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Modelling of trona based spray dry scrubbing of SO_2

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Abstract

The paper presents a model of the reaction between SO_2 and trona based slurries. Experimental findings relevant to the desulfurisation in a spray dryer were compared with the proposed mathematical model and were found in good agreement. It is observed that at high stoichiometric ratios, the diffusion of SO_2 from the gas bulk to the droplet interface is dominant for absorption and with decreasing stoichiometric ratio the absorption efficiency decreases. An increase in the temperature also causes a decrease in absorption efficiency. At the stoichiometric ratio (Na/S = 2), SO_2 removal efficiency was found to be around 90%.

Keywords: Trona; Desulfurisation; Spray dryer; Two-phase flow; Mathematical modelling

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1. Introduction

Spray drying produces particles by atomizing a solution or slurry and evaporating moisture from the resulting droplets by suspending them in a hot gas. The production of dry, spherical particles from a liquid feed in a single processing step makes spray drying a unique and important unit operation. Industrial and utility interest in the spray dry scrubbing concept for the control of SO_2 emissions has dramatically increased during past decade. The sorption efficiency of a spray dryer scrubber is generally considered to be quite high and these systems have much less operational difficulties as compared to the conventional wet systems. Limestone or lime slurry are the two varieties of alkaline slurry most frequently used in absorbers to remove SO_2 from flue gas [1]. Kaplan and Felsvang reported some data for the scrubbing of SO_2 in a spray dryer using lime as the reactive sorbent [2]. It is also reported that the sorption efficiency increased considerably with an increase in relative humidity [3]. Comparison of reactivities of various carbonates as absorbents of SO_2 was given in the review of Hartman [4]. Kimura and Smith reported that the rate of SO_2–Na_2CO_3 reaction was one or two orders of magnitude greater than the rate of limestone–SO_2 reaction [5]. Another important advantage of activated soda for SO_2 removal is its higher conversion than the conversion of CaO to $CaSO_4$. The conversion level of Na_2CO_3 to Na_2SO_3 and Na_2SO_4 approached to unity, while the maximum conversion of CaO to $CaSO_4$ is generally less than 0.4 in a dry system [5,6]. Doğu et al. reported that one of the major advantages of SO_2 removal by natural soda (trona) ore in the spray dryer is that the capital cost is about 25% less than of a limestone scrubbing system [7].

In that context, an alternative sorbent utilization, like as trona, can also be used to make the spray dry scrubbing process competitive against the established wet processes.

Recently, there has been an increased interest in the modelling of the spray dry scrubbing process [8–10]. Although the overall reaction chemistry of a natural soda based system is quite simple, the simultaneous mass and heat transfer is considerably complex. The reaction in the spray dryer involves several drying phases and, consequently, reaction conditions, which have to be approached differently. Furthermore, the method of gas–liquid contacting in the spray dryer must be taken into consideration. Taking as a basis the character of equations describing the process of spray drying in mathematical models presented in the literature [11], a classification of these models may be given.

The first group contains those models whose equations are so constructed that an analytical solution can be obtained. Such models are solved only for one particle diameter and it is assumed that the particle temperature is the temperature of adiabatic saturation during the whole drying process [11]. Models of this group allow the basic parameters of flue gas during drying

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Nomenclature

A	area (m^2)
$C_{Na_2CO_3}$	conversion efficiency of trona
C_p	heat capacity (J/(kg K))
C_{SO_2}	SO_2 concentration (kmol/m^3)
C_T	total gas concentration (kmol/m^3)
d	particle diameter (m)
D	diffusion coefficient (m^2/s)
h	heat transfer coefficient (W/(m^2 K))
H	humidity (kg/kg)
k	thermal conductivity (W/(m K))
Le	Lewis number
m_p	mass of a spherical particle (kg)
M	mass (kg)
M_s	mass of solids (kg/kmol)
N	mole flux (kmol/(m^2 s))
Nu	Nusselt number
Pr	Prandtl number
Q	heat (J)
Re	Reynolds number
S	unreacted soda fraction
Sc	Schmidt number
Sh	Sherwood number
SR	stoichiometric ratio
t	time (s)
T	temperature (K)
W	water content of droplet (kg water/kg solids)
x	residual water fraction in droplet
y	SO_2 mole fraction

Greek symbols

β	constant
Δ	gas bulk temperature – droplet surface temperature
η	SO_2 removal efficiency
λ	latent heat (J/kg)
ρ	density (kg/m^3)
τ	dimensionless temperature

Subscripts

0	initial
2	difference between inlet and outlet temperatures
a	gas bulk
i	inlet
o	outlet
s	droplet interface
w	water

to be determined but the number of simplifying assumptions is so large that the results are not fully reliable.

The second group encompasses the models in which the character of equations is similar to that in the first group, but the number of simplifying assumptions is smaller which causes that numerical solutions must be used. In this group the models are solved taking into account non-uniform spraying and hydrodynamics of drying agent in a dryer. In all models in this group, the parameters of the drying agent in a cross-sectional area of the dryer are taken as constant. Zbicinski et al. reported that results obtained in this group of models describe well the process of spray drying [11]. However, the application of overall heat and mass balance equations makes it impossible to determine local distributions of particular parameters for flue gas during spray drying.

In literature, the studies on trona based scrubbing in spray dryer are limited to experimental studies. The kinetic studies on the reaction between trona and SO_2 are also based only on TGA experiments and it is concluded that the reaction occurs at dry base. On the other hand, all model approaches for spray dryer scrubbing of SO_2 are based on lime or limestone slurries [8,9,12–14].

In this study, SO_2 removal efficiency of a spray dryer was investigated by using natural soda (trona) solutions. For modelling purposes, the contact of gas and liquid phases is considered under plug flow conditions. The model equations were derived assuming the reactions are occurring in aqueous phase. The verification of the model was also investigated by comparing the theoretical data with experimental results.

2. Model description

Natural soda ore, namely trona, is one of the main sources of the alkaline solutions which has the form of $Na_2CO_3NaHCO_3{\cdot}2H_2O$. It dissolves readily in water and can be used for SO_2 scrubbing. The following reactions are expected to take place in the spray dryer.

$$2NaHCO_3 \rightarrow Na_2CO_3 + CO_2 + H_2O \quad (1)$$

$$Na_2CO_3 + SO_2 \rightarrow Na_2SO_3 + CO_2 \quad (2)$$

$$Na_2SO_3 + (1/2)O_2 \rightarrow Na_2SO_4 \quad (3)$$

In the reaction of SO_2 with soda, the major product is Na_2SO_3. With an increase in temperature some Na_2SO_4 is also formed in the presence of oxygen. Doğu et al. reported that formation of Na_2SO_4 causes an additional diffusion resistance for the transport of SO_2 to the reaction surface [15]. At low temperatures, formation of $Na_2S_2O_5$ was also expected during the reaction [16].

Although the overall reaction chemistry is fairly simple, the simultaneous heat and mass transfer is considerably more complex. Furthermore, several drying phases in the spray dryer, representing quite different reaction conditions, are involved. The mixing conditions also play an important role in modelling.

From the standpoint of classical mass transfer theory, two limiting reaction concepts can be identified. At high stoichiometric ratios based on fresh soda, excess of ${CO_3}^{2-}$ ions will prevail in the whole droplet. Hence, the absorption of SO_2 will be controlled by mass transfer of SO_2 from the gas bulk to the droplet interface. However, if soda is consumed to a sufficient extent, SO_2 will penetrate into the droplet and reaction will take place in the droplet. Thus, the reaction rate is likely to be limited by the dissolution rate of the sorbent. In the case of Na_2CO_3–SO_2

reaction, the reactant solid (Na_2CO_3) is itself porous. The pore mouth closure and rapid activity decrease are not very significant in the SO_2–soda reaction [5].

During the drying of droplets, two separate drying phases can be identified: constant-rate drying phase and falling-rate drying phase. In the first, liquid droplets leaving the atomizers contain a large number of individual particles, which are in constant random motion within the droplet. As evaporation occurs, the droplet shrinks and the distance between the single particles is reduced until particle surfaces touch. Hence, the particulate motion within the droplet becomes restricted and the droplet diameter remains unchanged during subsequent drying. During the second, evaporation of moisture from the interior of the particle to the surface starts to predominate. Due to the nature of the solid crust, all the particles are immobile. Hence, physical and chemical properties of the particles may vary with their distance from the center of the drop since reaction products formed at one location cannot move to another. As a preliminary design consideration, the assumption of surface saturated condition and neglect of a drying falling rate period, with resulting constant droplet surface temperature, is probably adequate. In other words, the change in the solids content, due to chemical reactions during the drying process, is not taken into account.

2.1. Mass transfer and reaction

The two-phase flow inside the spray dryer is described with a steady state one-dimensional model.

The overall mass transfer and reaction Eq. (2) for sulfur dioxide absorption can be subdivided into the following steps:

(a) Diffusion of SO_2 from the bulk gas to the droplet surface.

(b) Absorption of SO_2 at the droplet surface:

$$SO_2(g) = SO_2(aq)$$

Trona is a double salt, which has two moles crystal water. This crystal water has no effect on production of OH^- ions during the reaction. On the other hand, $NaHCO_3$ decomposes as shown in Eq. (1) and it can be assumed that there are some OH^- ions to form sulfurous acid. In limestone based scrubbing studies it is reported that the relative humidity should be over 70% [3]. The FGD studies using trona solution showed that 5% relative humidity is enough to remove the SO_2 with efficiency of 99% [15]. That means the reaction occurs at dry base. Consequently, if the relative humidity is increased more than 5%, the particles will have excess OH^- ions to allow the formation of sulfurous acid:

$$SO_2(aq) + H_2O(l) \rightarrow H_2SO_3(aq)$$

(c) Dissociation of sulfurous acid into ionic sulfur species:

$$H_2SO_3(aq) \rightarrow H^+ + {SO_3}^{2-}$$

(d) Diffusion of liquid phase sulfur species towards the droplet center.

(e) Dissolution of Na_2CO_3 at the outer shell of the unreacted core:

$$Na_2CO_3(s) \rightarrow 2Na^+ + CO_2 + (1/2)O_2$$

It is assumed that the presence of CO_2 has no influence on the desulfurisation process [17].

(f) Precipitation of sodium sulfite:

$$2Na^+ + {SO_3}^{2-} \rightarrow Na_2SO_3(s)$$

The reactivity of the slurry droplets toward SO_2 is a strong function of how long the bulk water phase is maintained. Since these reactions are assumed to occur in aqueous phase, the removal of water by evaporation limits the reaction steps. In general, ionic reactions are instantaneous in aqueous phase, but extremely slow in the absence of water. The reactivity of the slurry is also dependent upon the droplet size. Smaller drops have a higher surface area than large drops and thus have higher SO_2 capture rates but their water evaporates faster. Thus, it can be said that the larger drops, having a longer wet lifetime, would be more reactive. In this study, droplets were assumed to be spherical, rigid and isothermal [8].

The drying begins with the constant rate period during which the drying rate is controlled by gas phase resistance for heat and mass transfer only. Afterwards the water recedes into the porous solid and the falling rate period starts. In this period an additional resistance lowers the drying rate.

The heat and mass transfer rates to the droplets are given by:

$$\frac{dQ}{dt} k\,Nu\,d(T_a - T_s) \tag{4}$$

where T_a is the air temperature and T_s is the droplet surface temperature.

$$\frac{dM}{dt} = D\rho_a\,Sh\,d(H_w - H_a) \tag{5}$$

where H_w is the humidity at the saturated surface and H_a is the humidity of air.

The Nu and Sh numbers have both been estimated from the Ranz–Marshall equation [18]:

$$Nu = 2 + 0.6\,Re^{1/2}\,Pr^{1/3} \tag{6}$$

$$Sh = 2 + 0.6\,Re^{1/2}\,Sc^{1/3} \tag{7}$$

Drying of the slurry droplets can be described as drying of pure water droplets [9]. The evaporation rate equation is:

$$\frac{dW}{dt} = \frac{Ah(T_a - T_s)}{\lambda} \tag{8}$$

Using the definition of mass of a spherical particle and Nu number we can obtain the evaporation rate of a single droplet containing insoluble solids as follows:

$$\frac{dW}{dt} = -\frac{1}{m_p}\left(\frac{Nu\,\pi dk\,\Delta T}{\lambda}\right) \tag{9}$$

During the evaporation process, soda is simultaneously being consumed by the absorbed SO_2. The stoichiometric ratio is defined as the ratio of sodium carbonate mole flux to sulfur dioxide mole flux at the inlet of the spray dryer:

$$\mathrm{SR} = \frac{N_{\mathrm{Na_2CO_3,i}}}{N_{\mathrm{SO_2,i}}} \tag{10}$$

The absorption efficiency is defined as the ratio of sulfur dioxide mole flux absorbed inside the spray dryer to the sulfur dioxide mole flux at the inlet of the spray dryer:

$$\eta = \frac{N_{\mathrm{SO_2,i}} - N_{\mathrm{SO_2,o}}}{N_{\mathrm{SO_2,i}}} \tag{11}$$

The inner core of the spray-dryer agglomerate contains unreacted Na_2CO_3 a consequence of the rate of water evaporation exceeding the rate of SO_2 absorption. Fraction of unreacted soda, S, is related to the stoichiometric ratio and absorption efficiency as follows:

$$S = \frac{N_{\mathrm{Na_2CO_3,o}}}{N_{\mathrm{Na_2CO_3,i}}} = 1 - \frac{\eta}{\mathrm{SR}} \tag{12}$$

The soda conversion rate can be described by the following equation:

$$\frac{\mathrm{d}S}{\mathrm{d}t} = -\frac{1}{m_{\mathrm{p}}}(Sh\,\pi d D C_{\mathrm{SO_2}} M_{\mathrm{s}}) \tag{13}$$

We can eliminate the time variable and obtain the following equation:

$$\frac{\mathrm{d}W}{\mathrm{d}S} = \frac{Nu}{Sh}\frac{k\,\Delta T}{D C_{\mathrm{SO_2}} M_{\mathrm{s}} \lambda} \tag{14}$$

The temperature gradient in the spray dryer may be described by the mean logarithmic temperature difference. We can also describe the $C_{\mathrm{SO_2}}$ term using total gas concentration (C_{T}) and mole fraction of SO_2 in gas ($y = y_{\mathrm{SO_2}}$) terms. Defining the residual water fraction in droplet as x related with the initial water content of droplet (W_0), Eq. (14) can be written as:

$$\frac{\mathrm{d}x}{\mathrm{d}S} = \mathrm{SR}\frac{(\Delta T/\Delta T_2)(y_0/y)}{\beta} \tag{15}$$

where

$$\beta = \frac{Sh}{Nu} Le \tag{16}$$

Considering the plug flow conditions, means the temperature and SO_2 concentration gradients are observed along the dryer's axis, accordingly we can define the unreacted soda fraction (S) in SO_2 mole fraction terms as follows:

$$S = 1 - \frac{(1 - y/y_0)}{\mathrm{SR}} \tag{17}$$

Since the fraction of unreacted soda (S) in the droplets varies with the SO_2 concentration in the gas flow, Eq. (15) should be written again using Eq. (17):

$$\mathrm{d}x = \left(\frac{\Delta T/\Delta T_2}{\beta}\right)\left(\frac{1}{y/y_0}\right)\mathrm{d}\left(\frac{y}{y_0}\right) \tag{18}$$

Table 1
Chemical analysis of Beypazarı trona ore

Na_2CO_3 (%)	45.28
$NaHCO_3$ (%)	35.88
Na_2SO_4 (ppm)	1860
NaCl (ppm)	630
SiO_2 (ppm)	1300
Al_2O_3 (ppm)	63
Fe_2O_3 (ppm)	80
CaO (ppm)	721
MgO (ppm)	413

Using the following boundary conditions we can integrate Eq. (18) to obtain the removal efficiency of the spray dryer at plug flow conditions (Eq. (19)):

$$x = 1 \rightarrow y = y_0$$

$$x = 0 \rightarrow y = y_0(1 - \eta)$$

and using the definitions given in Eqs. (12) and (17):

$$\eta = \frac{\beta\,\Delta T_2(1 - \mathrm{SR}\,C_{\mathrm{Na_2CO_3}})}{\Delta T} \tag{19}$$

where $C_{\mathrm{Na_2CO_3}}$ is the conversion efficiency of trona and the temperature gradients are the mean logarithmic temperature differences. Here, β depends on Nu and Sh numbers as described in Eq. (16). Also, Nu and Sh numbers, given in Eqs. (6) and (7) depend on the diameter and velocity of droplets. Masters pointed that the droplets decelerate rapidly to their terminal velocity and the deceleration period is negligible compared to the total duration of the drying phase with respect to the evaporation [19]. Hence, the Nu and Sh numbers can be assumed around 2 considering typical droplet diameter values. Based on the assumptions, β can be estimated using Eq. (16).

3. Experimental

The chemical composition of the trona ore, which is obtained from Beypazarı ore, is given in Table 1 [20]. Solutions containing 10%, 15% and 20% trona (on the basis of mass) were prepared and used in experiments. In this work, a pilot size spray dryer was used to investigate the sorption steps. In order to investigate the effect of Na/S ratio on the absorption efficiency, experiments were conducted with Na/S ratios changing between 0.5 and 2.5. Sulfur dioxide was introduced to the air stream before the heater of the spray dryer. The effect of outlet temperature on SO_2 scrubbing was also investigated running the dryer in the range of 85–120 °C. The experimental conditions are shown in Table 2.

Table 2
Experimental conditions for spray dryer

Feed rate of trona solution (l/h)	1.2–3.2
Feed temperature of trona solution (°C)	25
Feed rate of SO_2 (l/min)	0.6–2.4
Feed rate of air (l/min)	226
Trona solution concentration (%)	10–20
Na/S	0.5–2.5
Outlet temperature (°C)	85–120

Table 3
End product composition at two different outlet temperatures (10% trona solution)

T (°C)	SR	Na_2CO_3	$NaHCO_3$	Na_2SO_3	Na_2SO_4	SO_2 removal efficiency	Na_2SO_3/Na_2SO_4
85	2.5	9.75	35.06	46.60	8.30	91.6	5.6
100	2.5	8.45	33.03	48.87	7.39	85.6	6.6

The product samples were also analysed for Na_2O, Na_2CO_3, $NaHCO_3$, Na_2SO_3, Na_2SO_4.

4. Results and discussion

Experiments were performed to demonstrate the influences of stoichiometric ratio, gas temperature and inlet trona solution concentration. The results are discussed in terms of SO_2 removal efficiency and soda conversion efficiency. The effect of the stoichiometric ratio on the SO_2 removal efficiency for various inlet trona solution concentrations is illustrated in Fig. 1. With increasing stoichiometric ratio the removal efficiency is increasing. At the stoichiometric ratio (Na/S = 2), SO_2 removal efficiency was found to be around 90%. On the other hand, Fig. 1 shows that the feed solution concentration affects the SO_2 removal efficiency. With increasing solution concentration the absorption efficiency is decreasing. In experiments conducted with Na/S ratios greater than 2, considerable amounts of $NaHCO_3$ remained in the product. Results of two typical runs conducted with a SR of 2.5 are given in Table 3. In both these runs the Na_2SO_3 and Na_2SO_4 components exist in the product as 50–60% while SO_2 removal is about 90%. Wang et al. reported that the $NaHCO_3$ decomposition reaction starts at around 90 °C and became significant after 110 °C [21]. Our results also showed that the conversion of $NaHCO_3$ according to Eq. (1) was very small below 110 °C. In the figure, the reference line corresponding to complete trona conversion is also shown.

The conversion efficiency of trona is defined as the ratio of converted soda mole flux to soda mole flux at the inlet of the spray dryer:

$$C_{Na_2CO_3} = \frac{N_{Na_2CO_{3,i}} - N_{Na_2CO_{3,o}}}{N_{Na_2CO_{3,i}}} = 1 - S \qquad (20)$$

Fig. 2 shows that trona conversion increases as the stoichiometric ratio decreases. That means the unreacted soda fraction decreases and therefore $N_{SO_{2,o}}$ increases.

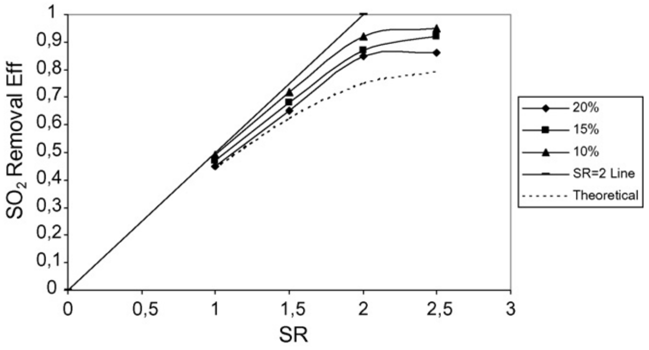

Fig. 1. Influence of stoichiometric ratio on SO_2 removal efficiency for various trona solution concentration.

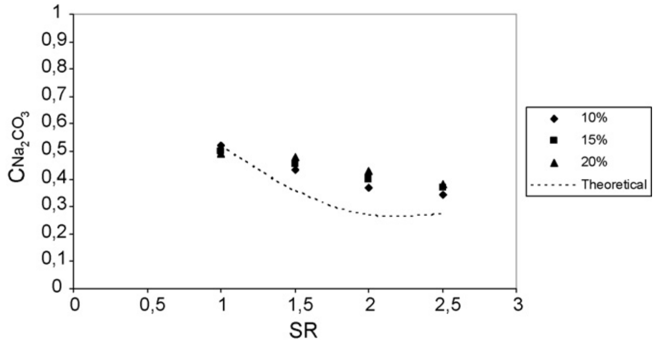

Fig. 2. Influence of stoichiometric ratio on sodium carbonate conversion for various trona solution concentration.

Increasing trona conversion causes to additional liquid phase mass transfer resistances for SO_2 which lower the absorption efficiency as shown in Fig. 1.

The influence of the gas temperature is illustrated in Fig. 3. The SR based on fresh trona feed is 2.5. Inlet temperature is 150 °C. The experimental and theoretical data for the sodium carbonate conversion efficiency are also shown in the same figure. The figure was produced as a function of the dimensionless temperature to compare the experimental data with the model given in Eq. (19), where $\Delta T_2/\Delta T$ can be assumed as the logarithm of dimensionless temperature. It is clearly shown that at the operating conditions the experiments yield higher efficiencies for both SO_2 removal and sodium carbonate conversion. On the other hand, increase of temperature caused a decrease in the conversion to Na_2SO_4 (Table 3). This may be attributed to possible further sorption of SO_2 on Na_2SO_3 to form $Na_2S_2O_5$.

The effect of outlet temperature was investigated for different temperature levels changing from 85 to 120 °C. Increase

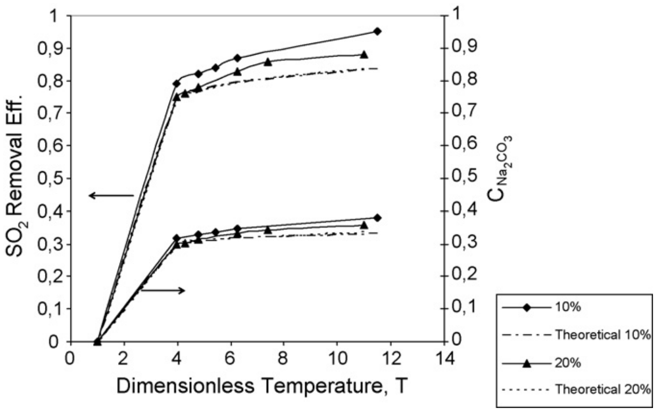

Fig. 3. Removal efficiency of SO_2 and conversion efficiency of sodium carbonate for dimensionless temperature ($\Delta T_2/\Delta T = \ln \tau$); inlet temperature, 150 °C; SR, 2.5.

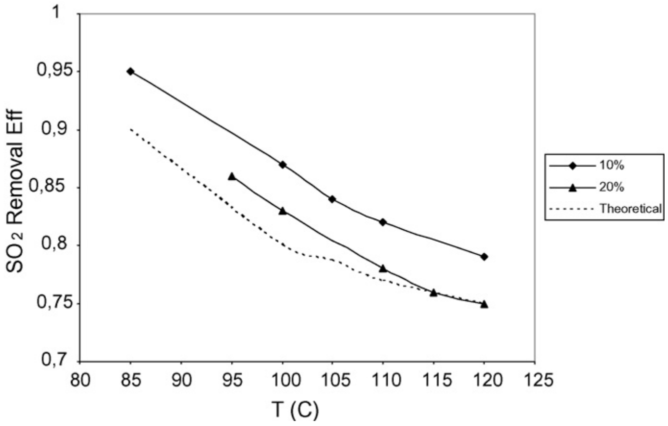

Fig. 4. Effect of outlet temperature on SO_2 removal efficiency (SR = 2.5).

of temperature caused a decrease in SO_2 removal efficiency as shown in Fig. 4. Since the absorption efficiency depends on the initial water content of the droplet, more concentrated trona solution shows lower SO_2 capture performance. On the other hand, at high stoichiometric ratio, the conversion of the trona is low and an increase in the temperature causes an increase in the drying rate and therefore the time period for absorption is decreased. Absorption efficiencies calculated from Eq. (19) agree well with the experimental data for more concentrated trona solution.

5. Conclusions

In this paper, a model approach for SO_2 removal by spray dry absorption with a trona slurry is presented. The model proposed shows good agreement with experimental results. It was concluded that trona based scrubbing of SO_2 could be achieved with an efficiency over 90%. The model showed that the absorption mechanism could be considered instead of dry-base mechanism at high relative humidities. As a result of the study, the Na/S ratio and temperature have direct effect on the removal efficiency. The trona solution concentration is found to be more effective at less concentrated solutions. At operation with excess of trona, the absorption efficiency is lowered by additional liquid phase mass transfer resistances for SO_2. It is also known that the particle size of the droplet is another important parameter for the removal efficiency and must be further considered.

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