EXPERIMENTAL EVALUATION OF THE REMOVAL EFFICIENCY OF SO_2 IN A SPRAY TOWER USING DIFFERENT SPRAY NOZZLES

Milene Costa Codolo, milenec@fem.unicamp.br
Waldir Bizzo, bizzo@fem.unicamp.br
Faculty of Mechanical Engineering – State University of Campinas - Mendeleyev St, 200, Campinas, SP - Brazil

Abstract. *This work presents an experimental evaluation of the removal efficiency of SO_2 in a spray tower. The experiments were carried out in different conditions, varying gas velocity and using different sprays nozzles. The influence of the height of tower on the removal efficiency was evaluated through experiments inside spray tower. In this study was used two sets of five nozzles, with diameter of orifice of 2.4 and 3.2 mm, and only one nozzle with diameter of orifice of 5.6 mm. The results showed the influence of the gas velocity and L/G ratio in the removal efficiency, the influence of the gas velocity on the volumetric gas side mass transfer coefficient and the influence of the height of the tower in the removal efficiency.*

Keywords*: spray tower, sulphur dioxide, absorption, and mass transfer.*

1. NOMENCLATURE

a specific interfacial area (m^2/m^3)
A area of the tower (m^2)
C_{inlet} SO_2 concentration at the inlet (mol/m^3)
C_{exit} SO_2 concentration at the exit (mol/m^3)
Ef removal efficiency of SO_2 (%)
Do diameter of nozzle orifice (mm)
G gas flow rate (m^3/h)
h height of the tower (m)
kg gas side mass transfer coefficient ($kmol/m^2$ s atm)
Kg overall gas side mass transfer coefficient ($kmol/m^2$ s atm)
kga gas side mass transfer volumetric coefficient ($kmol/m^3$ s atm)
L liquid flow rate (l/h)
L/G liquid/gas ratio (l/m^3)
M molecular weight (kg/kmol)
Vg gas velocity (m/s)
y_{inlet} mole fraction of SO_2 at the inlet
y_{exit} mole fraction of SO_2 at the inlet

Greek Symbols

ρ gas density (kg/m^3)

2. INTRODUCTION

The spray tower is a gas-liquid contacting equipment widely used in industry. In a spray tower the liquid is sprayed in fine droplets, to produce great interfacial area for mass transfer between the continuous phase and the dispersed phase. Some of the main advantages of the spray tower are the high capacity of treatment, low pressure drop and low investment cost (Pinilla et al., 1984; Tanniguchi et al.,1997; Turpin et al., 2008).

The performance of a spray tower is difficult to predict, because of droplet size and distribution, coalescence between the droplets, oscillation and distortion of droplets (Metha and Sharma, 1970; Taniguchi et. al, 1997; Turpin et al., 2008). The removal efficiency of spray tower depends mainly on the spray hydrodynamics, physic-chemical properties of the system, operating variables, as gas and liquid flow rates, and dimensions, as height and cross-sectional area of the spray tower (Bandyopadhyay and Biswas, 2007).

In literature there are several experimental studies using in spray towers. Schmidt and Stichmair (1991) carried out a study in concurrent spray tower for SO_2 absorption, the study showed that the gas velocity has little influence in the mass transfer rate. Taniguchi et al. (1997) carried out an experimental study of CO_2 absorption and the properties of spray, the results showed that the mean diameter of the droplets does not change appreciably with of the distance from the nozzle exit, but decreases with increase of liquid flow rate. In the work carried out by Bandyopadhyay and Biswas (2006), the results showed that the SO_2 concentration does not have significant effect in the removal efficiency. Turpin et al. (2008) carried out an experimental study of the removal efficiency of H_2S, they concluded that for a given liquid velocity, the interfacial area increase with an increasing gas velocity. The studies from Pinilla et al. (1984), Javed et al. (2006) and Turpin et al. (2009) showed that the volumetric gas side mass transfer coefficient (*kga*) increases continuously with increasing gas velocity.

When SO_2 is absorbed in water, the following reactions (1) and (2) occur in the liquid phase:

$$SO_{2(g)} + H_2O \Leftrightarrow H^+ + HSO_3^- \quad (1)$$

$$HSO_3^- \Leftrightarrow H^+ + SO_3^{2-} \quad (2)$$

When SO_2 is absorbed into aqueous NaOH solutions the following two reactions should be considered, in addition to hydrolytic reaction (1) and (2):

$$SO_{2(g)} + OH^- \Leftrightarrow HSO_3^- \tag{3}$$

$$HSO_3^- + OH^- \Leftrightarrow SO_3^{2-} + H_2O \tag{4}$$

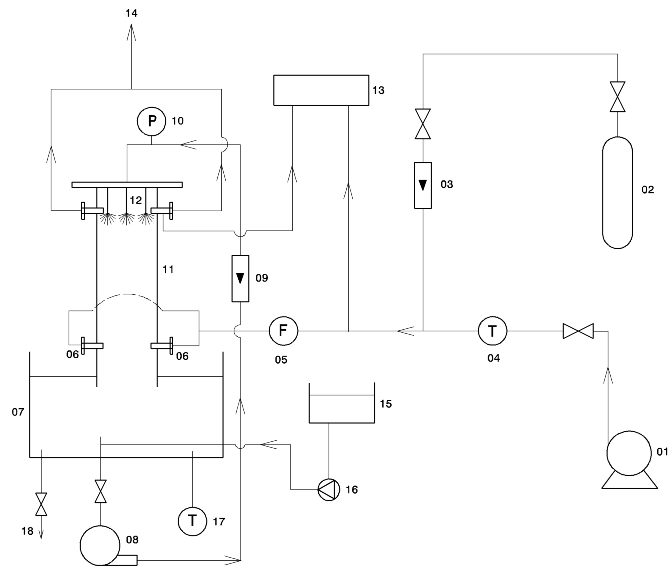
This work studies the removal efficiency of sulphur dioxide in spray tower with sodium hydroxide solution. The experimental work was carried out to evaluate the influence of the gas velocity, the diameter of nozzle orifice, the number of nozzles used in the spray tower and the profile of concentration along of the height of tower at different operation conditions. The gas side mass transfer volumetric coefficient (*kga*) was calculated from the experimental data and the effect of the gas velocity on the *kga* was analyzed.

3. EXPERIMENTAL FACILITIES AND PROCEDURES

Figure 1 shows a schematic diagram of the experimental apparatus. The plant consisted of an acrylic column with diameter of 0.29 m and height of 1.5 m. The fluids (liquid e gas) circulated in counter current in the tower. The liquid was distributed thanks to solid cone spray nozzle located in the top of the tower. The experiments were carried out with set of nozzles, with orifice diameter of 2.4 mm and 3.2 mm, and only one nozzle, with orifice diameter of 5.6 mm.

The experiments were conducted at gas flow rates of 95.1, 142.7, 190.2, 237.8, 285.3, 332.9 and 380.5 m^3/h, which results in gas velocity inside the tower from 0.4 to 1.6 m/s, and liquid flow rate of 1500 l/h. In these operation conditions, two set of five nozzles of orifice diameter of 2.4 and 3.2 mm and only one nozzle of orifice diameter of 5.6 mm were used in the experiments.

The experiments were carried out with hydroxide solution. The gas, mixture of air and SO_2, was prepared by injecting pure SO_2 in the air line. The SO_2 flow rate was measured by means of a calibrated rotameter to reach the necessary concentration.



1-Blower, 2-SO_2 cylinder, 3-SO_2 rotameter, 4-Gas temperature measurement, 5-Orifice plate, 6-Gas inlet, 7-Liquid storage tank, 8-Centrifugal pump, 9-Liquid rotameter, 10-Pressure measurement, 11-Spray tower, 12-Nozzles, 13-Gas Analyzer, 14-Gas outlet to atmosphere, 15-solution tank (NaOH), 16-Peristaltic pump, 17-Liquid temperature measurement and 18-Drainage

Figure 1. Schematic diagram of the experimental apparatus

Inside the tower the measurements were carried out to obtain the profile of SO_2 concentration. The measurements inside the tower were difficult, due to very large number of droplets. To collect the gas sample, it was built a probe, which was introduced inside the tower, through the top of the tower between the nozzles. The probe was connected to a flexible tube, which way the gas sample went to the gas analyzer. The probe was constituted of four modules and separated by a nylon mesh, with thickness of 0.3 mm and opening of 1.3 mm x 1.3 mm. The first measurement was carried out at 125 mm and the last at 1250 mm from the gas inlet. The concentrations in the inlet and exit tower were measurement out of column, without humidity interference. The experiments were conducted only in one gas velocity of 1 m/s. Figure 2 shows the probe for sample collection and Fig. 3 shows the probe inside the tower.

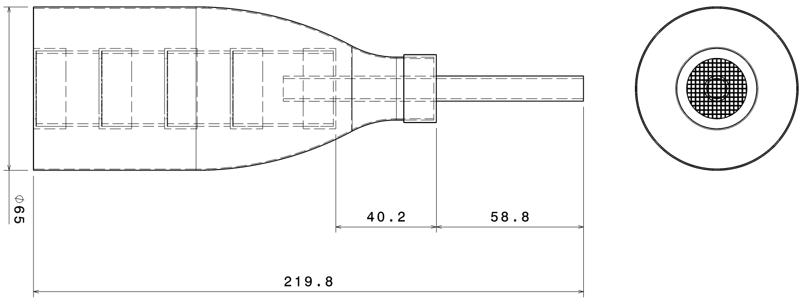


Figure 2. Probe to collect gas sample.

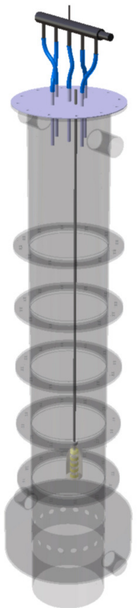


Fig 3: Probe for sample collection inside of the tower.

The measurements of SO_2 concentration were carried out by means of a gas analyzer HORIBA (ENDA-1000). In all experiments, the concentrations were measured five times, with 4% maximum deviation .

4. RESULTS AND DISCUSSION

From the experimental data was calculated removal efficiency of SO_2 in the spray tower by Eq (5):

$$Ef\,(\%) = \frac{C_{inlet} - C_{exit}}{C_{inlet}} \cdot 100 \tag{5}$$

Figure 4 shows the influence of the gas velocity and the *L/G* (liquid/gas ratio), in removal efficiency. As can be seen, the increasing gas velocity did not affect the removal efficiency, when the set of nozzles with *Do* 2.4 mm was used. However, the removal efficiency decreased with the increasing velocity for the set of nozzles with *Do* 3.2 mm and one nozzle with *Do* 5.6 mm. The removal efficiency decreased with the orifice diameter increase for the set of five nozzles and the removal efficiency was greater using only one nozzle with *Do* 5.6 mm than using the set of nozzle with *Do* 3.2 mm. In the last case the use of only one nozzle must have produced smaller droplets, generating larger interfacial area than using set of nozzles with *Do* 3.2 mm. It can be seen in the figure that a given *L/G* ratio may result in different removal efficiencies depending on the used nozzles. The choice of spray nozzles is of the great importance, whereas the nozzle produces the interfacial area available for mass transfer.

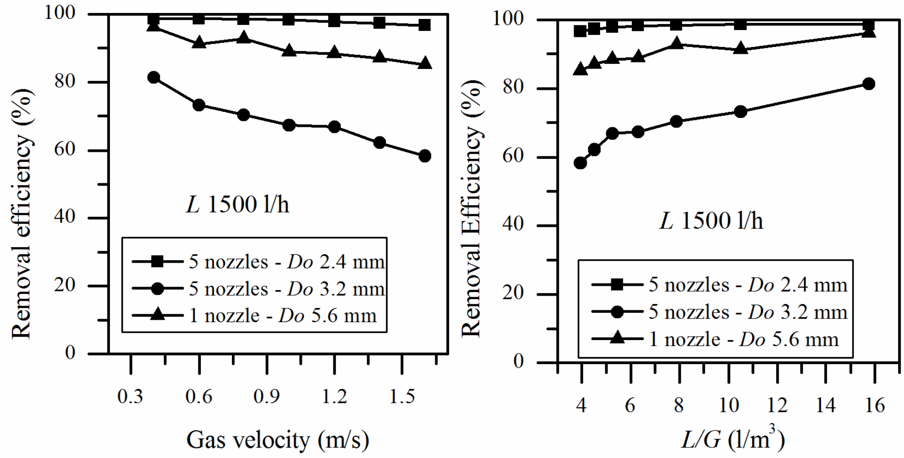


Figure 4. Influence of the gas velocity and *L/G* on the removal efficiency of SO_2.

The mass transfer coefficient *kg* and the interfacial area of the droplets are two important parameters of mass transfer in spray towers. According Danckwerts (1970), for systems which the gas phase resistance controls process of mass transfer and the reaction between gas and liquid is instantaneous and irreversible, and the mass transfer volumetric coefficient (*kga*) can be calculated by the Eq 6:

$$kga = \frac{G\rho}{AhM}\ln\left(\frac{y_{inlet}}{y_{exit}}\right) \tag{6}$$

The absorption of sulfur dioxide in aqueous sodium hydroxide solution is accompanied by an instantaneous chemical reaction between dissolved sulfur dioxide ions and OH^- ions (Hikita, et al., 1977). In this system, dissolved sulfur dioxide reacts with an excess reagent at the gas-liquid interface and the liquid phase resistance can be negligible (Chang and Rochele, 1981). In systems using highly soluble gases, such as SO_2, gas phase resistance controls the process mass transfer, therefore *Kg* can be considered approximately equal to *kg*.

Figure 5 shows the influence of gas velocity in the mass transfer volumetric coefficient. As can be seen in Fig.5, *kga* increases with increasing gas velocity. The velocity increase had the greatest influence in the set of nozzles with *Do* 2.4 mm. This can be due to the smaller diameter of droplets produced by the nozzles, whereas the nozzle produce a distribution of diameter and the smaller droplets can have stayed in suspension, what increased the interfacial area and consequently *kga* was increased.

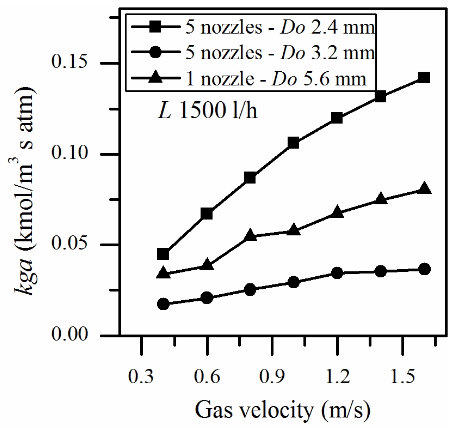


Figure 5. Influence of the gas velocity on the *kga*.

Figure 6 shows the evolution of the concentration inside the spray tower. In the first point inside the tower occurred great reduction of the SO_2 concentration, in the three studied cases. The profile of reduction of the concentration was similar for the set of nozzles with *Do* 2.4 mm and only one nozzle with Do 5.6 mm, nevertheless the reduction of the concentration, for the set of nozzles with *Do* 3.2 mm, was more discreet. The reduction of concentration occurred up to

1 m (from the gas inlet), and from this height of tower the SO_2 concentration was constant or increased lightly up to the end of the tower. The concentration increase must have occurred due to humidity inside the probe. The humidity can have affected the measurements by absorption of SO_2 inside the probe, thus the real concentration must be larger than the measured concentration. This can be clearly noticed in the last sampling point, whereas in this measurement the sample was collected out of tower, thus outside the spray zone, therefore without influence of the humidity inside the probe.

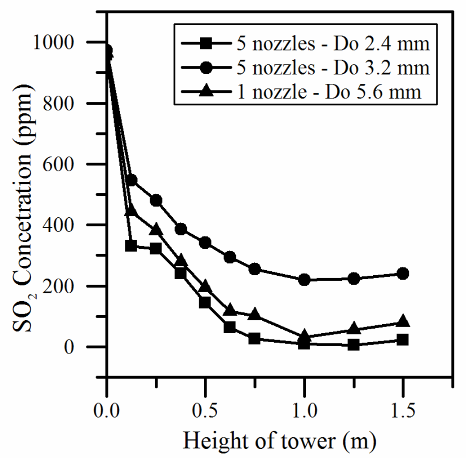


Figure 6. Influence of the height of the tower on SO_2 concentration.

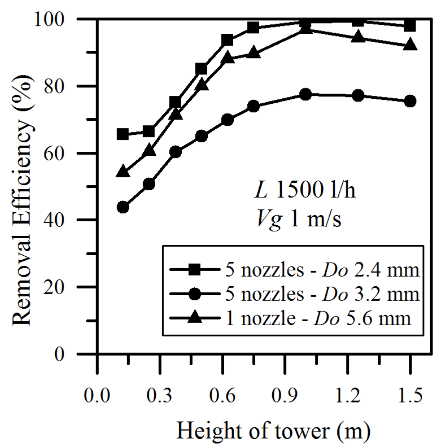


Fig. 7: Influence of the height of the tower on the removal efficiency of SO_2.

As shown in Figure 7 the efficiency increase was significant up to 1 m (from the gas inlet), the set of nozzles with *Do* 2.4 mm and only one nozzle with *Do* 5.6 mm showed higher efficiency and similar profile of the removal efficiency. From this point, the efficiency was constant or decrease slightly, due to measurement of the SO_2 concentration, as previously explained. In general, most of the SO_2 absorption occurred at the bottom of the tower, up to 1 meter from the gas inlet. The liquid, when leaves the nozzle, has high velocity, however, due to drag force the droplets decelerate along the tower. As the relative velocity between the droplets and gas decrease, the residence time of the droplets increases, increasing the interfacial area available or mass transfer. At the bottom of the tower, the cross section is completely covered by the droplets, and there is a turbulence zone due to the gas inlet by distribution chamber orifices, which also contribute for mass transfer.

5. CONCLUSIONS

The results showed the influence of the gas velocity in removal efficiency for the set of nozzles with *Do* 3.2 mm and only one nozzle with *Do* 5.6 mm and the influence of the nozzle on the removal efficiency, whereas a given *L/G* can produce different results, depending on the choice of the nozzle. The results also showed the great influence of the gas velocity in the mass transfer volumetric coefficient (*kga*), mainly in the set of nozzles with *Do* 2.4 mm.

The removal efficiency was significant up to height of 1 m, from this height the efficiency was constant or decrease. The efficiency decrease in the end of the tower showed the interference of the humidity inside the probe, whereas the last measurement was carried out in the tower exit, therefore out of the spray zone. The nozzles, which had higher efficiencies, showed profile of efficiencies more pronounced up to height of 1 m.

This experimental work showed the importance of the choice of the spray nozzles for spray towers**.** The diameter of the orifice had a great effect on the removal efficiency, whereas the set of nozzles with *Do* 3.2 mm obtained efficiency lower than the set of nozzles with *Do* 2.4 mm. The number of nozzles also showed influence on the removal, whereas only one nozzle obtained efficiency smaller than the set of nozzles with *Do* 2.4 mm, when only one nozzles is used in the tower the covering of the tower volume by droplets is smaller. The choice of the nozzles appears to be directly related to interfacial area available for mass transfer. Nozzles with larger orifice produce larger droplets and consequently smaller interfacial.

6. ACKNOWLEDGEMENTS

The authors gratefully acknowledge the financial support of Capes and Fapesp.

7. REFERENCES

Bandyopadhyay, A. and Biswas, M. N., 2006. "Prediction of the removal efficiency of a novel two-stage hybrid scrubber for flue gas desulfurization". *Chemical Engineering Technology*, Vol. 29, pp. 130-145.

Chang, C. S. and Rochelle, G. T., 1981. "SO_2 Absorption into Aqueous Solution". *Aiche Journal*, Vol. 27, pp. 292-298.

Danckwerts, F.R.S., 1970. *Gas – Liquid Reactions*. MacGraw-Hill, New York.

Hikita, H., Asi, S. and Tsuji, T., 1977. "Absorption of sulfur dioxide into aqueous sodium hydroxide and sodium sulfite solutions". *Aiche Journal*, Vol. 23, pp. 538-544.

Javed, K. H., Mahmud, T. and Purba, E., 2006. "Enhancement of mass transfer in a spray tower using swirling gas flow". *Chemical Engineering Research and Design*, Vol. 84, pp.465-477.

Mehta, K. C. and Sharma, M. M., 1970. "Mass transfer in spray columns". *British Chemical Engineering*, Vol. 15, pp.1440-1444.

Pinilla, E. A., Díaz, J. M. and Coca, J., 1984. "Mass transfer and axial dispersion in a spray tower for gas-liquid contacting". *The Canadian Journal of Chemical Engineering*, Vol. 62, pp. 617-622.

Schmidt, B. and Stichlmair, J., 1991. "Two phase flow and mass transfer in scrubber". *Chemical Engineering Technology*, Vol. 14, pp. 162-166.

Taniguchi, I., Takamura, Y. and Asano, K., 1997. "Experimental study of gas absorption with a spray column". *Chemical Engineering of Japan*, Vol. 30, pp. 427-433.

Turpin, A., Couvert, A., Laplance, A. and Paillier. A., 2008. "Experimental study of mass transfer and H_2S removed efficiency in a spray tower". *Chemical Engineering and Processing*, Vol. 47, pp. 886-892.

Turpin, A., Couvert, A., Laplance, A. and Paillier, A., 2009. "Mass transfer and deodorization efficiency in a countercurrent spray tower for low superficial gas velocities". *The Canadian Journal of Chemical Engineering*, Vol. 87, pp. 53-59.

8. RESPONSIBILITY NOTICE

The authors are the only responsible for the printed material included in this paper.