

# **AN ENGINEERING ANALYSIS OF CAPACITY IMPROVEMENT IN FLUE GAS DESULFURIZATION PLANT**

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## **ABSTRACT**

The overall process of desulfurization was divided into three partial processes (reactions). The mass balances of the partial processes of the industrial spray scrubbing process were presented in the equilibrium diagram for the  $\text{Ca}^{2+}$ - $\text{SO}_2$ - $\text{H}_2\text{O}$  system.

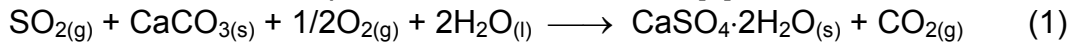
The mass transfer analysis of the partial processes was made and its results were in good agreement with the results based on the overall mass balance equation and the overall reaction.

Cases considering changes in input process parameters were also studied. A modification of some process parameters had to be made to satisfy the proper operation of the absorption unit.

## INTRODUCTION

The sulfur dioxide emissions from power plants and other sources are harmful to the environment and to humans as well. Large amounts of sulfur dioxide are released into the air by combustion of fossil fuels for power generation. The most common flue gas desulfurization technology is the absorption of SO<sub>2</sub> in a limestone slurry [1].

The overall reaction for this process can be written as [2]:



There has been considerable work done in the field of SO<sub>2</sub> absorption. The equilibrium diagram for the MgO–SO<sub>2</sub>–H<sub>2</sub>O system showing the concentrations of total SO<sub>2</sub> (c<sub>tot</sub>), of combined SO<sub>2</sub> (c<sub>com</sub>), of true free SO<sub>2</sub> (Y) and the pH value of the solution was constructed. A solubility curve for MgSO<sub>3</sub> was also presented [3]. An analogous model was applied to the Ca<sup>2+</sup>–SO<sub>2</sub>–H<sub>2</sub>O system, which enabled the construction of the equilibrium diagram for this system as well [2].

Some studies divide the desulfurization process into partial processes (reactions) [1, 4], the same approach was used in our previous work [2]. On the assumption that mass transfer is diffusion controlled a mass transfer analysis of the partial processes was also made.

The aim of the present work is to study the operation of the industrial spray scrubber under normal operating conditions and if the input conditions (parameters) are changed. In the latter case a possible change in some process parameters had to be considered in order to satisfy the mass balance of the scrubber.

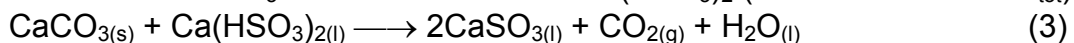
## PRESENTATION OF THE PARTIAL PROCESSES

The overall reaction (1) can be divided into three partial processes (reaction steps) [2];

- absorption of SO<sub>2</sub>, sulfite – bisulfite reaction (occurs at constant c<sub>com</sub>)



- dissolution of CaCO<sub>3</sub> and its reaction with Ca(HSO<sub>3</sub>)<sub>2</sub> (occurs at constant c<sub>tot</sub>)



- oxidation of CaSO<sub>3</sub> and crystallization into gypsum (occurs at constant Y)



The division into partial processes is an idealized assumption, they are also assumed to take place in series. These three partial processes are presented as a triangle in the equilibrium diagram (Figure 1).

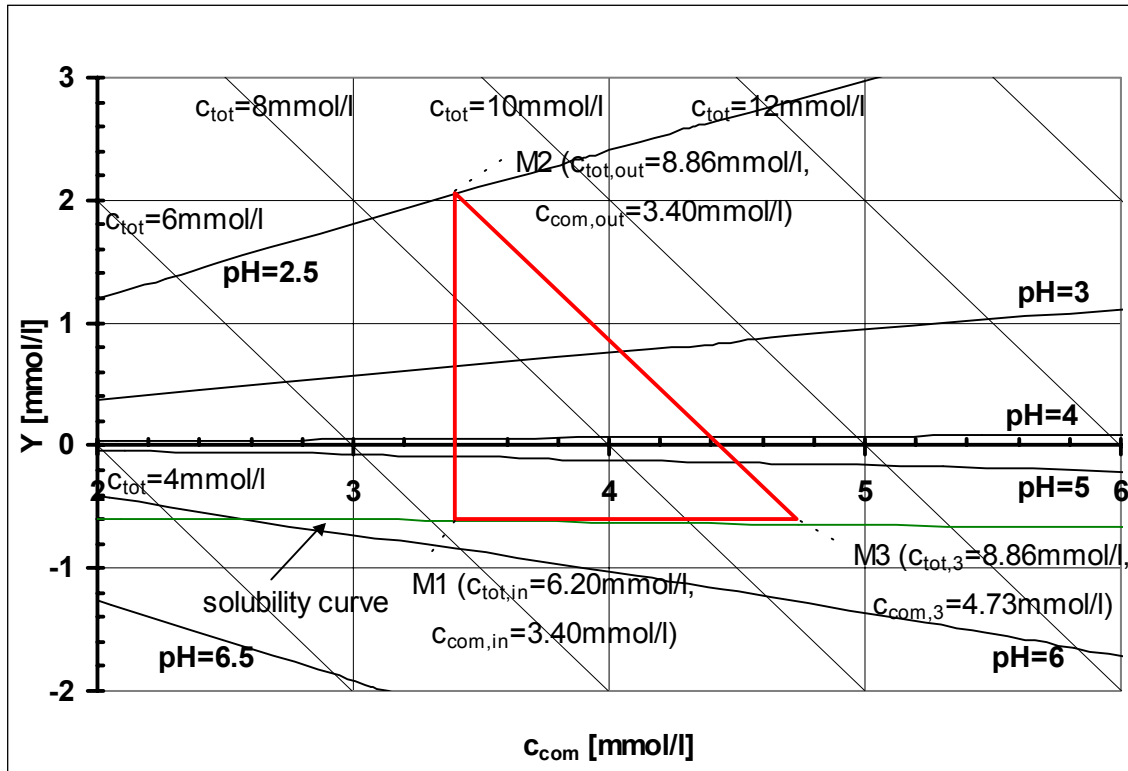


Figure 1: Idealized presentation of the partial processes of the spray scrubbing process in the equilibrium diagram (at 25°C)

$M_1M_2$  – absorption of  $SO_2$ , sulfite – bisulfite reaction

$M_2M_3$  – limestone dissolution, reaction with  $Ca(HSO_3)_2$

$M_3M_1$  – absorption of  $O_2$ , oxidation of  $CaSO_3$ , gypsum crystallization

The process data for the triangle points M1, M2 and M3 are taken from the industrial spray scrubbing plant (Figure 2), which removes  $SO_2$  from the flue gas from the 275MW power plant in Sostanj, Slovenia, using coal powder as the fuel. The flue gas enters the absorption unit, the limestone slurry is distributed in six levels, collected in the holding tank and recycled to the sprayers. Air is blown into the holding tank to ensure the oxidation of  $CaSO_3$  into sulfate. Calcium sulfate leaves the process as gypsum, which is separated from the recycle flow in hydrocyclones.

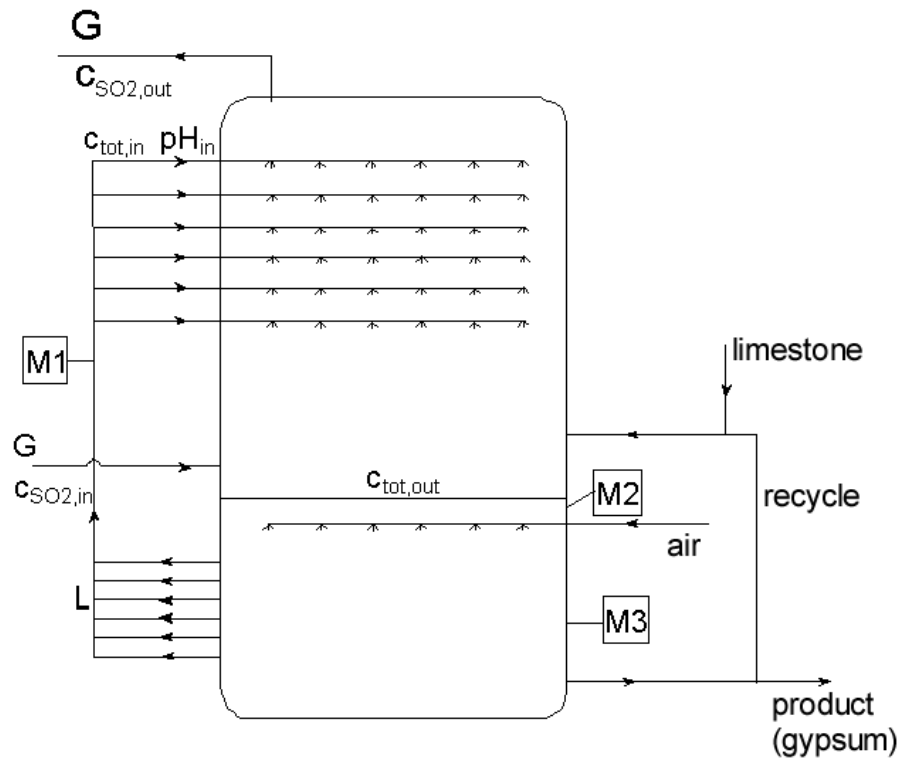


Figure 2: Schematic flow diagram of the industrial spray scrubbing plant

## MASS TRANSFER CALCULATIONS

The mass transfer analysis of the three processes was based on the industrial process data (Table 1), using the assumption that mass transfer is the rate determining process (the mass transfer is diffusion controlled).

Table 1: Basic process and triangle points data

Process data			
$G=1.1 \cdot 10^6 \text{ m}^3/\text{h}$	$c_{\text{SO}_2,\text{in}} = 0.125 \text{ mmol/l}$	$c_{\text{limestone}} = 6 \text{ g/l}$	
$L=5 \cdot 10^4 \text{ m}^3/\text{h}$	$c_{\text{SO}_2,\text{out}} = 0.0041 \text{ mmol/l}$	$G_{\text{oxid}} = 1.7 \cdot 10^4 \text{ m}^3/\text{h}$	
Triangle points			
	measured	calculated	
M1	$c_{\text{tot},\text{in}} = 6.20 \text{ mmol/l}$ , $\text{pH}_{\text{in}} = 5.8$	$c_{\text{com}} = 3.40 \text{ mmol/l}$	
M2	/	$c_{\text{tot},\text{out}} = 8.86 \text{ mmol/l}$ , $\text{pH}_{\text{out}} = 2.5$ , $c_{\text{com}} = 3.40 \text{ mmol/l}$	
M3	/	$c_{\text{tot},3} = 8.86 \text{ mmol/l}$ , $\text{pH}_3 = 5.6$ , $c_{\text{com},3} = 4.73 \text{ mmol/l}$	

All three partial processes were modeled by the equation

$$\dot{W} = K \cdot A \cdot \Delta c \quad (5)$$

The data for the calculation were taken from the industrial plant, some data and the appropriate correlations were taken from the up to date literature. The results of the calculation of the overall mass transfer coefficient  $K$ , the area  $A$  available for the mass transfer, the concentration potential  $\Delta c$  and the molar flow rate  $\dot{W}$  for the three partial processes are presented in Table 2.

Table 2: The results of the mass transfer analysis for the three partial processes

	K (m/s)	A (m <sup>2</sup> )	$\Delta c$ (mol/m <sup>3</sup> )	$\dot{W}$ (kmol/h)
absorption of SO <sub>2</sub>	$K'_g = 2.80 * 10^{-2}$	$A_{SO_2} = 6.67 * 10^4$	$\Delta c_{SO_2(ln)} = 2.02 * 10^{-2}$	$\dot{W}_{SO_2} = 136$
dissolution of CaCO <sub>3</sub>	$k_s = 2.95 * 10^{-3}$	$A_{CaCO_3} = 4.73 * 10^6$	$\Delta c_{dissol} = 2.51 * 10^{-3}$	$\dot{W}_{CaCO_3} = 126$
absorption of O <sub>2</sub>	$k_{L(O_2)} = 3.81 * 10^{-4}$	$A_{O_2} = 1.96 * 10^5$	$\Delta c_{O_2(ln)} = 2.61 * 10^{-1}$	$\dot{W}_{O_2} = 70$

The results of the mass transfer analysis were verified by the basic mass balance

$$G(c_{SO_2,in} - c_{SO_2,out}) = L(c_{tot,out} - c_{tot,in}) \quad (6)$$

and the overall reaction (1), and the results were within a 10% margin, which confirms that the calculation procedure is satisfactory.

## VARIATIONS IN INDUSTRIAL PLANT OPERATION

The industrial plant should be able to operate at changed input conditions and still ensure the same SO<sub>2</sub> removal. Therefore the following cases were studied;

Case 1: The increase in the input gas flow rate and in the input SO<sub>2</sub> concentrations.

The input gas flow rate and the input gas SO<sub>2</sub> concentration were increased by 20%. This case illustrates the increase in power plant operation. The spray scrubber can ensure the demanded SO<sub>2</sub> removal if some process parameters ( $c_{limestone}$ ,  $G_{oxid}$ ,  $L$ ) are reset. The process data are listed in Table 3.

Case 2: The increase in the input SO<sub>2</sub> concentration, whereas the input gas flow rate remains unchanged

The increase in gas SO<sub>2</sub> concentration was 20%. Such a case would happen if a different kind of coal with larger sulfur content was used in the power plant. Again the spray scrubber met the demands if the relevant process parameters ( $c_{limestone}$ ,  $G_{oxid}$ ) had been changed (Table 3).

Case 3: Use of a limestone with a different particle size distribution

In this case the input process parameters remained the same, however the limestone concentration had to be smaller (Table 3) since this limestone has much smaller particles which dissolve faster. The  $d_{90}$  of this limestone is 31.2 $\mu$ m compared to the  $d_{90}$ =63 $\mu$ m for the primary limestone.

The suspension flow rate, limestone concentration, and air flow rate, respectively, had to be modified in order to satisfy the new condition of the overall mass balance of the absorption process. As for the mass transfer: First for the partial process of the absorption of SO<sub>2</sub> this is ensured by increasing the interfacial area and the concentration potential, for the partial process of limestone dissolution the adequate interfacial area is achieved by a greater (cases 1 and 2) or smaller (case 3) limestone concentration. In case 3 the limestone particles are smaller which also increases interfacial area. The interfacial area for the partial process of absorption of O<sub>2</sub> is enlarged through a larger air flow rate. The capacity of the mechanical equipment is oversized to higher capacity conditions, thus no special rearrangements of pumps, compressors and hydrocyclones is necessary.

Table 3: Basic process data – cases 1–3

case 1	$G=1.32 \cdot 10^6 \text{ m}^3/\text{h}$	$c_{\text{SO}_2, \text{in}} = 0.150 \text{ mmol/l}$	$c_{\text{limestone}} = 8.5 \text{ g/l}$
	$L=6 \cdot 10^4 \text{ m}^3/\text{h}$	$c_{\text{SO}_2, \text{out}} = 0.0041 \text{ mmol/l}$	$G_{\text{oxid}} = 2.2 \cdot 10^4 \text{ m}^3/\text{h}$
case 2	$G=1.1 \cdot 10^6 \text{ m}^3/\text{h}$	$c_{\text{SO}_2, \text{in}} = 0.150 \text{ mmol/l}$	$c_{\text{limestone}} = 7 \text{ g/l}$
	$L=5 \cdot 10^4 \text{ m}^3/\text{h}$	$c_{\text{SO}_2, \text{out}} = 0.0041 \text{ mmol/l}$	$G_{\text{oxid}} = 1.9 \cdot 10^4 \text{ m}^3/\text{h}$
case 3	$G=1.1 \cdot 10^6 \text{ m}^3/\text{h}$	$c_{\text{SO}_2, \text{in}} = 0.125 \text{ mmol/l}$	$c_{\text{limestone}} = 1.5 \text{ g/l}$
	$L=5 \cdot 10^4 \text{ m}^3/\text{h}$	$c_{\text{SO}_2, \text{out}} = 0.0041 \text{ mmol/l}$	$G_{\text{oxid}} = 1.7 \cdot 10^4 \text{ m}^3/\text{h}$

The triangles representing the partial processes for the case 1 and case 2 can be presented in the equilibrium diagram (Figure 3). For both cases the measured point M1 has the pH=5,8. For the case 3 the presentation of the partial processes in the equilibrium diagram would be the same as for the normal operating conditions.

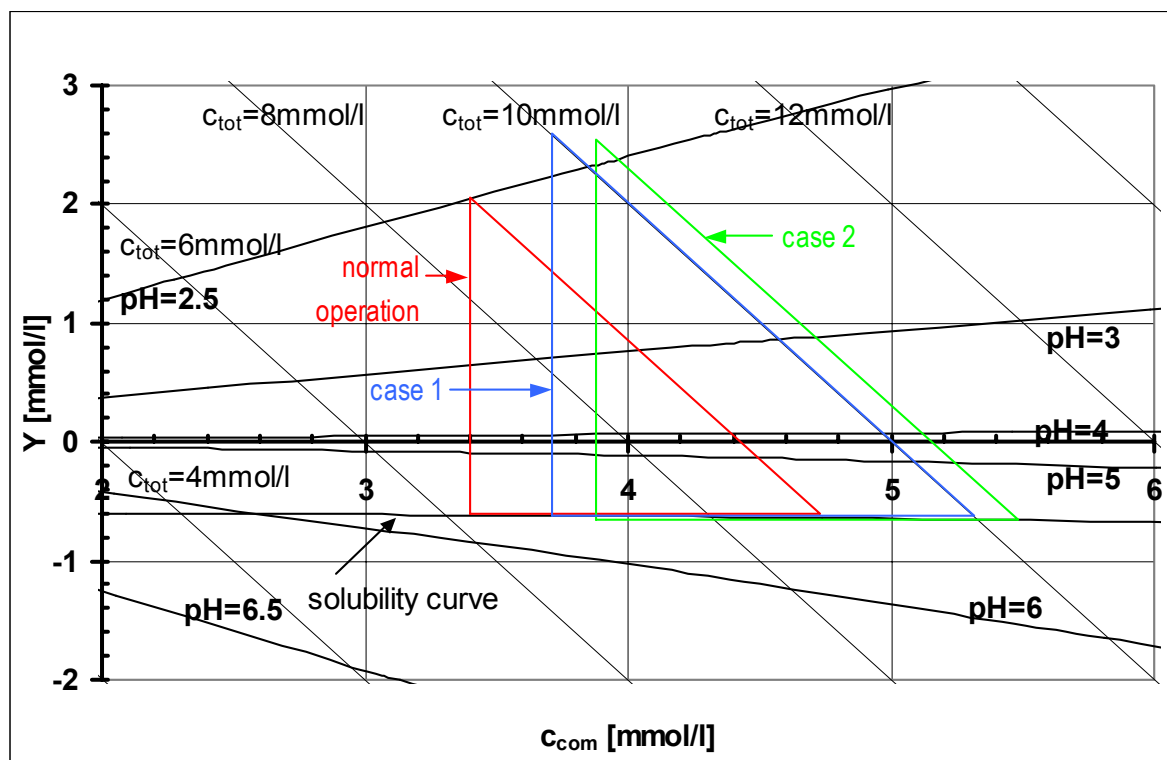


Figure 3: The presentation of the partial processes for the case 1 and case 2 in the equilibrium diagram (at 25°C). Also presented is the triangle for the normal spray scrubber operation.

## CONCLUSIONS

The process of the industrial desulfurization spray scrubbing process was divided into partial processes – absorption of  $\text{SO}_2$  and sulfite to bisulfite reaction, limestone dissolution and its reaction with  $\text{Ca}(\text{HSO}_3)_2$ , absorption of  $\text{O}_2$ , oxidation of  $\text{CaSO}_3$  and gypsum crystallization. Their mass balances were presented in the equilibrium diagram for the  $\text{Ca}^{2+}\text{--SO}_2\text{--H}_2\text{O}$  system.

The values obtained by the mass transfer analysis of the partial processes were in good agreement with the values calculated by the basic mass balance and the overall reaction.

The operation of the spray scrubber under changed operating conditions was also studied. The proper process parameters had to be changed in order to satisfy the mass balance and the mass transfer of the process and to ensure the demanded  $\text{SO}_2$  removal.

## NOMENCLATURE

c	concentration, mmol/l
$c_{\text{limestone}}$	steady-state limestone concentration, g/l
A	interfacial area, $\text{m}^2$
$\Delta c$	concentration potential, $\text{mol}/\text{m}^3$
G	flue gas flow rate, $\text{m}^3/\text{h}$
$G_{\text{oxid}}$	air flow rate for absorption of $\text{O}_2$ , $\text{m}^3/\text{h}$
K	overall mass transfer coefficient, m/s
$K'_g$	overall mass transfer coefficient for absorption of $\text{SO}_2$ , m/s
$k_{L(\text{O}_2)}$	liquid-side mass transfer coefficient for absorption of $\text{O}_2$ , m/s
$k_s$	liquid-side mass transfer coefficient for dissolution of $\text{CaCO}_3$ , m/s
L	suspension flow rate, $\text{m}^3/\text{h}$
$\dot{W}$	molar flow rate, kmol/h
Y	concentration of true free $\text{SO}_2$ , mmol/l

### *subscripts*

com	combined $\text{SO}_2$
dissol	dissolution
tot	total $\text{SO}_2$

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keywords:

absorption of SO<sub>2</sub>, flue gas, desulfurization, limestone, spray scrubbing