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Gas pollutants removal in a single- and two-stage ejector–venturi scrubber

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Abstract

The absorption of SO_2 and NH_3 from the flue gas into NaOH and H_2SO_4 solutions, respectively has been studied using an industrial scale ejector–venturi scrubber. A statistical methodology is presented to characterise the performance of the scrubber by varying several factors such as gas pollutant concentration, air flowrate and absorbing solution flowrate. Some types of venturi tube constructions were assessed, including the use of a two-stage venturi tube.

The results showed a strong influence of the liquid scrubbing flowrate on pollutant removal efficiency. The initial pollutant concentration and the gas flowrate had a slight influence. The use of a two-stage venturi tube considerably improved the absorption efficiency, although it increased energy consumption.

The results of this study will be applicable to the optimal design of venturi-based absorbers for gaseous pollution control or chemical reactors.

Keywords: Venturi scrubber; Absorption; Sulphur dioxide; Ammonia; Statistical modelling

1. Introduction

Several options are available for the control of particulate matter from flue gases such as cyclones, settling chambers, fabric filters, electrostatic precipitators, and various types of wet scrubbers. Amongst the wet scrubbers, the venturi scrubber is unique in that it is not only very efficient for the collection of particulates but can also function as a gas absorber.

The ejector–venturi scrubber is a special design of venturi-based absorbers. The main characteristic is the liquid injection mechanism, based on a mechanical atomization system.

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In this study, a pressure-swirl atomizer was used. The principle of operation involves a jet effect created by the water (or aqueous solution) spray nozzle. This spray nozzle is located on the top of the jet scrubber and creates a full cone-shaped spray. This is a relatively narrow-angle spray, which contacts the wall of the jet scrubber at a point above the throat. The result is an induced airflow through the scrubber. The gas and liquid enter the throat, where extreme turbulence is encountered, and continue through the diffuser section where partial separation of the gas and liquid occurs. The concurrent nature of this scrubber requires a separation device to be used to separate the gas completely from the liquid. Mechanical devices such as blowers are used when high gas flowrates are desired.

The scrubbing mechanism includes the cross-flow effect of the air being entrained through the spray plus the turbulence, which occurs at the throat area.

While the efficiency of venturi scrubbers for the removal of particulate matter has been extensively studied, only a few papers on gas pollutants absorption have been reported. Johnstone et al. [1] found that efficiencies for SO_2 removal by alkaline solutions in venturi scrubbers were proportional to the specific surface area of the droplets; they also found that the gas mass transfer coefficient increased substantially as the liquid injection rate increased. In the Russian literature, several papers related to venturi scrubbers can be found, most of them theoretical approaches (Kuznetsov and Oratovskii [2], Boyadzhiev [3], Elenkov and Boyadzhiev [4] and Volgin et al. [5]). The first paper related to complete venturi scrubber modelling was published by Uchida and Wen [6]. These authors performed mass, heat and momentum balances for SO_2 removal in an industrial scale venturi scrubber pilot plant. For mass transfer, they developed material balances in the droplet without chemical reaction. They solved the equation analytically and obtained the rate of physical absorption. Chemical reaction was applied as an enhancement factor derived from film theory.

Cooney [7] proposed a much more realistic model of the liquid-side mass transfer and reaction involving dividing the droplet into 50 concentric shells of equal thickness and writing down the flux of each species present into and out of the shell in finite difference form. Ravindram and Pyla [8] used a simpler hydrodynamic model by assuming homogeneous flow in which liquid and gas flow at the same velocity (no-slip assumption). They also neglected gas-side mass transfer resistance and adopted a solution given by Crank [9] for diffusion in a sphere accompanied by a first order reaction.

Atay et al. [10] developed empirical models to describe the fluid flow characteristics and gas absorption efficiency of an ejector–venturi scrubber. They also performed mechanical energy and material balances considering isothermal systems. This is the only work devoted to mass transfer in ejector–venturi scrubbers found in the literature. More recently, Hills [11] proposed solutions to the equations of absorption with first order chemical reactions in spheres. The author solved the equations under several boundary and initial conditions, although he neglected the hydrodynamics of the process. He used the experimental data of Ravindram and Pyla and found serious discrepancies with their conclusions. Finally, Talaie et al. [12] proposed a model for determining the absorption efficiency in venturi scrubbers based on a non-uniform droplet concentration distribution. They also used dispersion models for gas pollutant concentration, distribution and material balance in a differential control volume in liquid phase. Gas-phase mass transfer coefficient was calculated by empirical equations and film theory mass transfer coefficient was used for the liquid side. They neglected the droplet–scrubber walls interactions.

All these papers, except the last, consider that the entire amount of the liquid injected into the throat section is completely atomized into fine droplets of uniform diameter moving in plug flow towards the end of the diffuser. For jet–venturi scrubbers, where the liquid is sprayed into the gas flow in the venturi throat, initially all the liquid will be in the form of droplets. However, some of the liquid soon deposits to form a wall film. There can be re-atomization at the throat if the liquid is introduced as drops upstream of the throat. The existence of the liquid film promotes the appearance of several phenomena quoted by Azzopardi and co-workers [13–17]. The most important one is that the film has a much lower surface area per unit volume than the drops. Another consequence is that the film on the wall will act as a rough surface to the gas phase and thus, affect the pressure drop. To some extent, the geometry of the venturi allows the re-entrainment and deposition of droplets.

As we have seen, several theories have been put forward to describe the scrubbing action and predict its effectiveness, but to date they have been unable to come up with a single theory that predicts the results under varying operating conditions. The complexity of the coupled phenomena (mass transfer in two-phase systems and hydrodynamics) makes the study of venturi scrubbers very difficult to overcome by using mass, energy and momentum balances. Pilot plant studies are the principal means of obtaining data on this type of unit.

The aim of this study is to evaluate the suitability of an ejector–venturi scrubber for the removal of two common stack gases, sulphur dioxide and ammonia. From this work, it will be possible to determine the influence of several operating variables besides different venturi tube constructions. The conclusions and the methodology employed in this study will be useful for the design of these systems.

2. Experimental

2.1. Apparatus and procedure

Fig. 1 shows a schematic diagram of the jet–venturi scrubber equipment at pilot plant scale. A blower generated the gas flowrate and the gas pollutant was introduced from a cylinder. The mixture was passed through the venturi throat where atomization of the liquid absorbent occurs (via a pressure-swirl atomizer). The reservoir produced the gas–liquid phases separation.

The equipment was built in PVC and allowed several construction shapes, as shown in Fig. 2. The first configuration (Fig. 2a) consisted of a complete venturi tube with one nozzle. The main difference between configuration one and two (Fig. 2b) was that the latter was installed without a diffuser. This was done in order to examine the influence of the reactor volume in the absorption process. Configuration three (Fig. 2c) consisted of a two-stage jet–venturi scrubber with two nozzles, and finally, configuration four (Fig. 2d) was the same as configuration three but equipped with the upper nozzle only. Experiments were carried out with two common stack gases, sulphur dioxide and ammonia. The specifications and operating conditions for the absorption process are given in Table 1.

Gas pollutant concentrations have been selected from usual values in several types of industrial flue gas compositions, so different magnitudes of sulphur dioxide and ammonia

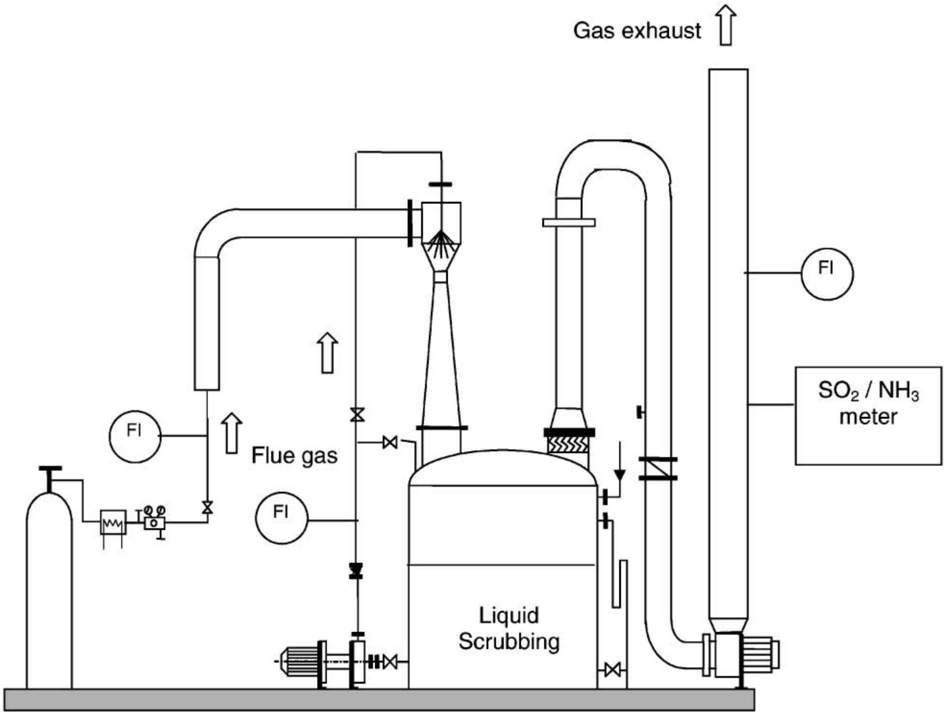

Fig. 1. Schematic diagram of the jet–venturi scrubber pilot plant showing the basic instrumentation used in the experiments.

concentrations were used. In spite of this, most of the conclusions of the present study are common for both stack gases.

The amount of sulphur dioxide or ammonia absorbed was obtained from the difference in the concentration of the flue gas between the venturi tube entrance and the exit duct. The concentration was measured by using an electrochemical continuous sensor. A Pitot tube was used to determine the air flowrate, and the liquid flowrate was measured with a rotameter.

2.2. Numerical analysis

As pointed out by Rappaport et al. [18], chemical and environmental industry processes are frequently not well understood. Therefore, experimentation is needed both to improve existing processes and to develop new ones. The information obtained in pilot plants has to be of high quality. The use of statistical modelling and design clearly improves the quality of the data. An empirical modelling technique named response surface methodology (RSM) was used to evaluate the relationship between the set of controllable factors and the experimental results. RSM is used to determine the optimum factor values for the maximum absorption efficiency. The model evaluated the effect of each independent variable on a

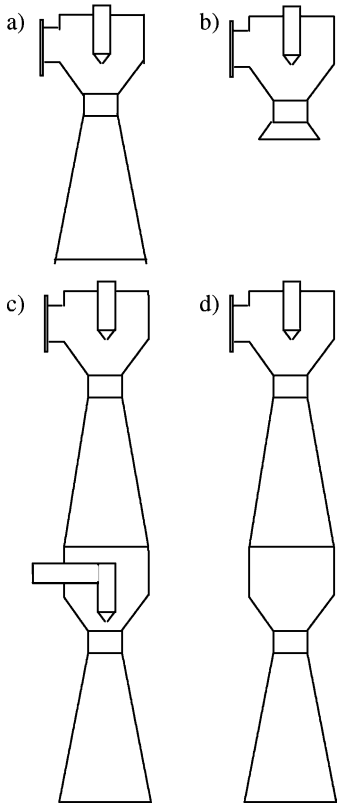

Fig. 2. Venturi tube configurations: (a) complete venturi tube with one nozzle; (b) venturi tube without diffuser; (c) two-stage jet–venturi with two nozzles; (d) two-stage jet–venturi and single nozzle.

Table 1
Specifications and operating conditions for absorption tests in the jet–venturi scrubber

Flue gas	
Temperature (°C)	15–18
Liquid scrubbing stream	
Temperature (°C)	15–18
Liquid scrubbing concentration	
NaOH (kmol/m^3)	0.5
H_2SO_4 (kmol/m^3)	0.4
Pipe diameter (mm)	250
Throat diameter (mm)	150
Diffuser length (mm)	1000
Nozzle type	Pressure-swirl atomizer
Nozzle dispersion angle (°)	90

response. Moreover, the interactions between variables were also included. The mathematical relationship between the independent variables and the response can be calculated by the quadratic polynomial equation:

$$y = \beta_0 + \sum_{i=1}^{K} \beta_i X_i + \sum_{i=1}^{K} \beta_{ii} X_i^2 + \sum_{i=1}^{K-1} \sum_{j=i+1}^{K} \beta_{ij} X_i X_j \quad (1)$$

where y is the response variable; X the main factor; K the number of factors; β_0 the constant value of the regression; β_i the linear coefficients; β_{ii} the quadratic coefficients; and β_{ij} the interaction coefficients. The computation was carried out by multiple regression analysis making use of the least squares method. The multiple correlation coefficients, determined by computation with the commercial software package, STATGRAPHICS, were used to determine the optimal model fitting, and the Student's t-value obtained showed the significant effects and interactions. The confidence level used for the determination of the coefficients was >95%.

3. Results and discussion

The main factors affecting the absorption efficiency were: gas flowrate, F_G (m^3/h); liquid scrubbing flowrate, F_L (m^3/h); and pollutant gas concentration, C_{A0} (ppm). It was observed that the liquid scrubbing concentration did not affect the absorption rate due to the close relationship between the molar liquid scrubbing flowrate and the molar gas flowrate. Consequently, the relationship between the controllable factors (F_G, F_L, C_{A0}) and the response of the system (removal efficiency, RE (%)) was evaluated for two different gas pollutants and four configurations.

Table 2 shows the experimental field for the operating variables employed in the present work.

Table 3 shows, as an example, the experimental matrix and the response obtained for the gas SO_2 and configuration 1. The same procedure was carried out for the rest of the configurations and gas pollutants.

3.1. Statistical results

For each set of experiments, a mathematical model describing the effects of the related variables on the removal efficiency was derived. The regression coefficients are summarised

Table 2
Experimental field for the operating variables and codification for the statistical analysis

Code	Variable	Values
X_1	Gas concentration (ppm)	280–1300 (SO_2), 10–40 (NH_3)
X_2	Gas flowrate (m^3/h)	500–1800
X_3	Liquid flowrate (m^3/h)	2.5–7.3
Y	Removal efficiency (%)	0–100

Table 3
Experimental matrix for the SO_2 removal efficiency using configuration 1

No.	C_{A0} (ppm)	F_G (m^3/h)	F_L (m^3/h)	RE (%)
1	287	1060	2.5	72.64
2	287	1060	5.0	78.72
3	287	1060	7.3	85.56
4	287	1700	2.5	75.43
5	287	1700	5.0	81.57
6	287	1700	7.3	86.18
7	750	500	2.5	76.47
8	750	500	5.0	85.29
9	750	500	7.3	93.23
10	750	1060	2.5	67.98
11	750	1060	5.0	75.32
12	750	1060	7.3	82.96
13	750	1700	2.5	68.87
14	750	1700	5.0	76.5
15	750	1700	7.3	81.2
16	1319	500	5.0	78.87
17	1319	500	7.3	85.53
18	1319	1060	2.5	72.21
19	1319	1060	5.0	77.04
20	1319	1060	7.3	84.69

in Table 4. The analysis of variance (ANOVA) for the three operational variables indicated that removal efficiency could be well described by polynomial models. Furthermore, the statistical analysis showed that three factors had significant effects on the response and among them, the liquid flowrate was always the most significant. This factor produced a positive linear effect in all cases. Quadratic effects are slightly significant. In addition, simple interactions between factors were not observed in the majority of the experiments. In this sense, the response variability for configuration 2 must be attributed to the linear and quadratic terms because all the interaction parameters are non-significant.

None of the models obtained includes the independent term (β_0). This is because during the multivariable regression analysis, the regression coefficient increased when this term was removed from the model. This means that the response variability depends only on the factors considered.

On the other hand, the small values obtained in the variance–covariance matrix diagonal and the almost zero remaining values indicated that the parameters were obtained with high confidence. This is due to the quasi-orthogonality of the proposed experimental design.

ANOVA for the full regressions was used to estimate the buoyancy of the models; Table 5 shows the excellent regression coefficients (R) obtained.

The high values of T (Snedecor test parameter) ensure that the model explains not only the random variation of the response but also the effect of the factors. The excellent agreement between experimental data and the model is also verified by the high squared regression coefficients. In addition, the standard deviations for the model predictions are, in all cases, less than 2%.

Table 4
Regression coefficients obtained by multiple regression analysis from the experimental data[a]

Gas	Configuration	β_1	β_2	β_3	β_{11}	β_{22}	β_{33}	β_{12}	β_{23}	β_{13}
SO_2	1	−0.013	0.085	6.985	1.2×10^{-5}	-2.8×10^{-5}	−0.259	-6.7×10^{-6}	N/S	-8.1×10^{-4}
SO_2	2	−0.019	0.084	7.642	1.1×10^{-5}	-3×10^{-5}	−0.415	N/S	N/S	N/S
SO_2	3	0.146	0.015	9.445	-1.4×10^{-4}	N/S	−0.402	N/S	N/S	−0.00121
SO_2	4	−0.007	0.098	5.114	3.4×10^{-6}	-3.5×10^{-5}	−0.259	-7.8×10^{-6}	N/S	N/S
NH_3	1	−0.416	0.121	8.007	0.008	-5.7×10^{-5}	−0.625	N/S	N/S	0.00137
NH_3	2	0.281	0.116	6.517	−0.005	-5.1×10^{-5}	−0.402	N/S	N/S	N/S
NH_3	3	N/S	0.096	8.458	N/S	-3.7×10^{-5}	−0.463	N/S	−0.008	N/S
NH_3	4	−0.763	0.125	6.767	0.012	-4.8×10^{-5}	−0.423	-1.2×10^{-6}	N/S	N/S

[a] N/S: Non-significant.

Table 5
ANOVA table for the full regressions

Gas	Configuration	R^2	T-ratio	s_n (%)
SO_2	1	0.9999	12574	0.866
SO_2	2	0.9997	1250	1.478
SO_2	3	0.9999	18354	0.781
SO_2	4	0.9998	5568	1.150
NH_3	1	0.9999	2503	0.808
NH_3	2	0.9999	11335	0.954
NH_3	3	0.9997	7501	1.755
NH_3	4	0.9999	8697	0.945

3.2. Experimental results

Figs. 3–6 show the response surface for the removal of SO_2, and Figs. 7–10 show the same for NH_3. The response surfaces show removal efficiencies greater than 65%, reaching maximum values at about 96%. These maximum values were obtained in scrubbing ammonia and are related to its higher solubility in aqueous solutions. This physical property is the most critical when one is using venturi-based scrubbers. The limited residence times found suggests that only gas pollutants having moderate or high solubility on the scrubbing liquid are appropriate.

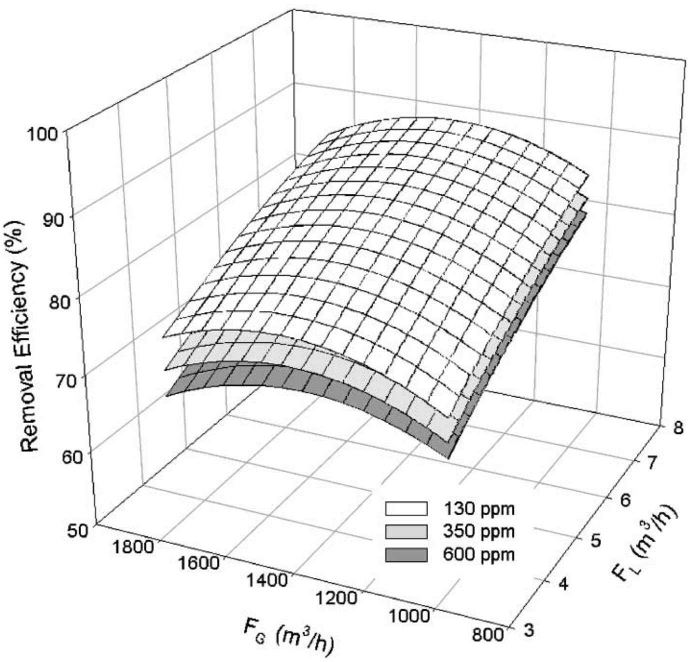

Fig. 3. SO_2 removal efficiency response surface vs. liquid and gas flowrates by using the configuration 1.

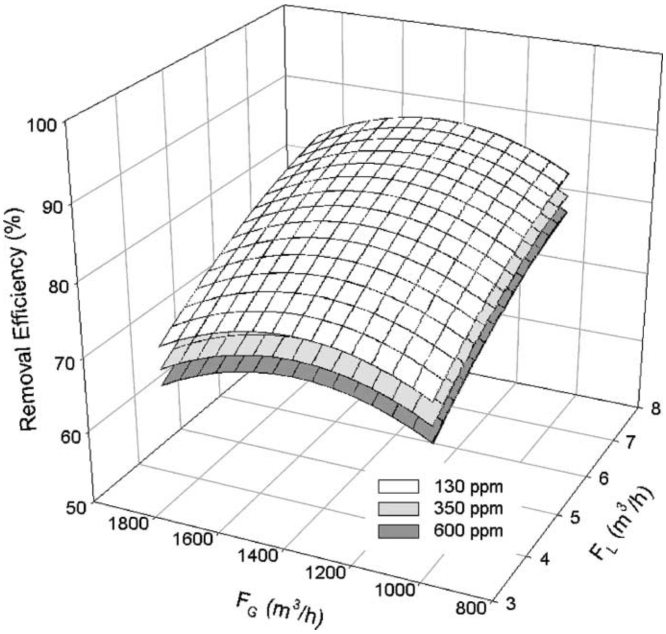

Fig. 4. SO_2 removal efficiency response surface vs. liquid and gas flowrates by using the configuration 2.

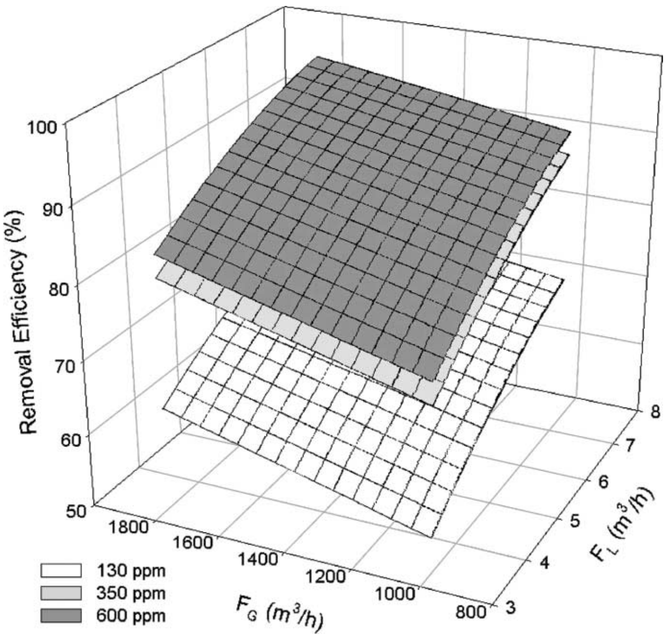

Fig. 5. SO_2 removal efficiency response surface vs. liquid and gas flowrates by using the configuration 3.

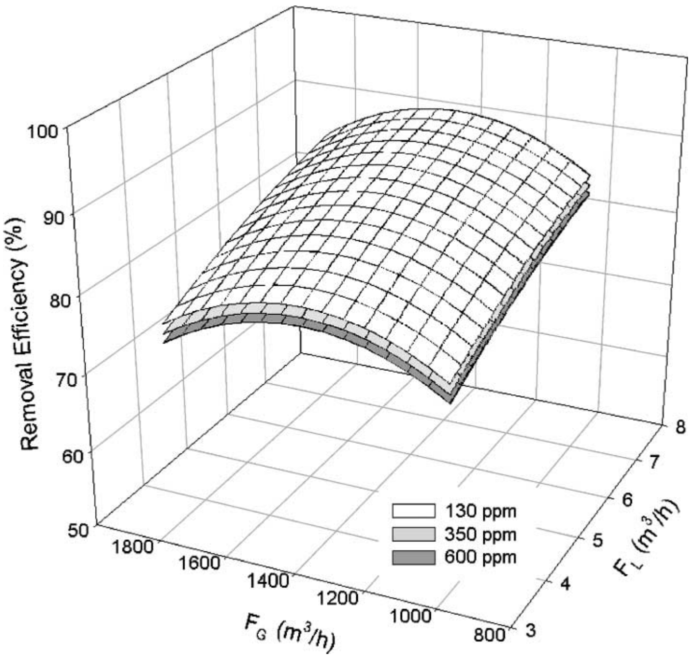

Fig. 6. SO_2 removal efficiency response surface vs. liquid and gas flowrates by using the configuration 4.

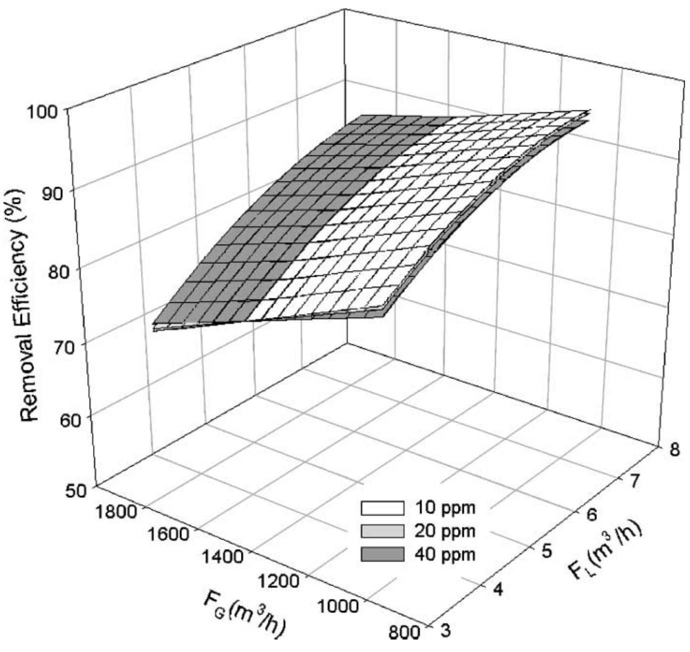

Fig. 7. NH_3 removal efficiency response surface vs. liquid and gas flowrates by using the configuration 1.

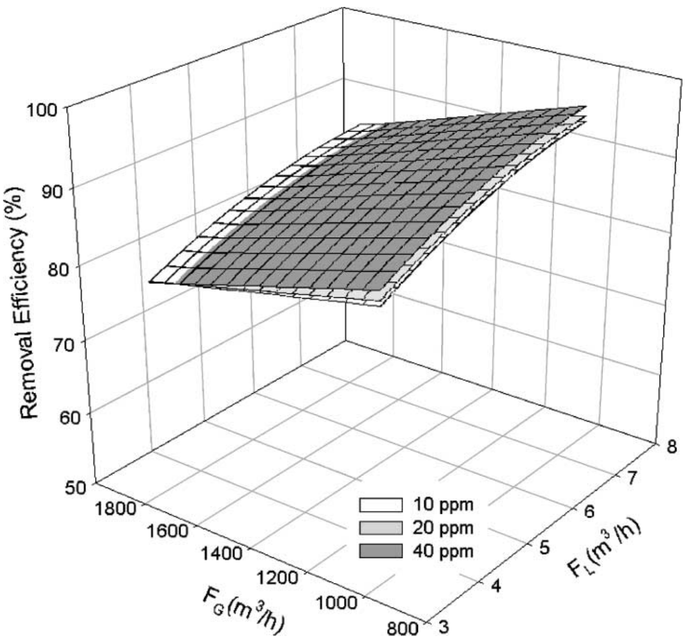

Fig. 8. NH_3 removal efficiency response surface vs. liquid and gas flowrates by using the configuration 2.

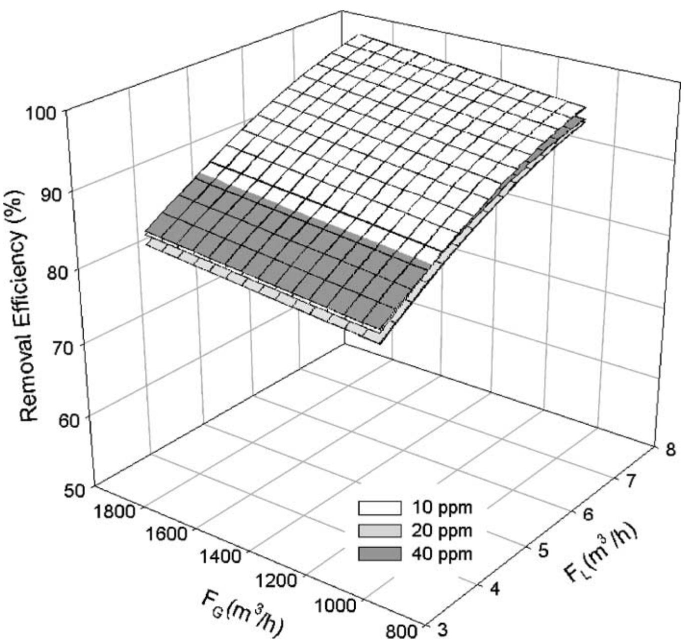

Fig. 9. NH_3 removal efficiency response surface vs. liquid and gas flowrates by using the configuration 3.

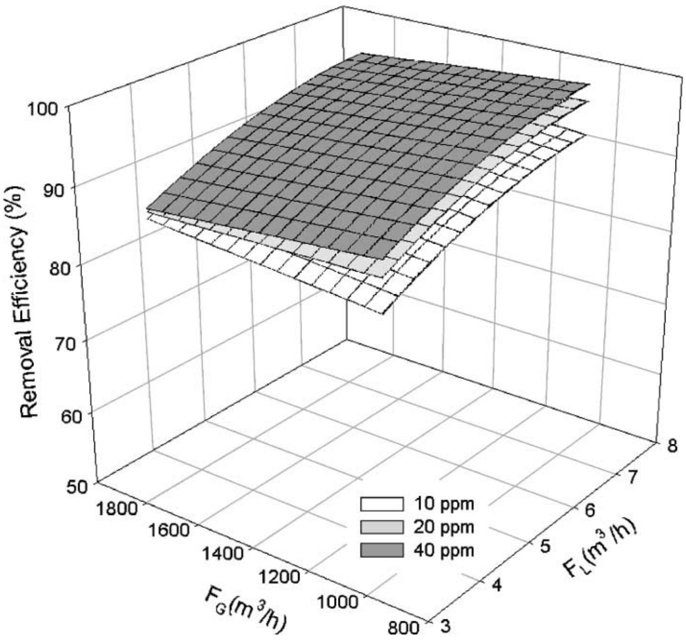

Fig. 10. NH_3 removal efficiency response surface vs. liquid and gas flowrates by using the configuration 4.

A common trend in all figures is the increase in scrubbing efficiency when the liquid absorber flowrate is increased. However, in all the response surfaces shown in Figs. 3–10, this tendency suggests a quadratic relationship, so a maximum is expected to appear. This was also observed by Atay et al. [10]. This is an important issue in optimising the power consumption efficiency because a common trend in industry is to use the maximum liquid scrubbing flowrate allowed by the pumping system. This is not usually necessary and wastes energy. The lack of non-linearity in the models proposed makes its optimisation process quite easy.

Configuration 2 is less effective in the scrubbing process. However, the efficiencies obtained were only slightly worse than those of configuration 1. The non-existence of a diffuser limited the reactor volume. High efficiencies can be explained by observing the spray shape. The full cone-shaped spray conserved its identity until it reached the liquid reservoir surface (Fig. 11), allowing a larger mass transfer surface area (and promoting a lot of turbulence). This behaviour is similar to that observed in impinging reactors. The absence of the diffuser caused the pressure recovery to be lower than in any other configuration, so from the point of view of power consumption, this was one of the most unfavourable. This was found in a previous work [19], in which the pressure drop across the venturi tube was measured in all the configurations and in different operating conditions. Fig. 12 shows the total pressure drop ($-\Delta P$) for $F_L = 5.0\,m^3/h$. It can be seen that the maximum values for pressure drop, and therefore energy consumption, were obtained for configurations 3 and 4. Configuration 2 offers a considerably higher pressure drop than configuration 1.

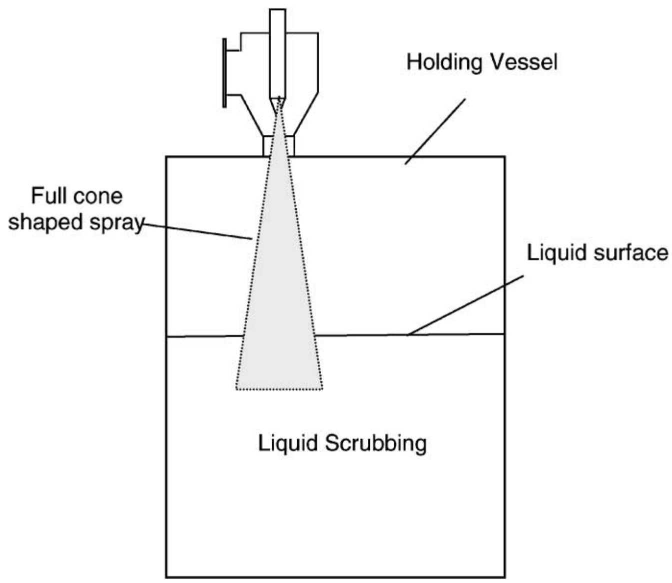

Fig. 11. Full cone spray shape formation for configuration 3.

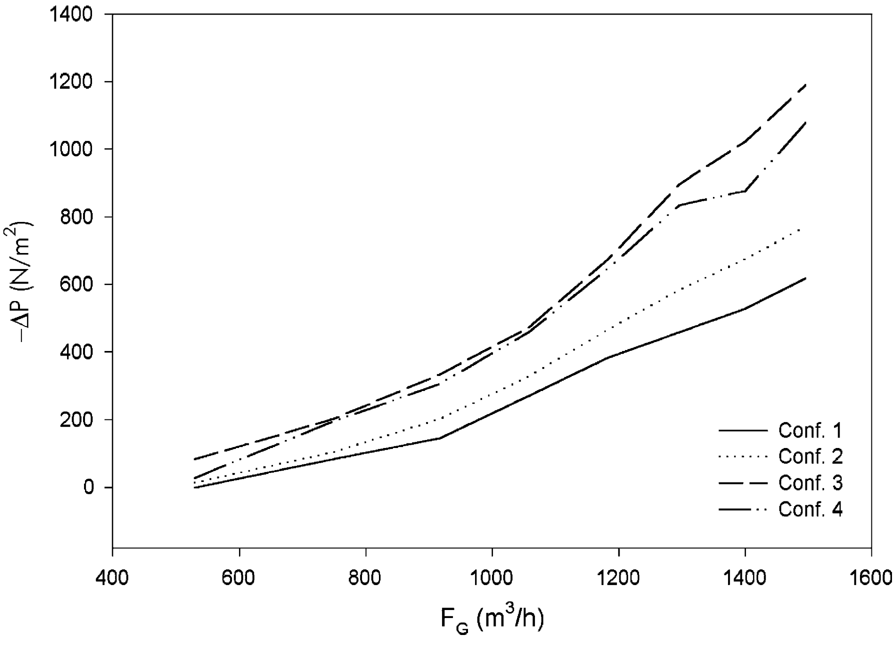

Fig. 12. Total pressure drop for all the configurations against the gas flowrate ($F_{\mathrm{L}} = 5.0\,\mathrm{m^3/h}$)

Regarding the influence of the gas flowrate for the SO_2 scrubbing efficiency, Figs. 3, 4 and 6 show maximum removal at about 1500 m^3/h. This trend is not observed in any of the configurations for NH_3 and configuration 3 for SO_2. This is due to an interaction between gas flowrate and the initial pollutant concentration, showing the maximum molar flowrate allowable for absorption.

Configuration 4 for NH_3 shows an increase in scrubbing efficiency when the gas flowrate is increased, but only at a low gas pollutant concentration. This could be explained by the fact that a fraction of the liquid absorbing solution is flowing as a liquid film along the diffuser walls. Azzopardi and Govan [13] suggested that the liquid entrainment rate was a function of the interfacial shear stress between the liquid film and the gas core. Thus, an increase in the gas flowrate will cause an increase in this stress and accordingly, an increase in droplet entrainment. Therefore, the specific mass transfer surface area is enhanced, so an improvement in gas pollutant removal is observed. At higher gas pollutant concentrations, due to the small droplet diameter of the re-atomized liquid scrubbing solution, depletion of reactant may occur, giving a drop in absorption efficiency.

Configuration 3 was expected to be the best one, based on the use of a two-stage jet–venturi scrubber. Experimental performance showed generally lower scrubbing efficiencies than those for configuration 4, though they were acceptable. As observed in Fig. 5, gas pollutant concentration has a strong effect on the scrubbing efficiency. Moreover, the gas flowrate effect in ammonia scrubbing (Fig. 9) was negligible. These results demonstrate the difficulty in understanding the interactions between two different but coupled phenomena, mass transfer with chemical reaction and two-phase flow in cone-shaped tubes.

When two nozzles (pressure-swirl atomizers) were used, high coalescence effects were observed. This coalescence has two important consequences. Firstly, it promotes the formation of higher diameter droplets, which means a fall in surface area. Secondly, the second atomizer produces a gas core deceleration, diminishing the re-atomization rate.

It is also remarkable that gas flowrate (or gas velocity) only has a slight influence on the process. This is the main difference between jet–venturi scrubbers and other venturi-based scrubbers. This performance difference is due to the liquid injection system. In the present study, the scrubbing solution is injected at high kinetic energy while in other venturi scrubbing systems, gas flowrate is the main source of kinetic energy. Thus, only re-atomization in the venturi tube is a direct consequence of gas flowrate.

Considering only scrubbing efficiency improvement, configuration 4 was the most suitable, although the results did not show major enhancements. The influence of the main operating variables is similar to that in configuration 1. The most important improvement is observed at higher gas pollutant concentrations in the case of ammonia scrubbing, due to the effect of the higher driving force. The presence of the second venturi promotes the re-atomization of liquid scrubbing solution when high air flowrates are used.

Observing the experimental pressure drops (Fig. 12), it is possible to conclude that the improvement on the removal efficiencies due to the use of two-stage venturi scrubbers is not broad enough to compensate for the associated increased power consumption. The use of this technique in higher gas flowrates is not recommended.

Work in progress in our laboratory aims to measure the absorption in the jet–venturi scrubber by varying the angle of dispersion of the pressure-swirl atomizer and the venturi throat diameter and length.

4. Conclusions

An easy-to-use empirical method was proposed to characterise the amount of gas pollutants removed in an industrial scale ejector–venturi scrubber. The empirical model was found to adequately represent gas absorption as reflected in the excellent correlation coefficients evaluated.

Experiments carried out in the present work showed an increase in absorption efficiency by using two-stage jet–venturi scrubbers, while removal levels lower than expected were observed when two-stage two-nozzle ones were used.

The use of two-stage venturi scrubbers is strongly limited by the increase in pressure drop, and thus, the associated energy consumption.

References

[1] H.F. Johnstone, R.B. Feild, M.C. Tassler, Ind. Eng. Chem. 46 (8) (1954) 1601.
[2] M.D. Kuznetsov, V.I. Oratovskii, Int. Chem. Eng. 2 (2) (1962) 85.
[3] Kh. Boyadzhiev, Int. Chem. Eng. 4 (1) (1964) 22.
[4] D. Elenkov, Kh. Boyadzhiev, Int. Chem. Eng. 7 (2) (1967) 191.
[5] B.P. Volgin, T.F. Efimova, M.S. Gofman, Int. Chem. Eng. 8 (1) (1968) 113.
[6] S. Uchida, C.Y. Wen, Ind. Eng. Chem. Process Des. Dev. 12 (4) (1973) 437.
[7] D.O. Cooney, Chem. Eng. Commun. 35 (1985) 315.
[8] M. Ravindram, N. Pyla, Ind. Eng. Chem. Process Des. Dev. 25 (1986) 35.
[9] J. Crank, The Mathematics of Diffusion, Oxford University Press, Oxford, 1997.
[10] I. Atay, G. Lewandowski, R. Trattner, Environ. Prog. 6 (3) (1987) 198.
[11] J.H. Hills, Ind. Eng. Chem. Res. 34 (12) (1995) 4254.
[12] M.R. Talaie, J. Fathikalajahi, M. Taheri, J. Air Waste Manage. Assoc. 47 (1997) 1211.
[13] B.J. Azzopardi, A.H. Govan, Filtr. Separat. 3 (1984) 196.
[14] B.J. Azzopardi, S.F.C.F. Texeira, A.H. Govan, T.R. Bott, Trans. IChemE B 69 (1991) 237.
[15] B.J. Azzopardi, Chem. Eng. J. 49 (1992) 55.
[16] B.J. Azzopardi, Chem. Eng. Sci. 48 (15) (1987) 2807.
[17] J.H. Hills, B.J. Azzopardi, A.S. Barhey, Trans. IChemE A 74 (1996) 567.
[18] K.D. Rappaport, N. Kettaneh, S. Wold, Am. Stat. 52 (2) (1998) 152.
[19] X. Gamisans, Internal Report (1998) EMRN-C2607-4.