

Wet Flue Gas Desulfurization Using a New O-Element Design Which Replaces the Venturi Scrubber

P. Lestinsky, D. Jecha, V. Brummer, P. Stehlik

Abstract—Scrubbing by a liquid spraying is one of the most effective processes used for removal of fine particles and soluble gas pollutants (such as SO₂, HCl, HF) from the flue gas. There are many configurations of scrubbers designed to provide contact between the liquid and gas stream for effectively capturing particles or soluble gas pollutants, such as spray plates, packed bed towers, jet scrubbers, cyclones, vortex and venturi scrubbers. The primary function of venturi scrubber is the capture of fine particles as well as HCl, HF or SO₂ removal with effect of the flue gas temperature decrease before input to the absorption column. In this paper, sulfur dioxide (SO₂) from flue gas was captured using new design replacing venturi scrubber (1st degree of wet scrubbing). The flue gas was prepared by the combustion of the carbon disulfide solution in toluene (1:1 vol.) in the flame in the reactor. Such prepared flue gas with temperature around 150°C was processed in designed laboratory O-element scrubber. Water was used as absorbent liquid. The efficiency of SO₂ removal, pressure drop and temperature drop were measured on our experimental device. The dependence of these variables on liquid-gas ratio was observed. The average temperature drop was in the range from 150°C to 40°C. The pressure drop was increased with increasing of a liquid-gas ratio, but not too much as for the common venturi scrubber designs. The efficiency of SO₂ removal was up to 70 %. The pressure drop of our new designed wet scrubber is similar to commonly used venturi scrubbers; nevertheless the influence of amount of the liquid on pressure drop is not so significant.

Keywords—Desulphurization, absorption, flue gas, modeling.

I. INTRODUCTION

IN recent years, progress in the field of heterogeneous catalysts, especially catalytic filtration, has led to the use dry of flue gas cleaning in municipal waste incineration. The SO₂ is usually removed using calcium carbonate or limestone (dry flue gas desulphurization) and the product of this reaction is filtrated on catalytic filters. The problem of dry flue gas cleaning (with catalytic filtration) is that the removal of each pollutant (i.e. NO_x, VOC, SO₂ and dioxins) has its optimal removal efficiency at different ranges of temperature. Therefore, it is appropriate in technological units to place a wet scrubber after the unit of catalytic filtration. When emission limits are exceeded after a unit of catalytic filtration, flue gas is subsequently treated to the required level in the wet scrubber. The primary aim of the venturi scrubber is to remove fine dust particles. When catalytic filtration is used, the

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removal of dust particles using venturi scrubber is not necessary. However, the temperature of the flue gas is still high for treatment in the absorption column. The O-element is a new design replacing the venturi scrubber and is suitable for flue gas cleaning at high temperatures (100-300°C).

The O-element works as a venturi scrubber and also as a static mixer. This equipment can be used not only as the 1st stage of a wet scrubber, but also for secondary flue gas treatment after the catalytic filtration is used.

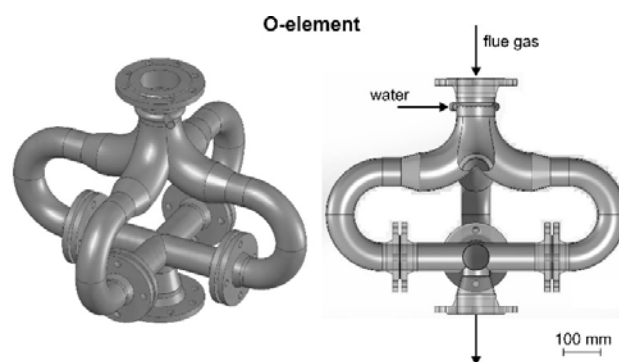


Fig. 1 The new design for the 1st stage of the wet scrubber - O-element

Emission limits for SO₂ are decreasing all over the world [1], [2]. Research and development into the technology for desulphurization focus on improving the removal efficiency and minimizing consumptions of energy and water [3]. In this paper, SO₂ removal is evaluated using the wet scrubber new design i.e. the O-element (wet technology). A 1D separated flow model of the external mass-transfer has been developed on both the gas and liquid sides. This was with the effects of a sulfite concentration in the liquid phase to estimate the SO₂'s removal efficiency.

II. MODEL

In the wet flue gas desulfurization process, SO₂ diffuses through the gas phase to the liquid surface, where it dissolves and is transferred by diffusion or convective mixing into the liquid phase. The rate at which SO₂ is transferred depends on a number of factors, such as the solubility of SO₂ in the liquid, temperature and mixing in the fluid flow. A number of models have been proposed to describe this transfer across a phase boundary. The two resistance theory described the transfer as being confined to two thin stagnant films on either side of the

gas-liquid interface. This model assumes that all the resistance to mass-transfer is contained in the two films and none at the interface.

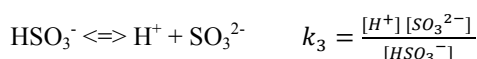
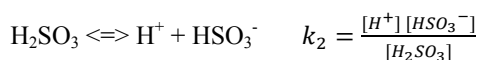
The interface concentrations (c_i) are in equilibrium and can be determined from the following relationship:

$$c_{SO_2} = H_{SO_2} p_{SO_2} \quad (1)$$

where p_A is the partial pressure of SO_2 on the interface phases, H is the thermodynamic equilibrium constant, whose value can be obtained by [4]:

$$H_{SO_2} = \exp\left(\frac{32143.3}{T} + 198.14 \ln T - 0.3384 T - 1135.62\right) \quad (2)$$

The SO_2 is initially absorbed into the liquid and converted into H_2SO_3 (sulfurous acid). A succession of equations between the vapor pressure of SO_2 and the concentration of sulfite in the liquid phase can be considered [5]:



The equilibrium constants are $k_2 = 1.32 \cdot 10^{-2} \text{ kmol/m}^3$ and $k_3 = 6.41 \cdot 10^{-8} \text{ kmol/m}^3$.

The total sulfur dioxide concentration (c_{SO_2}) in the liquid phase can be represented as:

$$c_{SO_2} = [H_2SO_3] + [HSO_3^-] + [SO_3^{2-}] \quad (3)$$

The SO_2 concentration (c_A) in the liquid is affected by $[H^+]$ and the partial pressure of SO_2 in the gas phase. The Henry constant can be estimated at a constant pH value of the liquid phase [6]:

$$H_{SO_2}^* = H_{SO_2} \left(1 + \frac{k_2}{[H^+]} + \frac{k_2 k_3}{[H^+]^2}\right) \quad (4)$$

where $[H^+]$ can be calculated from the pH of the solution:

$$[H^+] = 10^{-pH} \quad (5)$$

The theoretical prediction of SO_2 removal is based on the following simplifying assumptions [7], [8]:

- gaseous reactants have low solubility in the liquid;
- the reaction between the two species is instantaneous and irreversible;
- the influence of the product species is negligible;
- there are no resistances to particle dissolution and interface of gas-liquid phase;
- the droplet is spherical;
- the heat of reaction and dissolution can be neglected.

The absorption rate can be calculated through the interfacial surface for the retention time of flue gas in the O-element as:

$$\frac{dN_{SO_2, film}}{dL} = K_{G, film} S_{L, film} t_G \quad (6)$$

$$\frac{dN_{SO_2, drop}}{dL} = K_{G, drop} S_{L, drop} t_G \quad (7)$$

The resistance of mass transfer between the phases is concentrated in the gas phase. The overall mass transfer coefficient is calculated as the sum of mass transfer resistances:

$$K_{G, film} = \frac{1}{\frac{1}{k_{G, film}} + \frac{1}{H_{SO_2, film}^* k_{L, film}}} (p_{SO_2} - p_{SO_2, film}) \quad (8)$$

$$K_{G, drop} = \frac{1}{\frac{1}{k_{G, drop}} + \frac{1}{H_{SO_2, drop}^* k_{L, drop}}} (p_{SO_2} - p_{SO_2, drop}) \quad (9)$$

The balance of SO_2 in the flue gas dL is calculated after the elementary step as:

$$N_{SO_2} = (n_G t_G y_{SO_2}) - N_{SO_2, film} - N_{SO_2, drop} \quad (10)$$

The physical-chemical properties of the gas and liquid are calculated by their temperature and chemical composition at the each step dL .

B. Slip Ratio for Liquid Film and Entrainment Droplets

The next important parameter for calculating mass transfer is knowledge of the behavior of the two-phase flow in the Q-element equipment. The gas superficial velocity (u_G) and slip ratio (SR) are important for the determination of liquid velocity (u_L). Slip ratio is the ratio between gas and liquid velocity (u_G / u_L). The slip ratio for the liquid film in the annular two-phase flow can be calculated as [9]:

$$SR_{film} = (1 - X (1 - \frac{\rho_L}{\rho_G}))^{0.5} \quad (11)$$

The slip ratio for entrainment droplets can be calculated as [10]:

$$SR_{drop} = \frac{\rho_L X (1 - \varepsilon)}{\rho_G (1 - X) \varepsilon} \quad (12)$$

where X is mass fraction and ε is volume fraction of the gas phase in the two-phase flow.

Fore and Dukler [11] measured the diameter of entrainment droplets at the annular two-phase flow in a vertical tube. The Sauter mean diameter of entrainment droplets was 400 μm for the conditions in our experiment ($u_g = 10\text{-}30 \text{ m/s}$, $Re_L = 100\text{-}3000$).

Sawant et al. [12] measured the amount of entrainment droplets at the annular two-phase flow in the vertical tube. For the air-water system, the following equation can be used:

$$E = 9 \cdot 10^{-6} We_{modif}^{1.25} \quad (13)$$

where the modified Weber number is calculated as:

$$We_{modif} = \frac{\rho_G u_G^2 d}{\sigma (\frac{\rho_L - \rho_G}{\rho_G})^{1.4}} \quad (14)$$

The mass transfer coefficients are calculated using data of the flow rate and physical-chemical parameters as written below.

C. Mass Transfer in the Liquid Falling Film

The liquid mass flow along wall:

$$\Gamma_L = \frac{m_{L, film}}{\pi d} \quad (15)$$

The liquid velocity of the falling film is affected by flowing gas (slip ratio):

$$u_{L, film} = \frac{u_G}{SR_{film}} \quad (16)$$

The thickness of the falling liquid film on the wall of tube:

$$dL = \frac{\Gamma_L}{\rho_L u_{L, film}} \quad (17)$$

The calculation of the Sherwood number is dependent on Reynolds number [13]:

$$Re_L > 1600 \quad Sh_L = 7.7 \cdot 10^{-5} Re_L Sc_L^{0.5} \quad (18)$$

$$Re_L < 300 \quad Sh_L = 0.888 Re_L^{0.45} Sc_L^{0.5} Ga_L^{-0.166} \quad (19)$$

$$300 < Re_L < 1600 \quad pp = \frac{3.2 - \log Re_L}{1.47} \quad (20)$$

$$Sh_L = 1.21 \cdot 10^6 \cdot 0.909^{pp} Re_L^{\frac{pp}{3} - 2.18} Sc_L^{0.5} Ga_L^{-\frac{pp}{3}} \quad (21)$$

where there is a Reynolds number, the Schmidt number and Galileo number are calculated as:

$$Re_L = \frac{u_{L, film} dL \rho_L}{\eta_L} \quad (22)$$

$$Sc_L = \frac{\eta_L}{\rho_L D_L} \quad (23)$$

$$Ga_L = \frac{g \rho_L^2 dL}{\eta_L^2} \quad (24)$$

The mass transfer coefficient in the falling liquid film:

$$k_{L, film} = \frac{Sh_L D_L}{dL} \quad (25)$$

The Sherwood number for gas phase flowing along the falling liquid film [14]:

$$Sh_G = 0.023 Re_G^{0.83} Sc_G^{0.44} \quad (26)$$

where Reynolds number and Schmidt number for gas phase are calculated as:

$$Re_G = \frac{u_G d_G \rho_G}{\eta_G} \quad (27)$$

$$Sc_G = \frac{\eta_G}{\rho_G D_G} \quad (28)$$

The mass transfer coefficient in the gas phase flowing along the falling liquid film:

$$k_{G, film} = \frac{Sh_G D_G R T_G}{d} \quad (29)$$

D. Mass Transfer in the Liquid Droplet

The Sherwood number for mass transfer in the sphere [15]:

$$Sh_L = 0.992 Re_L^{0.33} Sc_L^{0.33} \quad (30)$$

where Reynolds number and Schmidt number are calculated as:

$$Re_L = \frac{u_{L, drop} d_p \rho_L}{\eta_L} \quad (31)$$

$$Sc_L = \frac{\eta_L}{\rho_L D_L} \quad (32)$$

The mass transfer coefficient for liquid droplet:

$$k_{L, drop} = \frac{Sh_L D_L}{d_p} \quad (33)$$

The Sherwood number for mass transfer in the gas phase around liquid droplet can be calculated by the Frossling correlation [16]:

$$Sh_G = 2 + 0.6 Re_L^{0.5} Sc_G^{0.33} \quad (34)$$

where Reynolds number and Schmidt number are calculated as:

$$Re_G = \frac{(u_G - u_L) d_p \rho_G}{\eta_G} \quad (35)$$

$$Sc_G = \frac{\eta_G}{\rho_G D_G} \quad (36)$$

The mass transfer coefficient in gas phase flowing around sphere particle (droplet):

$$k_{G, drop} = \frac{Sh_G D_G R T_G}{d_p} \quad (37)$$

III. EXPERIMENTAL PART

The flue gas was prepared by combustion of the carbon disulfide dissolved in toluene, in a proportion volume of 1:1 (vol.). The solution was then injected (800 cm³/h) with air, which was blown using a fan into the reactor with the burner. The reactor contains heat recovery ventilation (heat recuperation) for easier heating of flue gas with a small consumption of natural gas. This SO₂ polluted flue gas, with a temperature of around 150°C, was transported from the reactor to the O-element using under pressure created by the fan at the end of the pipeline. The concentration of SO₂ was from 360 to 460 ppm on the inlet.

The flue gas enters the O-element through a tube with a diameter of DN 100. The water was sprayed in the place

where flue gas enters the O-element (see Fig. 1). Then, the flue gas is divided into four tubes with a diameter of DN 70. All of four flows are mixed into the one. The incurred two-phase mixed flow leaves the O-element in a tube with a diameter of DN 100 into the tank.

As absorption liquid was used fresh water. The waste water from the tank is flushed away into the sewerage. Two gas volumetric flow rates (V_G) were used 500 and 600 Nm³/h and liquid volumetric flow rates (V_L) were used from 0.05 to 1 Nm³/h.

A simplified layout of experimental devices is shown in Fig. 2 with measurements and gauges. The flue gas composition was measured with the flue gas analyzer, ABB EL 3020 (infrared photometer). The experiment took 30 min. The pressure, temperature and flow rate were constant for this time. The concentrations of SO₂ from the analyzer were recorded every second. The values of the inlet/outlet concentration were calculated as median.

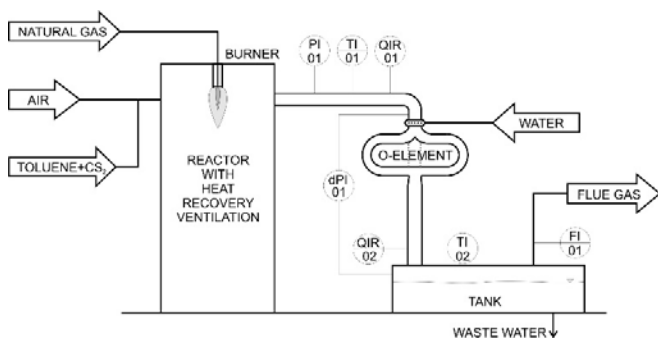


Fig. 2 The layout of experimental devices (dP – pressure drop, P – relative pressure, T – temperature, F – flow rate, Q – flue gas composition)

IV. RESULTS AND DISCUSSION

The enthalpy balance of cold water and hot flue gas's mixing was performed to determine the flue gas's outlet temperature. The temperature difference between the outlet gas and liquid was supposed to be 1°C. The results of the enthalpy balance were compared with the temperatures measured. The maximum temperature error of output flue gas was only 2°C in the prediction (see Fig. 3).

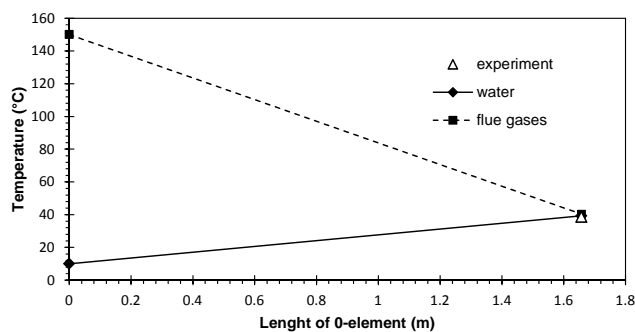


Fig. 3 A temperature linear trend for water and flue gases in the O-element in the conditions: $V_G = 500 \text{ Nm}^3/\text{h}$ and $V_L = 0.5 \text{ Nm}^3/\text{h}$

The temperature difference between the inlet and outlet was predicted using a linear trend according to the length of the absorption unit (O-element). This linear trend was used to calculate the physical-chemical properties in the O-element.

The pH profile is changed along the complete length of the O-element as can be seen in Fig. 4. As the SO₂ concentration increased in the liquid, the values of pH decreased. The concentrations of SO₂ in the liquid film and in the droplets were averaged in the place where all four flows are mixed to the one.

The maximum efficiency to remove SO₂ was 70% for a liquid/gas ratio of 0.002 (see Fig. 5). The maximum absolute error between the mathematic model and the experiment was 25%. When the lower flows of water were used, the evaporation of water from the droplets was bigger. 15% of mass was evaporated from the droplets to the flue gas when the water flow rate was 0.05 m³/h. Such a level of evaporation caused an increase in mass transfer resistance through the interfacial area and is the explanation for the experimental efficiency of desulphurization was smaller than in the mathematic model.

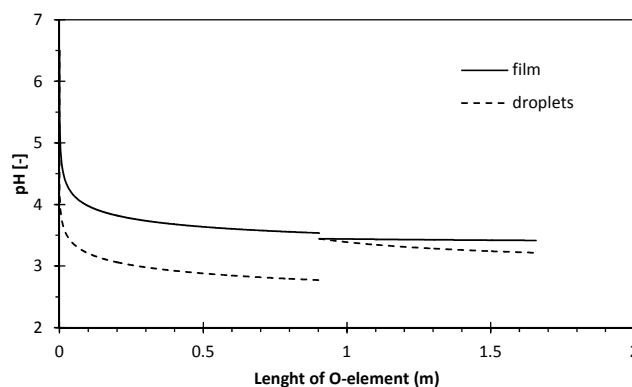


Fig. 4 The change of pH in the O-element in the conditions: $V_G = 500 \text{ Nm}^3/\text{h}$ and $V_L = 0.5 \text{ Nm}^3/\text{h}$

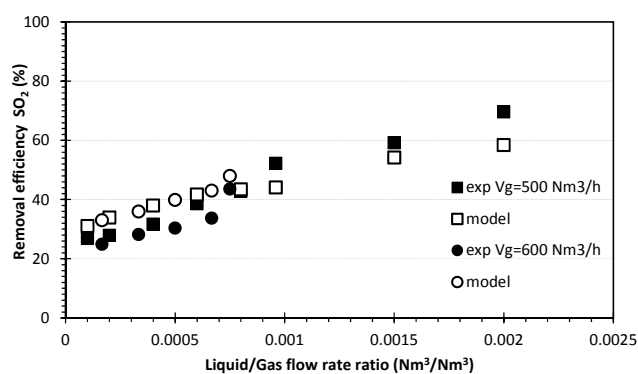


Fig. 5 The dependence of SO₂ removal efficiency on liquid/gas volumetric flow ratio

V. CONCLUSION

The mathematical mass transfer model was used to predict the behavior of a wet scrubber named the O-element. The results of the mathematic model were compared with an

experimental desulphurization process on this equipment. The maximum efficiency to remove SO₂ was 70% for a liquid/gas ratio of 0.002. The pressure drop was 3.5 kPa in the condition used. This equipment is appropriate for a 1st stage wet scrubber.

The temperature of the flue gas outlet was predicted using enthalpy balance with heat loss at an accuracy of 95%. The maximum absolute error for predicting the removal efficiency of SO₂ was 25 %. However, the resistance of mass transfer was not included in the mass transfer model due to evaporation from the interface surface. Other gases, such as CO and CO₂, were absorbed into the liquid but in smaller quantities than the SO₂ (the removal efficiency of CO was up to 15% and CO₂ up to 1%). The absorption of these gases was not modeled although it could reduce the absorption capacity of the liquid.

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