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Scrubbing of HCl Gas from Synthesis Gas in a Multistage Dual-Flow Sieve Plate Wet Scrubber by Alkaline Solution

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Abstract

The synthesis gas leaving from the coal gasification process is having particulate matter and other gaseous pollutants majorly hydrogen sulphide and hydrogen chloride (HCl). Wet scrubbers can simultaneously remove the gaseous pollutants and dust particles from effluent gas streams. Dual-flow sieve plate column wet scrubbers can be used for the removal of acid gases from the exhaust gases. A three stage lab scale dual-flow sieve plate scrubber has been designed, fabricated and installed. Experiments were conducted to remove the HCl from the synthesis gas at different gas-liquid ratios within the gas flow rates (Q_G) 8.297 × 10^{-4} Nm^3/s, 13.829 × 10^{-4} Nm^3/s and 19.36 × 10^{-4} Nm^3/s, liquid flow rates (Q_L) 20.649 × 10^{-6} m^3/s, 34.415 × 10^{-6} m^3/s and 48.183 × 10^{-6} m^3/s and at different inlet HCl concentrations ($C_{HCl,i}$) 100 ppm, 200 ppm and 300 ppm range using sodium hydroxide solution as scrubbing liquid. The maximum percentage removal of HCl ($\%\eta_{HCl}$) 93.98% are observed at $Q_G = 8.297 \times 10^{-4}$ Nm^3/s, $Q_L = 48.183 \times 10^{-6}$ m^3/s and $C_{HCl,i} = 300$ ppm with 0.005 $kmol/m^3$ NaOH solution. A semi empirical model has been developed to predict the efficiency of the HCl scrubbing using the experimental results and the variables which show the impact on the scrubber performance. Almost 80% of the experimental results are matched well with the predicted values with less than ± 15% deviation.

Keywords: Air pollution; Control; Efficiency; Scrubbing; Model; Removal; Sodium hydroxide

Introduction

Coal is a combustible substance composed of carbon along with several impurities like sulphur, chlorine, and nitrogen and metal compounds. These impurities get converted to gaseous species such as H_2S, HCl and NH_3 when this coal is processed. Therefore, removal of these impurities is essential to protect the energy production equipment and to meet environmental standards before entering into the atmosphere. During the gasification process, the chlorine present in the coal is volatilized to HCl or metal chlorides. The chloride content is high in US coals with 0.01% to 0.5% and during coal gasification process these chlorides will produce HCl [1]. In the gasification process, the chlorine content of coal volatilizes to metal chlorides or HCl. But major portion of chlorine is converted into HCl. Reports show that there is a wide variation in the HCl concentrations in synthesis gas streams. Bakker and Perkins [2] reported that the HCl concentration is five times higher in coal gas than in coal-fired boiler combustion gas stream. The chlorine content of coal, the gasification temperature and the type of gasifier are the factors on which the HCl vapour concentration depends and the most reliable data was found from the fixed-bed gasifier operation at General Electric Corporate Research and Development at Schenectady, New York. At that facility, about 300 ppmv HCl vapour concentration was obtained using coal with 0.24 wt% chlorides [3]. The concentrations of HCl are reported as high as 500 ppm in coal syngas from gasifier effluents [4,5]. Perkins et al. [6] reported that the chloride deposited on syngas coolers accelerate the corrosion of the heat exchanger material and HCl reacts with the deposited scum of slag, forming low-melting iron chlorides, thereby expediting the corrosion rate.

HCl gas destroys the metal and lime stone building structures due to its highly corrosive nature. HCl is harmful to aquatic organisms when it is highly dissolved in water and many times it kills them. The emitted HCl into the environment is quickly washed out by moisture in the air and rain. If the HCl levels in these acid rains are above the exposure limits then the lakes and soils will be affected [7]. Exposure to high concentrations of HCl emissions leads to various health issues such as skin colour change, rapid breathing and fluid accumulation in the lungs. HCl gas ingestion seriously destroys the gastrointestinal tract and in extreme cases it causes to death also.

Frank and Nancy [8] have reported that the wet scrubbers are effective air pollution control devices which can simultaneously remove both gaseous pollutants and dust particles. Wet scrubber removes particles by capturing them in liquid droplets, and they remove gases by dissolving or absorbing them into the liquid [9]. The gaseous pollutants will be removed in the wet scrubbers by absorbing or dissolving them into the scrubbing liquid and for HCl gas removal this scrubbing liquid is normally water or a weak alkaline solution. In many cases, weak basic solutions give higher HCl gas removal efficiencies than ordinary water. Furthermore, the efficiencies of dry scrubbing methods are relatively low compared with wet scrubbing methods.

There are different types of scrubbers reported in the literature to choose the suitable scrubber for particulate and gaseous pollutant removal with higher removal efficiencies [10-14]. Rajmohan et al. [10] have reported almost 99.99% SO_2 removal efficiency of twin fluid air-assist atomized spray column. Meikap et al. [11] have reported the water scrubbing and alkali (NaOH) scrubbing of SO_2 gas using small droplets in a horizontal co-current flow ejector system with 98.62% and 100% removal efficiencies respectively. The SO_2 gas scrubbing has also been studied with greater removal efficiencies in a modified multistage bubble column scrubber without using any additives or pre-treatment [12]. Bandopadhyay et al. [13] have achieved almost 100% removal efficiencies with alkali scrubbing of SO_2 gas in a Tapered

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Bubble Column Scrubber. Bangwoo Han et al. [14] have observed the HCl removal in a packed bed scrubber for four different packing at various liquid-gas ratios. There is very less literature found for gas absorption in multistage dual-flow sieve plate columns. The most commonly used perforated tray column without a down comer is one of the dual-flow tray columns, which is used for its simple design and its ease of installation and maintenance. In many applications the dual-flow trays were used where there is no need of wide range of operating conditions. In the operating range, with the less capital investment it gives high mass transfer efficiency. The alternating counter-current passage of gas and liquid through the holes provides a self-cleaning of the device in fouling systems [15,16]. This paper presents the work done for the removal of HCl gas from an effluent gas stream with a three stage dual-flow sieve plate column wet scrubber and the effect of different operating variables on HCl removal efficiency of scrubber.

Materials and Methods

Materials

Hydrochloric acid solution (35%), sodium hydroxide pellets of 97% purity, oxalic acid dehydrate of 99% purity were supplied by Merck Specialities Pvt. Ltd. and used without further purification. Aqueous solution of hydrochloric acid, dilute oxalic acid solution (0.025 k mol/m^3) and dilute solutions of sodium hydroxide (0.00125 k mol/m^3, 0.0025 k mol/m^3, 0.00375 k mol/m^3, 0.005 k mol/m^3 and 0.05 k mol/m^3) have been prepared using distilled water. Dilute Hydrochloric acid solution was prepared by dissolving 35% pure concentrated hydrochloric acid in distilled water (0.6 k mol/m^3 - 2 k mol/m^3).

Methods

The schematic representation of the experimental set-up is shown in Figure 1. A three stage dual-flow sieve plate column is made up of a transparent, vertical Perspex glass with 2.6 m length and 0.1524 m ID and fitted with an outlet at top in fructo-conical shape. The air contaminated with HCl, enters through the inlet of the column. The inlet is fitted with a diffuser on the inner side of the column to facilitate the flow of the gas in a uniform manner throughout the column. The plates and diffuser consist of 396 holes of 3 × 10^{-3} m size. The experimental air for the operation is supplied by an air compressor which was prepared by sucking the ambient air. The outlet of the air compressor is fitted

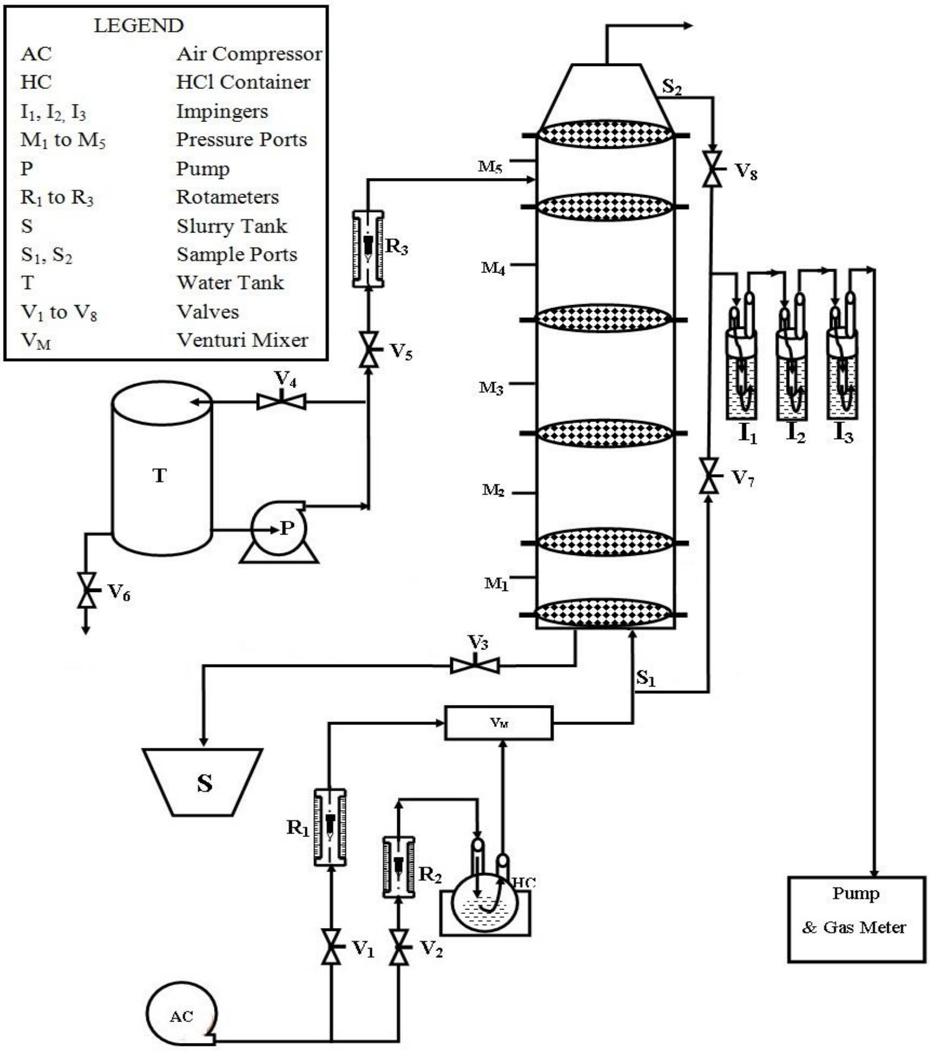

Figure 1: Schematic diagram of experimental set up.

to an air rotameter of range 0-27.65 × 10^{-4} Nm^3/s. The outlet of the rotameter is connected to the inlet of the column through a venturi mixer. Liquid HCl of known concentration is placed in a 2-neck round bottom flask of one litre capacity. Part of the experimental air was entered into the flask from one end and bubbled into the concentrated liquid HCl to form the HCl vapours. The generated HCl vapours were then allowed to escape through the other end to enter into the venturi mixer where they were mixed with the other part of the experimental air and enter to the column at desired concentrations. The concentrations of HCl are maintained at desired values with less than ± 5% deviations. A tank of 20 × 10^{-3} m^3 capacity is used as liquid reservoir from which the scrubbing liquid is pumped out by a 0.5 HP centrifugal pump to the column at the top where a liquid distributor plate disperses the liquid to the tray below in a shower manner. The flow rate of the scrubbing liquid is controlled through a rotameter of range 0-68.83 × 10^{-6} m^3/s.

The experimental conditions can be seen in Table 1. The experiments were conducted at different gas to liquid ratios at different inlet HCl concentrations. The samples were collected at S_1 and S_2 which are sample ports at inlet and outlet respectively. For iso-kinetic sampling, 60 × 10^{-6} m^3 of 0.05 k mol/m^3 NaOH solutions was placed in the three impingers in series in which the first impinger inlet was connected to the sample port and the third impinger outlet was connected to vacuum pump and gas meter. As the experiment proceeded, the HCl vapours were passed through the impingers and got absorbed into the NaOH solution. The NaOH reacts with the HCl in the contaminated air, thereby, neutralizing it. Treated air is then discharged into the atmosphere. As the reaction proceeds, the concentrations of NaOH solution in the impingers get reduced and they were estimated by titrating against Oxalic Acid. The amount of HCl absorbed is evaluated by back calculation and the concentrations of HCl at inlet ($C_{HCl,\ inlet}$) and outlet ($C_{HCl,\ outlet}$) were calculated. The percentage removal of HCl ($\%\eta_{HCl}$) of the column is calculated from the equation (1).

$$\%\eta_{HCl} = \frac{C_{HCl,inlet} - C_{HCl,outlet}}{C_{HCL,inlet}} \times 100 \qquad (1)$$

Results and Discussion

The experimental study was conducted to observe the removal of HCl in three stage dual-flow sieve plate column wet scrubber with water and different concentrations of NaOH solutions as scrubbing liquids. Effects of various parameters such as flow rate of gas, flow rate of liquid, inlet concentration of HCl and concentration of NaOH solution on percentage removal of HCl are keenly observed. Experimentation has been done at various liquid rates of 20.649 × 10^{-6} m^3/s, 34.415 × 10^{-6} m^3/s and 48.183 × 10^{-6} m^3/s and gas rates of 8.297 × 10^{-4} Nm^3/s, 13.829 × 10^{-4} Nm^3/s and 19.36 × 10^{-4} Nm^3/s at different inlet HCl concentrations 100 ppm, 200 ppm and 300 ppm. Water and different concentrations of NaOH solutions of 0.00125 k mol/m^3, 0.0025 k mol/m^3, 0.00375 k mol/m^3 and 0.005 k mol/m^3 are used as scrubbing liquids. HCl removal efficiency percentage has been estimated using equation (1).

Parameters	Values
Temperature	300 ± 1 K
Pressure	1 atm
Liquid flow rate	20.649 × 10^{-6}, 34.145 × 10^{-6} and 48.183 × 10^{-6} m^3/s
Gas flow rates	8.297 × 10^{-4}, 13.829 × 10^{-4} and 19.36 × 10^{-4} Nm^3/s
Scrubbing liquids	Tap Water and NaOH Solutions of different concentrations
NaOH concentration	0.00125, 0.0025, 0.00375 and 0.005 kmol/m^3
Inlet HCl concentrations	100, 200 and 300 ppm

Table 1: Operating conditions for the Experimental Study of HCl removal in Multistage Dual-flow Sieve Plate Column.

Effect of liquid flow rate and HCl loading on the percentage removal of HCl

The scrubbing liquid flow rate effect on HCl percentage removal has been shown in Figures 2-4, for 0.005 kmol/m^3 NaOH solution and for different constant gas flow rates at 100 ppm, 200 ppm and 300 ppm inlet HCl concentrations respectively. It is observed from the results that the HCl percentage removal increased with the increase in liquid flow rate. As the liquid flow rate increases the availability of mass transfer surface increases which increases the interfacial contact between the liquid and gas [17] and the high reactive nature of NaOH reacts with HCl vapours as soon as they are absorbed into the liquid phase NaOH (scrubbing liquid). Due to this fact, the HCl removal efficiency of the dual-flow sieve plate scrubber increases with increasing the flow rate of liquid. The mechanism involves in the dual-flow sieve plate column is that the liquid and gas flows through the holes in counter current manner alternatively. There are two zones in each stage namely froth and spray. In the froth zone, the up flowing gas passes through the thin liquid layer in small bubbles form and in the spray zone the down flowing liquid from the above tray flows in streams and breaks into droplets. Many times the mass transfer occurs in both the zones significantly [15]. It is also observed that the efficiency increased with the increase in concentration due to the fact that the increased concentration increases the gradient of concentration between the gas phase and liquid phase HCl concentrations which directly improves the driving force for absorption of HCl into the film of scrubbing liquid on the tray and the liquid droplets coming from the above tray. Similar trends of results are obtained and observed with the NaOH solutions of 0.00125 k mol/m^3, 0.0025 k mol/m^3 and 0.00375 k mol/m^3 concentration.

Effect of gas flow rate and HCl loading on the percentage removal of HCl

From the Figures 5-7, the effect of gas flow rate on HCl percentage

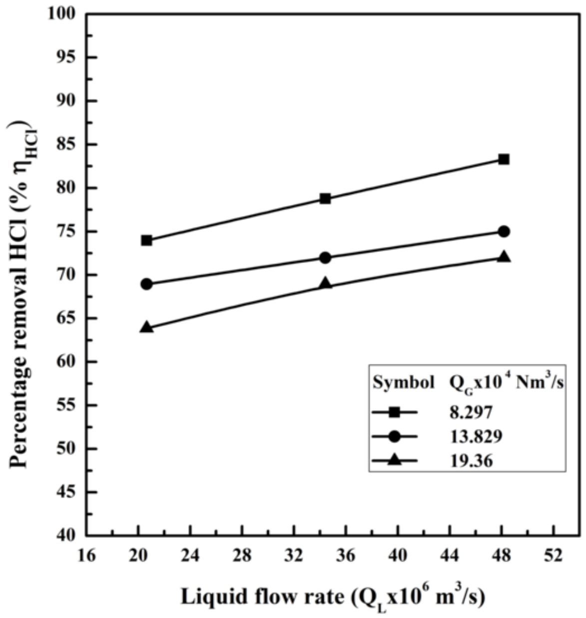

Figure 2: Effect of liquid flow rate on the η_{HCl} for HCl scrubbing with 0.005 kmol/m^3 NaOH solution at different constant gas flow rates and $C_{HCl,i}$=100 ppm.

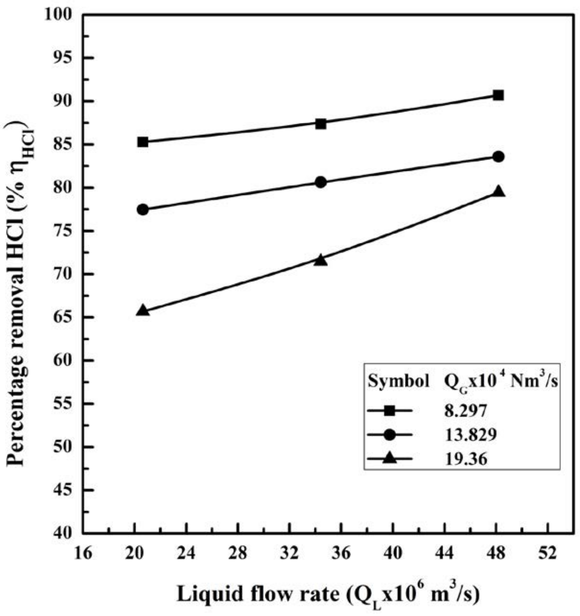

Figure 3: Effect of liquid flow rate on the ηHCl for HCl scrubbing with 0.005 kmol/m3 NaOH solution at different constant gas flow rates and CHCl,i=200 ppm.

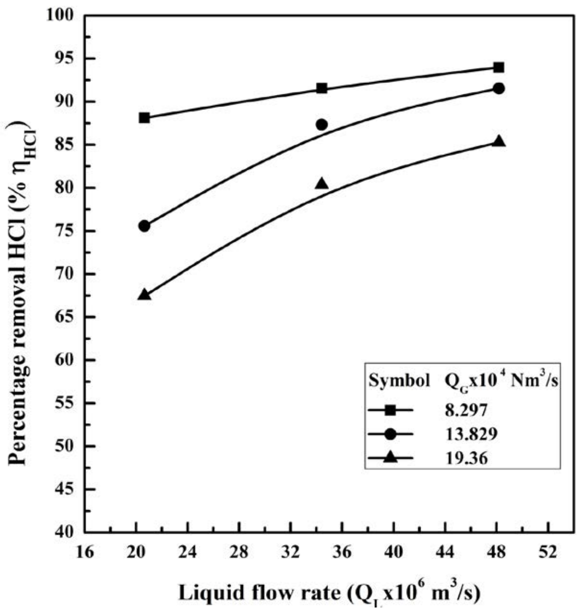

Figure 4: Effect of liquid flow rate on the η_{HCl} for HCl scrubbing with 0.005 kmol/m^3 NaOH solution at different constant gas flow rates and $C_{HCl,i}$=300 ppm.

removal can be seen for 0.005 kmol/m^3 NaOH solutions and for different constant liquid flow rates at 100 ppm, 200 and 300 ppm inlet HCl concentrations respectively. The results show that the increase in gas flow rate decreased the HCl percentage removal. This is due to the fact that the increase in gas flow rate decreases the residence time of gas phase in the column which further decreases the contact time between the down flowing liquid droplets and the up flowing gas in each stage and increases the bubble rise velocity through the scrubbing liquid layer, therefore the gas phase HCl absorption into the liquid phase decreases, which directly decreases the HCl percentage removal. The percentage removal of HCl is increased with the inlet HCl concentrations.

Effect of NaOH concentration on the percentage removal of HCl

It can be seen in the Figure 8 that the effect of NaOH solution

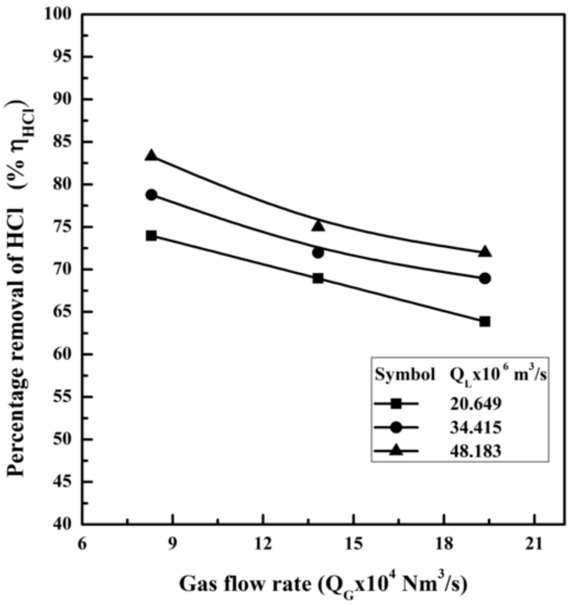

Figure 5: Effect of gas flow rate on the η_{HCl} for HCl scrubbing with 0.005 kmol/m^3 NaOH solution at different constant liquid flow rates and $C_{HCl,i}$=100 ppm.

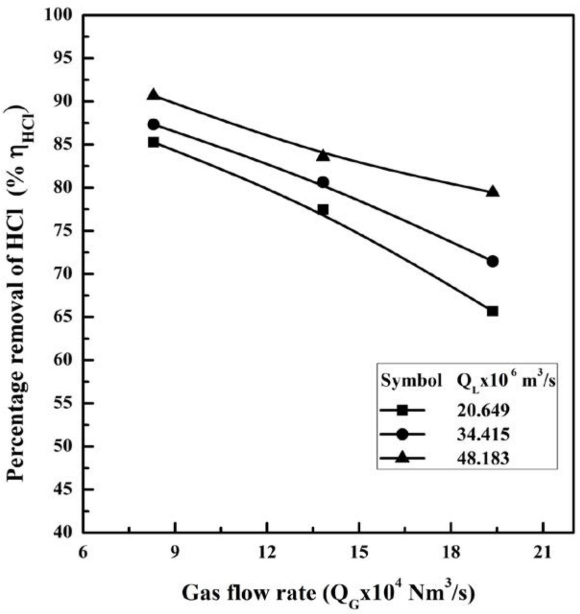

Figure 6: Effect of gas flow rate on the η_{HCl} for HCl scrubbing with 0.005 kmol/m^3 NaOH solution at different constant liquid flow rates and $C_{HCl,i}$=200 ppt.

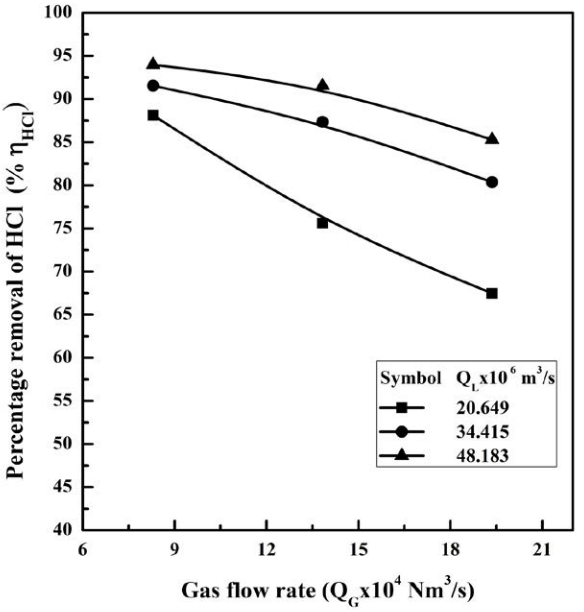

Figure 7: Effect of gas flow rate on the η_{HCl} for HCl scrubbing with 0.005 kmol/m^3 NaOH solution at different constant liquid flow rates and $C_{HCl,i}$=300 ppm.

concentration (C_{NaOH}) on percentage removal of HCl at different inlet concentrations of HCl and at 8.297 × 10^{-4} Nm^3/s gas flow rate, 48.183 × 10^{-6} m^3/s liquid flow rate and it can also be observed that the HCl percentage removal efficiency increased with the increase in concentration of NaOH solution. It due to the fact that as the concentration of NaOH increases the absorbed HCl into the liquid reacts as soon as it enters and directly enhances the mass transfer into the liquid phase from the gas phase. Therefore as the NaOH concentration increased the percentage removal of HCl in the multistage dual-flow sieve plate column scrubber increased.

Correlation development for removal efficiency prediction for water and NaOH scrubbing of HCl

The liquid flow rate and gas flow rate effect on HCl percentage removal for 100 ppm, 200 ppm and 300 ppm constant inlet HCl concentrations at 8.297 × 10^{-4} Nm^3/s gas flow rate and 48.183 × 10^{-6} m^3/s liquid flow rate can be seen in Figures 9 and 10 respectively. From the experimental data, Buckingham π theorem and choosing physical, geometric and flow parameters an empirical equation has been developed to predict the removal efficiency of HCl. The parameters which may influence the HCl removal efficiency (η_{HCl-H_2O}) are:

(a) Geometrical parameters – scrubber height H (m), diameter of the column D_C (m) and sieve tray hole diameter d_h (m), (b) Flow parameters – velocity of gas V_G (m/s) and velocity of liquid V_L (m/s), (c) Physical properties – HCl inlet concentration C_{HCl} (kg/m^3), density of liquid ρ_L (kg/m^3), density of gas ρ_G (kg/m^3), viscosity of gas μ_G (kg/ms), viscosity of liquid μ_L (kg/ms), diffusion coefficient of HCl in water D_{HCl}, (m^2/s) and gravitational acceleration g (m/s^2). The inlet concentration of HCl is expressed in kg/m^3 for dimensional analysis purpose.

Therefore, the removal efficiency of HCl becomes function of twelve parameters. If the relation exists theoretically between the HCl removal efficiency and the above system parameters then may be η_{HCl} written in the below form:

$$\eta_{HCl-H_2O} = f\left(d_h, D_c, H, V_G, \rho_G, V_L, \rho L, \mu_G, \mu_L, D, C_{HCl}, g\right) \quad (2)$$

Estimation of correlation coefficients of dimensional groups was done with multiple linear regression analysis. Nearly the best fitted correlation with 0.1182 root-mean-square error (rmse) and 0.913 coefficient of regression. The obtained model with the estimated coefficients is given in equation (6).

$$\eta_{HCl-H_2O} = k_0\left[\frac{D_C V_G \rho_G}{\mu_G}\right]^a\left[\frac{D_C V_L \rho_L}{\mu_L}\right]^b\left[\frac{D_{HCl}\, C_{HCl}}{\mu_L}\right]^c\left[\frac{H}{D_C}\right]^d\left[\frac{d_h}{D_C}\right]^e\left[\frac{\mu_G}{\mu_L}\right]^f\left[\frac{\rho_G}{\rho_L}\right]^g \quad (3)$$

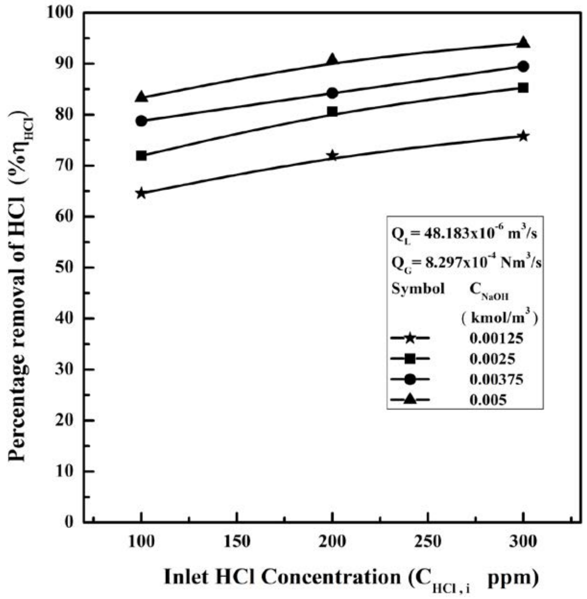

Figure 8: Effect of C_{NaOH} on percentage removal of HCl (%η_{HCl}).

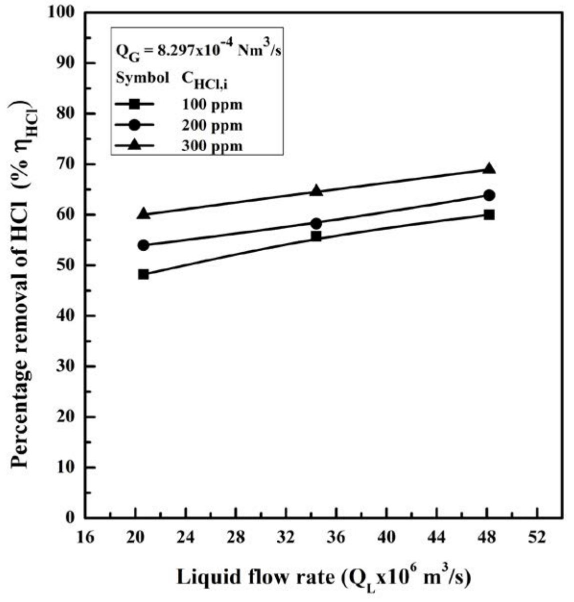

Figure 9: Effect of liquid flow rate on the η_{HCl} for water scrubbing at different constant $C_{HCl,i}$ and Q_G=8.297 × 10^{-4} Nm^3/s.

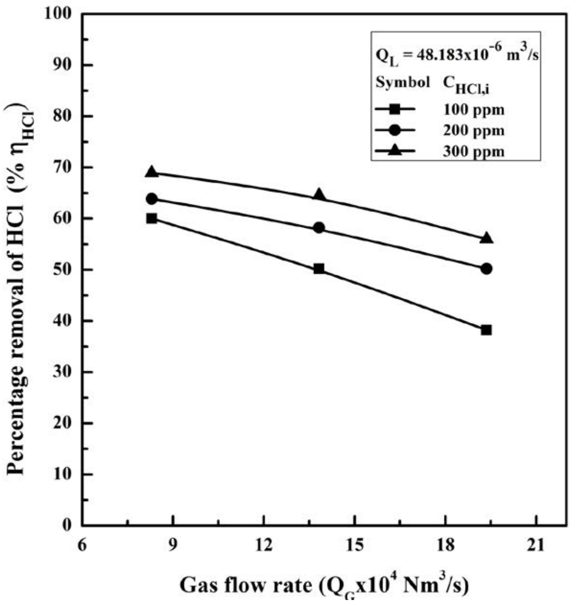

Figure 10: Effect of gas flow rate on the η_{HCl} for water scrubbing at different constant $C_{HCl,i}$ and Q_L=48.183 × 10^{-6} m³/s.

$$\eta_{HCl-H_2O} = k_0 \left[Re_G\right]^a \left[Re_L\right]^b \left[Sc\right]^c \left[\frac{H}{D_C}\right]^d \left[\frac{d_h}{D_C}\right]^e \left[\frac{\mu_G}{\mu_L}\right]^f \left[\frac{\rho_G}{\rho_L}\right]^g \tag{4}$$

In equation (4), as the values of $\left[\frac{d_h}{D_C}\right]^e \left[\frac{\mu_G}{\mu_L}\right]^f \left[\frac{\rho_G}{\rho_L}\right]^g$ and $\left[\frac{H}{D_C}\right]^d$ remained constant or their variation were negligible.

$$\eta_{HCl-H_2O} = k_0 \left[Re_G\right]^a \left[Re_L\right]^b \left[Sc\right]^c \tag{5}$$

$$\eta_{HCl-H_2O} = 7.87\times10^2 \left[Re_G\right]^{-0.471} \left[Re_L\right]^{0.344} \left[Sc\right]^{0.298} \tag{6}$$

Where k_0 and k are correlation constants and f is function of variables.

The major objective of this work is to analyze the HCl absorption in alkaline systems and prediction of the HCl removal efficiency for these systems. When HCl gas is absorbed into the water, reaction (A) occurs:

$$HCL_{(g)} + H_2O \rightarrow H^+_{(aq)} + Cl^-_{(aq)} \tag{A}$$

In particular situation of HCl absorption into aqueous NaOH solution, reaction (B) should be taken into account along with the reaction (A):

$$H^+_{(aq)} + OH^-_{(aq)} \rightleftarrows H_2O \tag{B}$$

With the overall reaction as

$$HCl + NaOH \rightarrow NaCl + H_2O \tag{C}$$

The stoichiometric factor of reaction (C) is 1, therefore the mole ratio, x, is 1.

The interfacial concentration of HCl can be calculated using equation (7). The expression which has been used to estimate HCl interfacial concentration is,

$$C_{HCL,int} = \frac{P_{HCl}}{H} \tag{7}$$

Where $C_{HCl,int}$ is the interfacial of HCl (kmol/m³), H is the Henry's law constant (atm.m³/kmol), P_{HCl} is HCl partial pressure in bulk gas (atm).

In the scrubber, the removal efficiency of HCl is depends on the ratio of HCl mass transfer rates with and without chemical reaction. The HCl percentage removal for alkaline scrubbing $(\eta_{HCL\text{-}NaOH})$ can be written as [13,18,19]

$$\eta_{HCl-NaOH} = \eta_{HCl-H_2O} \cdot \left[1 + \left[\times \frac{D_{NaOH} C_{NaOH}}{D_{HCl} C_{HCl}, int}\right]\right] \tag{8}$$

Where η_{HCl-H_2O} is HCl percentage removal of water scrubbing and can be calculated using Equation (6) and D_{NaOH} is diffusion coefficient of NaOH in water (m²/s). The right-hand side second term in Equation (8) generally gives the mass transfer enhancement factor [20]. The comparison and deviation between the predicted and experimental values of HCl percentage removal are shown in Figures 11 and 12 respectively. The calculated HCl percentage removal values with Equation (8) have been taken as 1.0 for the values which exceed 1.0 because, the 100% or more removal cannot be achieved in practical situations.

Conclusions

The percentage removal of HCl is increased with the increase in scrubbing liquid flow rate and decreased with the increase in gas flow rate at constant inlet concentrations of HCl. It can also be concluded from the results that the percentage removal of HCl increased with inlet concentration of HCl which increases the solute concentration gradient between gas and liquid phase. This gradient in solute concentration increases the driving force for absorption into the liquid phase where the reaction between HCl and NaOH occurs. Similar trends are observed with different concentrations of sodium hydroxide solutions and the efficiency range is increased for HCl removal with increase in concentration of NaOH solution. The maximum percentage removal of HCl of the dual-flow sieve plate column wet scrubber is observed as 93.98% at 8.297 × 10^{-4} Nm³/s gas flow rate, 48.183 × 10^{-6} m³/s liquid flow rate and 300 ppm inlet HCl concentration with 0.005 kmol/m³ NaOH solution. A model has also been developed for HCl

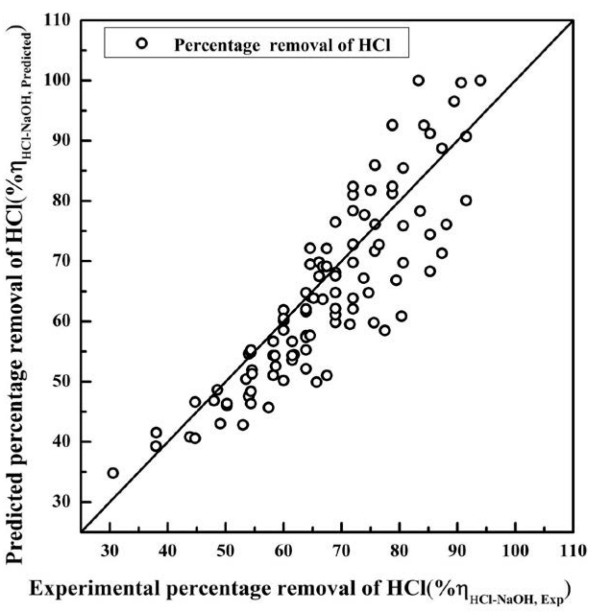

Figure 11: Comparison between the predicted and experimental values of %$\eta_{HCl\text{-}NaOH}$.

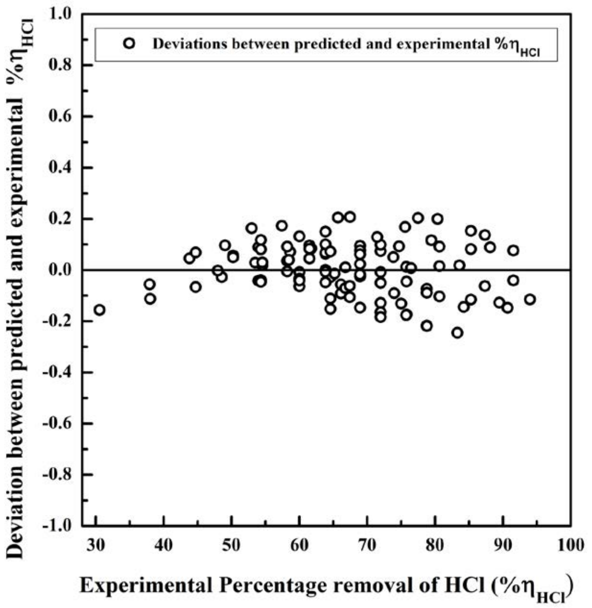

Figure 12: Deviation between the predicted and experimental values of $\%\eta_{HCl\text{-}NaOH}$.

scrubbing using the experimental results and the parameters which affect the percentage removal of HCl. Almost 80% of the experimental results matched well with the predicted values with less than ± 15% deviation and rest matched well with less than ± 25%.

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