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Experimental and modelling analysis of seawater scrubbers for sulphur dioxide removal from flue-gas

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ABSTRACT

Scrubbing with seawater is a reliable technology for flue-gas desulfurization in coal-fired power plants or in marine applications. The introduction of new regulations for emission control in Europe and the due increment of operational efficiency are pushing forward the optimization of scrubber's design, for which packing columns may be a better solution compared to spray columns. The design of a seawater scrubber mainly relies on a correct assessment of the equilibrium conditions and of the mass transfer coefficients in the absorber. In this work, experiments are performed on a model flue-gas $(32 \, \text{m}^3 \cdot \text{h}^{-1})$ having a SO₂ concentration in the range $500-2000 \text{ ppm}_{v}$, treated in a packed column filled with a structured packing (Mellapak 250X^{*}) at 1 atm and 25 °C, with liquid-to-gas ratio between 1.06 and 3.44 kg·kg⁻¹. Three different absorbing solutions are investigated: (a) a seawater; (b) a basic solution obtained by adding $200 \text{ mg} \text{L}^{-1}$ NaOH solution to the seawater and (c) distilled water, used as benchmark. In order to analyze the packed column tests, SO₂ equilibrium absorption tests at low concentrations (100-2000 ppm_v) are carried out in a feed-batch reactor, using the same absorbing solutions tested in column tests. The experimental tests in packed column indicate that, for SO₂ concentration as low as 500 ppm, an absorption efficiency above 98% can be achieved by using liquid-to-gas mass ratio about to 2.91 kg/kg^{-1} . In this condition, few differences appear among solutions (a) and (b). For the highest SO2 concentration (i.e. 2000 ppm_v), a maximum of 85% efficiency is observed for liquid-to-gas mass ratio of 3.44 kg kg⁻¹. Modeling of both the equilibrium and the dynamic data are implemented in Aspen Plus[®] V 8.6. However, while modeling assures a very good prediction of the equilibrium data, it is not able to properly describe the dynamic test results, with a systematic underestimation for tests having a removal efficiency higher than 85%.

1. Introduction

Sulphur dioxide (SO_2) is one of the primary air pollutants having extensive and proved damages to human health and the Environment. For this reason, stringent regulations for emission control have been enacted in many countries to limit the amounts emitted by anthropogenic activities.

The main sources of SO₂ emissions are related to the combustion of fossil fuels containing sulphur, such as coal, oil or gas. Li et al. [1] investigated the air quality and pollutant emissions from fifteen Chinese coal-fired power plants having a power generation capacity ranging from 125 to 600 MW_{th}. The sulphur content of the burnt coal was between 0.32 and 2.63% and the SO₂ concentration in the flue-gas before desulfurization units resulted on average between 210 and 1540 ppm_v.

The European Union regulations in force (2001/81/CE and 2003/ 35/CE) provide the guidelines for pollutant emission control. The current European regulation (2010/75/UE) on SO₂ emission limits in coalfired power plants installed before 2013 posed a limit of $200 \text{ mg} \text{ m}^{-3}$ (71 ppm_v) for units with power from 50 to 100 MW_{th} and 75 mg m⁻³ (26.5 ppm_v) for units with power higher than 300 MW_{th}.

Very recently, the Directive 2016/2284/CE established the entry in force of new regulations for SO₂ emissions for the years 2020–2029 and after 2030. On average, SO₂ emission cuts are planned to be within 8–83% for the different EU member states. This will generate a relevant increase in the required removal efficiency. For example, at moment, a coal fired plant with an installed power of 90 MW_{th}, emitting 2000 ppm_v of SO₂, has to comply with a 71 ppm_v limit, corresponding to a removal efficiency about 95.2%. For the same plant, the new regulation limit (35.5 ppm_v), assuming a 50% cut of the allowed emission on an average basis, will lead to a required efficiency greater than 97.6%. In Europe, the database of the European power plant infrastructure [2] shows that most of the installed and commissioned power plants are powered by fossil fuels (mainly coal and natural gas). Moreover, most of them are located near the main fuel transport routes,

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Nomenclature

List of symbols

	2	11-x,1(aq
A_i	concentration of a generic anion, $mol m^{-3}$	
a_e	wet effective surface of packing, m ² ·m ⁻³	M_S
a_n	nominal surface of packing, m ² ·m ⁻³	MW_S
$a_{i,i}$	activity of the generic i-th specimen in the j-th reaction	NTU
<i>b</i> _i	stoichiometric coefficient of atom X in the i-th specimen	Р
	$M_{x,i}$	Q
C_i	concentration of a generic cation, $mol m^{-3}$	Re_G
$C_{SO_2}^o$	initial gas concentration of sulphur dioxide, ppm _v	S
C_{SO_2}	final gas concentration of sulphur dioxide, ppm _v	S_c
C_E	surface renewal factor, –	Sc_G
\mathscr{D}_{G}	gas diffusivity, $m^2 \cdot s^{-1}$	Т
\mathscr{D}_L	liquid diffusivity, $m^2 s^{-1}$	t*
d_{ea}	equivalent diameter, m	$t_{I_{c}}$
E	enhancement factor, –	X
F_G	loading factor, Pa ^{0.5}	
F_{SE}	surface enhancement factor, -	$x_{s,m}^*$
F_t	correction factor for total liquid-holdup, –	S(IV)
G	gas molar flow rate, mol·s ⁻¹	$x_{S(n)}$
HTU	height of transfer unit, m	v_{aa}^*
HTU_{σ}	height of transfer unit related to gas-phase mass transfer	J SO ₂
8	resistance, m	v_{aa}^{o}
HTU_l	height of transfer unit related to liquid-phase mass transfer	V SO2
	resistance, m	Z
k_G	gas mass transfer coefficient, $m \cdot s^{-1}$	Z_i
k_L	liquid mass transfer coefficient, $m \cdot s^{-1}$	
k_{ov}	global mass transfer coefficient, mol·m ⁻² ·s ⁻¹	Greek
k_r	liquid mass transfer coefficient, $mol m^{-2} s^{-1}$	
k,	gas mass transfer coefficient, $mol m^{-2} s^{-1}$	$\eta_{so_{r}}$
Ĺ	liquid molar flow rate, $mol s^{-1}$	ρ_G
L/G	liquid-to-gas ratio, $kg kg^{-1}$	ρ_{I}
M _{r i}	molar mass of the generic molecular specimen containing	π
л,1		

Fuel 214 (2018) 254–263

		the atom <i>X</i> , mol
	$M_{x,i_{(g)}}$	molar fraction of the generic molecular specimen con-
	м	taining the atom X in the gas phase, mormor
	$M_{x,i(aq)}$	molar fraction of the generic molecular specimen con- taining the storn V in the liquid phase mol mol^{-1}
	м	mass of the absorbing solution for batch tests, a
	MIZ	mass of the absorbing solution for batch tests, g
n	NTI	number of transfer units
non	D	number of transfer units, –
ICII	P	gas pressure, and h_{1}
	Q D-	Pourselde number in see above
	Re _G	Reynolds number in gas phase, -
	S S	section of the packing column, m
	S _c	side dimension of packing, m
	Sc_G	schindt number in gas phase, –
	1	temperature, K
	L.	saturation time, s
	t_L	liquid exposure time of packing, s
	X	total atomic concentration of a specimen in solution, $molm^{-3}$
	$x^*_{S_{(IV)}}$	mole fraction of sulphur dioxide at thermodynamic equi- librium mol-mol ⁻¹
	$x_{S(m)}$	liquid mole fraction of total sulphur, mmol·mol ^{-1}
	v_{20}^{*}	mole fraction of sulphur dioxide at thermodynamic equi-
fer	J SU2	librium, mol·mol ^{-1}
	y_{so}^{o}	initial gas mole fraction of sulphur dioxide, $mol mol^{-1}$
fer	y_{SO_2}	final gas mole fraction of sulphur dioxide, $mol mol^{-1}$
	Z^{2}	packed height of column, m
	z_i	charge of the generic ion
	Greek sy	mbols
	nao	removal efficiency of sulphur dioxide. –
	ρ_{c}	molar gas density, $mol m^{-3}$
	ρ _τ	molar liquid density, $mol m^{-3}$
ino	π	Pi Greek. –
0		· · · · · · · · · · · · · · · · · · ·

such as sea, river or lakes [2].

In order to comply with the emission limits, different post-combustion desulphurization systems, commonly referred as to flue-gas desulfurization (FGD) systems, have been proposed. In general, the choice of an appropriate FGD system depends on both plant size and emission targets of flue-gas.

Dry scrubbers are among the most established technologies, as they are used for flue-gas cleaning in coal power plants since the early 1970. In this process, SO_2 removal is carried out by scrubbing the flue-gas with a limestone suspension [3–5]. When a removal efficiency higher than 90% is needed, wet scrubbers are preferred. These systems provide a wider choice of chemical reagents as absorbing liquids, such as sodium hydroxide [6], sodium carbonate and bicarbonate [7], sodium chlorite [8–11], hydrogen peroxide [12]. Alternatively, ozone oxidation systems can be also used [13].

A technically viable and economically feasible alternative to chemicals is the use of seawater as absorbing solution [14–16]. Recently, seawater scrubbers have been proposed for coastal installations and marine diesel engine applications, due to the large availability and favorable chemical properties (e.g. the intrinsic alkalinity). The presence of alkaline compounds such as carbonates and bicarbonates in equilibrium generate a buffering effect that allows preserving the pH conditions favorable to SO₂ absorption [17–19]. Some authors also observed that the high salinity content in seawater solution, mainly as sodium chloride, further improved the SO₂ absorption [20].

The absorption process generates an acid wash-water having a pH in the range 2.5–4. Hence, wash-water has to be corrected for pH values before discharge, usually with a minimum allowed pH of 0.5 points below that of the inflow seawater. This is achieved by either direct dilution of the scrubber wash-water with seawater or with NaOH addition.

For this reason, it was sometime considered more useful to add NaOH to the same absorption seawater feed, in order to support SO_2 removal while preserving wash-water pH level close to neutrality. The main issue related to this strategy is the pH limitation induced by precipitation of metal hydroxides in seawater, which, depending on salinity, typically occurs for pH above 9.7–10.

The main advantage of seawater scrubbers compared with conventional lime or limestone scrubber is the absence of chemicals and the less tendency to nozzle clogging and scaling over the scrubber walls or over packing. On the other hand, seawater is more corrosive and higher flow rates are required to comply with regulations due to the lower concentration of hydroxides and carbonates. In practice, piping and pumps are the primary capital costs of a seawater scrubber plant and strategies to minimize the specific water flow rate are therefore needed to reduce both operation and capital process costs. In particular, the ratio between seawater molar flow rate, L, and gas molar flow rate, G, depends on the equilibrium conditions, which identify the minimum value of L/G to operate the absorber. Besides, the L/G ratio depends on the efficiency of liquid-gas contact, in terms of mass transfer rate, which defines the scrubber height and diameter.

Spray columns are easy and largely preferred for limestone scrubbers because they have less scaling problem limitations compared to packed towers. One of the main issue of spray columns is the difficulty in predicting the mass transfer rates, which strongly depends on the droplet size distribution, but also on the occurrence of droplets Download English Version:

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