

## **RESEARCH ARTICLE**

#### MARINIZATION OF A TWO-STAGE MIXED STRUCTURED PACKING SCRUBBER FOR SOX **ABATEMENT AND CO2 CAPTURE.**

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#### ..... Manuscript Info

Manuscript History Received: 03 February 2019 Final Accepted: 05 March 2019 Published: April 2019

Key words:-SOx reduction, IMO requirements, Wet scrubbers, packed beds, Pressure drop, CO2 capture.

Abstract

..... In this paper, a practical investigation to evaluate the impact of a twostage mixed structured packing scrubber for SO<sub>x</sub> abatement and CO<sub>2</sub> capture using amine solutions is presented. The practical test consists to connect the two-stage packing column to the diesel engine (DE) exhaust outlet. The first stage of the scrubber, fed with a sodium hydroxide solution, served as a SO<sub>X</sub> absorber, where the liquid flow rate and the sodium concentration are the operating variables. Gasses are then transported from the first stage to the second stage, where they encounter an amine solution that can be recycled in a closed loop to absorb residual CO<sub>2</sub> streams. Tests are conducted with a 250kW diesel generator (DG) filled with a 0,5% Sulphur content fuel. Results revealed a significant decrease of SO<sub>x</sub> by an amount of 89% while CO<sub>2</sub> capture has shown a decrease by an amount of 49%. However, drop pressure was detected when DG operates at high loads due to the fouling structured packing.

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#### Introduction:-

Maritime transport emissions represent around 10-15% of global Sulphur oxide (SO<sub>x</sub>) and nitrogen oxide (NOx) emissions, and approximately 3,1% of global carbon dioxide (CO<sub>2</sub>) emissions [1], [2]. However, several studies confirmed that air pollutants could travel thousands of mile before deposition and contamination took place [3]. At the end 1980s, the international maritime organization (IMO) began its work and research on prevention of air pollution from ships and has adopted in 1997 the air pollution Annex VI (MARPOL convention).

This Annex came into force on 19 May 2005 after achieving the essential number of endorsers [4]. The Annex includes the establishment of emission control areas (ECAs) to scale down emissions in specified sea zones with a gradual reduction in emissions of  $NO_x$ ,  $SO_x$  and particulate matter (PM), figure 1 [5], [6]. Since then, several measures have been taken into consideration. A tier system has been adopted to reduce NO<sub>x</sub> levels, while SO<sub>x</sub> will be reduced from current 3, 50% to 0, 50% beginning from 1 January 2020 in international waters and to 0,1% for ECA. Table 1 summarizes the regulatory requirements to reduce ship emissions of Sulphur oxides for ship categories 1, 2 and 3, [7].



Figure 1:- Existing and potential new ECAs around the globe [6].

However, there are different feasible methods to meet Annex VI requirements. This can be accomplished by switching to a low-sulfur fuel or by applying exhaust after-treatment through absorption. As an alternative to low-sulfur fuel, MARPOL Annex VI recognizes exhaust gas cleaning systems (EGCS) that reduces sulfur emissions as efficiently as they do with low-sulfur fuel. The cost and benefit analysis of EGCS versus fuel changes was examined in [8-12]. Results have encouraged shipowners to choose primarily the EGCS option due to the ascending fuel price.

Starting year		Category 1 & 2			
(January 1 <sup>st</sup> )	Oceans	Control Areas	EU Ports	Coastal	ships
2010	4,5%	1,0%	0,1%	$0,5\%^{*}$	0,05%
2012	3,5%	1,0%	0,1%	0,1%	0,0015%
2015	3,5%	0,1%	0,1%	0,1%	0,0015%
2020-(2025)**	0,5%	0,1%	0,1%	0,1%	0,0015%

Table 1:- Low Sulfur Phase-In Dates [7].

0, 5%<sup>\*</sup>: Marine Gas Oil, or 0, 1% Marine Diesel Oil

#### (2025)\*\*: Implementation of Oceans limit at 0, 5% Sulphur

In the last decade, application of absorbers for Sulphur dioxide (SO<sub>2</sub>) elimination has increased considerably in marine transport and they are generally called scrubbers [13]. Scrubbers are categorized as dry scrubbers, using dry lime and other calcium-based pH control minerals, or as wet scrubbers using an alkaline solution, figure 2 [14]. The open loop scrubber is the easiest system, where water is supplied from the sea, pumped, filtered and sprayed into the scrubber using nozzles that diffuse water into droplets. However, open loop scrubber is only profitable if the water is alkaline. This can be accomplished by adding an alkali chemistry or by using seawater with a natural alkalinity extracted from the bicarbonate ion (HCO<sub>3</sub><sup>-</sup>) existing in the seawater [14]. The water is released back into the sea after particular matters are eliminated. Figure 3 illustrates a schematic of an open loop wet scrubber [15]. However, operation of the open loop scrubber system in fresh water can restrict scrubbing of SO<sub>x</sub> due to the weak alkalinity of the water [16].



Figure 2:-The hierarchy of SO<sub>X</sub> Scrubber systems [14].



Figure 3:- Schematic for an open loop scrubber system. Particles are eliminated from the water and released back into the sea [15].

For this, it is therefore more interesting to develop closed loop technology for shipowners sailing in fresh water such as the St. Lawrence River (Canada) and the Great Lakes (USA and Canada). Fresh water treated with an alkaline chemical like caustic soda is employed for neutralization in a closed loop scrubber system (including hybrid SO<sub>x</sub> scrubbers when operating in closed loop mode). Fresh water scrubbers are used when high efficiency cleaning is required or when the varying alkalinity associated with seawater prevents the use of marine scrubbers [16]. Nevertheless, closed loop fresh water scrubber systems have much smaller discharge rates than open loop sea scrubber systems by an amount approximately of 0,1 to 0,3 m<sup>3</sup>/MWh and occurs a smaller volume of effluent [17]. Moreover, closed loop fresh water scrubber system can periodically be operated in zero discharge mode without discharging any overboard wash water. Figure 3 shows the schematic for a closed loop fresh water scrubber system [15].



Figure 3:-Schematic for a closed loop wet SO<sub>X</sub> scrubbing system [15].

Finally, hybrid wet  $SO_X$  scrubber systems either can operate in open loop mode or closed loop mode offering advantages that sodium hydroxide is used only when required, reducing handling and storage costs. However, hybrid wet  $SO_X$  scrubber systems have more complex design. On the other hand, dry  $SO_X$  scrubber systems have

been generally applied in land-based industry since 1970. Dry  $SO_X$  scrubber use calcium hydroxide granules (Ca  $(OH)_2$ ) which react with sulfur oxides to form gypsum (Ca $SO_4$ -2H<sub>2</sub>O). Contrary to wet  $SO_X$  scrubbers, dry scrubbers do not require wash water treatment making them ideal for areas where there is an increased sensitivity to discharge to the sea. However, as with closed loop operation of a wet system, consumables need to be stored and handled. Granules used must also be stored offshore before disposal.

The objective of this paper is to evaluate the effect of a vertical two-stage mixed structured closed loop-packing scrubber using a sodium hydroxide solution for  $SO_X$  abatement and  $CO_2$  capture in aqueous Monoethanolamine (MEA) solutions. Indeed, the closed loop technology is much more interesting to develop for shipowners sailing in fresh waters and who have hybrid or closed loop with a 0,5% sulfur content fuel. Attention was given to the drop pressure in the scrubber column due to the fouling structured packing and on the level of  $SO_X$  abatement and  $CO_2$  capture.

#### **Experimental:-**

The experimental setup is based on the absorption principle of contacting the gas phase with the liquid phase at room temperature and atmospheric pressure. A vertical two-stage gas-liquid counter-current packed bed (1) is used as absorbers and connected to the diesel engine exhaust outlet (Fig.4). The scrubber consists of an inox steel column (2), which is assembled up to a nominal height of 200cm around the 250kW diesel generator's main exhaust gas (3). The stainless steel fan (4), which is specifically designed to operate at high temperatures (up to 400°C), is connected to the main exhaust of the diesel engine, the main purpose of which is to redirect the part of the gasses emitted to two heat exchangers (5), in order to reduce the temperature of the gasses below 115°C, so that the amine solution can have an effect on the CO<sub>2</sub> capture. To adjust and control the amount of gas passing through the packaging column (10 l/min), a needle valve (6) with a rotameter (7) are used. Additionally, two pumps were used, the first (8) to feed the packing column with NaOH solution (0,11/min) to capture the SO<sub>x</sub>, while the second (9) was used to feed the second packing column with a Monoethanolamine (MEA) solution (0,11/min) to reduce CO<sub>2</sub> emissions. Finally, at the inlet and outlet of the column, gas sensors (10) are installed to assess the CO<sub>2</sub> and SO<sub>x</sub> levels before and after treatment with a differential pressure drop sensors (11).



Figure 4:-Experimental setup representing the various components to assess the impact on the reduction of  $SO_X$  and  $CO_2$  emissions.

Table 2 summarizes the fluid properties, the range of operating conditions and the specifications of the packed bed, while table 3 illustrates the technical specifications of the diesel generator used during the test.

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	Parameter	Value/Range
(	Gas velocity, $U_g$	0,0016 - 0,10m/s
L	iquid velocity, $U_l$	0,0025 - 0,005m/s
Liqui	id surface tension, $\sigma_L$	0,072N/m
	Gas density, $\rho_g$	1,2Kg/m <sup>3</sup>
	Bed porosity, ε	0,395
	Bed length, L	0,3m
Co	olumn diameter, D	0,058m
Dies	sel Sulphur Content	0,5%
App	lied Load for testing	30%, 50% and 85%

#### **Table 2:-** Operational conditions and system properties ranges

The installation was conducted in a liquid phase recycling approach supplied and controlled by two peristaltic pumps. Two multipoint liquid distributors were used, consisting of 9 needle orifices with an internal diameter of 1mm for NaOH and MEA supply. The evolution of gas state for  $SO_X$  and  $CO_2$  as well pressure drops through the bed were measured in real time and transferred to the PC via a data acquisition system

Table 3:-	Generator set	specifications	according to	Caterpillar	manufacturer
	Obrierator Det	opeen en ono	ave of and to	Caronpinan	

Description	Value/Range
Genset power rating with fan@0,8p.f.	250kW
Open generator set	1800rpm/60Hz/600V
Fuel consumption@100% load with fan	71L/hr
Fuel consumption@75% load with fan	57,6L/hr
Fuel consumption@50% load with fan	39,2L/hr
Exhaust stack gas temperature	426°C

### **Results and discussion:-**

Evaluation of the  $SO_X$  and  $CO_2$  content at the outlet of the scrubber without the intervention of the NaOH and MEA solutions for different applied loads

The main purpose of this section is to assess the amount of  $SO_x$  in the inlet of scrubber as well as the  $CO_2$  level after combustion at 30%, 50% and 85% of applied loads. A  $SO_2$ -B4 sensor from alphasense air manufacturer was used for  $SO_2$  detection, while a MH-410D sensor from winsensor manufacturer for CO2 detection was employed. Table 4 shows the  $SO_x$  and  $CO_2$  level at the outlet scrubber. In addition, photos were taken with a thermal camera to assess temperature variation in heat exchangers in order to ensure a reduction in exhaust gas temperature as shown in figure 5 (a), (b) and (c).

<b>Function</b> of the box and cog levels at different folds during 6 minutes of test.										
Applied load	$Q_{Gas}$	SO <sub>X</sub> (ppm)				CO <sub>2</sub> (ppm)				
(%)	(L/min.)	2 min.	4 min.	6 min.	8 min.	2 min.	4 min.	6 min.	8 min.	
30		44	43	42	42	1,09	1,08	1,08	1,08	
50	10	33	32	33	33	0,97	0,97	0,97	0,97	
85		18	19	20	20	0,78	0,78	0,78	0,78	

Table 4:- Evaluation of the SO<sub>x</sub> and CO<sub>2</sub> levels at different loads during 8 minutes of test.



Figure 5:-Temperature in the heat exchanger at 30% of load in (a), at 50% of load in (b) and 85% of load in (c).

According to table 4, we can notice that the SOx and CO2 levels are lower when the charge increases. This can be explained by the fact that the combustion becomes better and the majority of the particles are burned under high loads.

Evaluation of the SOx content at the outlet of the scrubber with the intervention of the NaOH solution for different applied loads

 $SO_2$  associates with a salt in freshwater scrubbers and therefore does not react with the natural seawater bicarbonate. There are the coming reactions:

 $\begin{array}{ll} 2\text{NaOH} + \text{SO}_2 \rightarrow \text{Na}_2\text{SO}_3 + \text{H}_2\text{O} \ (\text{Sodium Sulfite}) & (1);\\ \text{Na}_2\text{SO}_3 + \text{SO}_2 + \text{H}_2\text{O} \rightarrow 2\text{NaHSO}_3 \ (\text{Sodium Hydrogen Sulfite}) & (2);\\ \text{SO}_2 \ (\text{Gas}) + \text{H}_2\text{SO}_4 \rightarrow \text{NaHSO}_4 + \text{H}_2\text{O} \ (\text{Sodium Hydrogen Sulfate}) & (3);\\ 2\text{NaOH} + \text{H}_2\text{SO}_4 \rightarrow \text{Na}_2\text{SO}_4 + 2\text{H}_2\text{O} \ (\text{Sodium Sulfate}) & (4). \end{array}$ 

A freshwater scrubber usually discharges 250 times less water than an open loop seawater scrubber. The bleed off is significantly smaller for fresh water units  $(0.1-0.3m^3/MWh)$  and as a result, the concentration of pollutants is higher, making washwater cleaning easier [18].

Table 5 shows the results of the reduction, followed by a comparison before and after NaOH's intervention, see table 6 and figure 6. It should be noted that only 0.1 liters of NaOH solution has been injected into the packing column.

Applied load	Q <sub>Gas</sub>		SO <sub>X</sub>	(ppm)		Pump (NaOH solution)
(%)	(L/min.)					l/min.
		2 min.	4 min.	6 min.	8 min.	
30		5,77	5,16	5,09	5,11	
50	10	3,27	3,45	3,35	3,34	0,1
85		1,89	1,90	1,89	1,88	

Table 5:-Evaluation of the SO<sub>X</sub> abatement with NaOH intervention

In order to compare results, we calculated the average of  $SO_x$  emissions before and after NaOH intervention and this for eight minutes of testing. Table 6 shows the average of  $SO_x$  emissions, while figure 6 shows the comparison.

Without intervention of NaOH						, I	With the	interventi	ion of Na	.OH				
Applied	Q <sub>Gas</sub>		SO <sub>X</sub> (ppm)			$SO_X(ppm)$ Average $SO_X(ppm)$					(ppm)	Average		
load	(L/min.)	2	4	6	8	(ppm)	2	4	6	8	(ppm)			
(%)		min.	min.	min.	min.		min.	min.	min.	min.				
30		44	43	42	42	42,75	5,77	5,16	5,09	5,11	5,28			
50	10	33	32	33	33	32,75	3,27	3,45	3,35	3,34	3,35			
85		18	19	20	20	19,25	1,89	1,90	1,89	1,88	1,89			

Table 6:-Evaluation of the average of SO<sub>X</sub> emissions before and after NaOH intervention



Figure 6:- Influence of the NaOH intervention on the  $SO_X$  abatement with a 0,5% sulfur content fuel.

According to table 6 and figure 6, the reduction of the  $SO_X$  emissions is 89% when the scrubber is fed with the NaOH solution. It can also be noted that due to better combustion in the piston chamber, the  $SO_X$  emission rate is further reduced at high load. However, an increase in pressure drop in the packing column has been detected due to the fouling structured packing under a high load. Figure 7 shows the microscopic Sulphur particles stuck on the packed bed causing an increase in the pressure drop.



**Figure 7**:- in (a) a small particles of sulphur appeared under a low load (30%) with the NaOH intervention; in figure 3(b) the particles of sulphur appeared more on the packed bed under a load of 50%, whereas in figure 3(c) and 3(d) the particles are thicker and further blocks the flow of exhaust gas creating an increase in pressure drop.

#### Pressure drop

The overall pressure drop consists of gas-liquid interactions and static head of the liquid phase in the packed beds with gas-liquid simultaneous flow. According to table 7, the pressure drop (inlet Vs outlet of the scrubber) increase by 9,93% after 3 minutes of operation between a low load (30%) and a medium load (50%), while it is 16,6% under a high load (85%). This can be explained by the increase in the flow and pressure of exhaust gasses when the load increase. Furthermore, the amount of the sodium sulfate (Na<sub>2</sub>SO<sub>4</sub>) increases further due to the reaction causing the fouling of the packed beds in the column. Figure 8 shows the pressure drop across the different applied loads.

Time (s)			
	30% of load	50% of load	85% of load
0	0	0	0
20	4	5	6
40	8	9	10,5
60	12	12,6	13
80	12,22	13	13,6
100	12,41	13,2	13,99
120	12,65	13,9	14,4
140	12,88	14	14,96
160	12,97	14,37	15,22
180	12,97	14,4	15,55
200	12,98	14,4	15,69

**Table 7**:-Evaluation of the pressure drop ( $\Delta P$ ) in the column at different loads



Figure 8:- Illustration of the pressure drop of low, medium and high loads in the packed beds for 3 minutes.

Evaluation of the  $CO_2$  content at the outlet of the scrubber with the intervention of the monoethanolamine (MEA) solution for different applied loads

Carbon dioxide is considered as potential source for greenhouse gas (GHG) development. Different studies have evaluated that 3 to 4 percent of universal  $CO_2$  emissions are attributable to marine vessels [19]. A 10 percent reduction in cruise speed is an effective operational methodology that offers a significant reduction in  $CO_2$  and fuel consumption up to 20 percent over the same distance [20].

In this section, attention is given to reduce the carbon dioxide level at the exit of the scrubber by injecting a 0,11/min of MEA solution. The following reversible reaction provides the basic reaction chemistry for an aqueous Monoethanolamine solution and  $CO_2$  [21]:

# $C_2H_4OH NH_2 + CO_2 + H_2O \underset{Cold}{\overset{Hot}{\rightleftharpoons}} C_2H_4OH NH_3^+ + HCO_3^-$ (5)

This is an exothermic reaction and per mole of  $CO_2$  absorbed in MEA solution, 72KJ of thermal energy is released. Absorption normally occurs around 50°C [21].

Table 8 shows the results of the reduction with a comparison before and after MEA solution intervention. It should be noted that only 0.1 liters of MEA solution has been injected into the packing column. Figure 9 illustrates the percentage reduction of the  $CO_2$  after MEA solution intervention.

Table 8:-Evaluation and Comparison of the CO<sub>2</sub> reduction using MEA solution

	Without the intervention of MEA solution						With the intervention of MEA solution					
Appl	lied	$Q_{Gas}$		CO <sub>2</sub> (%)			Average	CO <sub>2</sub> (%)				Average
loa	ıd	(L/min.)	2	4	6	8	(%)	2	4	6	8	(%)
(%	5)		min.	min.	min.	min.		min.	min.	min.	min.	
30	0		0,78	0,78	0,78	0,78	0,78	0,36	0,36	0,36	0,36	0,36
50	0	10	0,97	0,97	0,97	0,97	0,97	0,48	0,48	0,48	0,48	0,48
85	5		1,09	1,08	1,08	1,08	1,082	0,59	0,58	0,58	0,58	0,582



Figure 9:- Influence of the monoethanolamine solution on CO2 levels after intervention

According to table 8 and figure 9, the CO2 rate has decreased by an average of 49%. However, CO2 level increased with the load. This can be explained by the fact that the turbo absorbs a larger amount of air when the load increases.

#### **Conclusion:-**

Ship emissions are one of the major issues affecting those interested in the maritime domain, as they have a negative impact on the marine environment. The present paper discussed the various wet scrubber technologies, which could carry out to reduce those emissions.

A two-stage packed bed closed loop scrubber systems for sulfur abatement and CO2 capture was examined. For the evaluation of the efficiency of the system, a soda solution (NaOH) for sulfur abatement and monoethanolamine solution for CO2 capture were used. Tests are conducted using a 250KW diesel generator filled with a 0, 5% sulfur content fuel. The results showed the possibility to achieve valuable emission reduction percentage for the SO<sub>x</sub> and CO<sub>2</sub> by an average of 89% and 49%. However, the accumulation of the sodium sulfate (Na<sub>2</sub>SO<sub>4</sub>) in the packed bed due to the chemical reaction between the SO<sub>x</sub> and the NaOH solution, served further to the pressure drop in the scrubber by an amount of 17%.

On the other hand, the application of this methodology by ship operators allows them to meet the requirements set by IMO established in 2015 and that is to burn fuel with a sulfur content of less than 0,1% percent in ECAs starting by January 2020.

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