb

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