# MANAGEMENT OF THE CO<sub>2</sub> CAPTURE PROCESS INTEGRATION IN THE GLASS TECHNOLOGY INDUSTRY

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Abstract: The main objective of this study is the development of an inventory system of  $CO_2$  emissions in the processing and manufacturing of the glass, as well as ways to reduce them. Considering the characteristics of these types of processes: energy-intensive and with an increased impact on the environment (e.g. climate change), it is important to know the amount of  $CO_2$  generated in atmosphere. Today, the industrial processes consume more than one third of the global primary energy, while 70 % of this is provided by fossil fuels. Consequently, around 40 % of the globally CO2 emissions are generated by the industrial processes. One method to reduce the  $CO_2$  emissions is based on the integration in the glass fabrication process of the  $CO_2$  capture post-combustion technology. In this work, the integration of  $CO_2$ capture technology by chemical absorption and its integration effects on the glass technology performance were studied. The  $CO_2$  capture efficiency considered was of 90 %. The weight concentration of monoethanolamine and the  $CO_2$  lean loading solvent presented variations. The values of the thermal heat required for solvent regeneration obtained for cases studied were in the range 2-6 GJ/t<sub>CO2</sub>. The heat of waste gases from the glass process was used in two steam turbines, one of medium pressure and one of low pressure. The thermal energy required for solvent regeneration was extracted from the low pressure steam turbine. Initially, the thermodynamic efficiency of the steam turbines cycle was of 30 %, after the integration of the chemical absorption process the efficiency penalty was of 11-26 %. The usage of one system to recovery the heat of the waste gases reduces the negative impact of the chemical absorption process integration in the glass production.

*Keywords*: glass industry,  $CO_2$  emissions,  $CO_2$  capture technology, chemical absorption process.

## **1 INTRODUCTION**

In industry is important to make the distinction between the emissions generated from the energy production necessary of the process and the emissions generated from the manufacturing process of the products. The process emissions are produced in the chemical reactions due to the raw materials composition, generally this emissions cannot be avoided [1]. The amount of  $CO_2$  provided by the raw materials is directly related to the production of glass and is calculated based on the amount of carbonates in the raw materials [2], [3].

In the glass industry, the stages generation of emissions are the same, regardless of the technological type of process applied. The inventory of the pollution sources from the technological processes of glass manufacturing leads to the separation of the technological process in three stages: handling and mixing of raw materials, melting and shaping, and finishing (Figure 1) [4]. Although the raw materials used to produce different types of glass are mostly natural minerals widespread in nature, which generally do not have a significant impact on environmental factors, the handling of these materials presents a significant potential of dust pollution of the atmospheric air [5]. Thus, the diffuse emissions of particles in the storage and handling areas of the raw materials can vary according to the applied technologies and the technical condition of the equipments used in these technological processes. The glass manufacturing processes also require the use of the fossil fuels for production of the heat required for the melting of raw materials, dyes and organic solvents to perform painted glass products [6]. Generally, the household glass industry uses a very wide range of auxiliary materials that give the products different characteristics. The quantitative value of the final products varies between 75 % (crystals) and 90 % (soda-lime glass) of the raw materials entering in the process according to the physic-chemical processing of the shaping and finishing stages [5]. The ratio of raw and auxiliaries materials used in glass manufacturing technologies varies in a relatively large range depending on the final products obtained.



Figure 1: Technological flow of household glass production

The technological process of glass requires an amount of heat in the melting phase of the raw materials. To achieve the glass products the temperature required in the melting furnace is in the range 1500-1800°C [7]. To have the optimal temperature in the melting furnace, an amount of fossil fuel is required (natural gas, oil etc.). The fossil fuel used generates carbon dioxide emissions.

One method to reduce the amount of  $CO_2$  emissions generated by glass production consists of the integration of CCS technology (Capture and Storage  $CO_2$ ). More technologies for the separation the carbon dioxide were developed in the

last years. The most developed technology for separating the  $CO_2$  is the chemical absorption process [8], [9]. The main advantage of this technology is that it can be applied to separate the carbon dioxide provided from many sources, as the  $CO_2$  from fossil fuel combustion, the  $CO_2$  from different industries (glass, cement, iron, etc.). The main drawback of the chemical absorption process using amines consists of the thermal heat required for solvent regeneration [10]. In the case, the chemical absorption process is integrated into a power plant, the steam required for solvent regeneration is taken by low pressure body turbine leading to a decrease in the global efficiency with 10-20 % [11], [12]. In the glass production, the waste gases have a high temperature. The waste gases are cooled down to 50 °C, the temperature of the chemical absorption process. The heat of the waste gases can be used to cover the heat required for regeneration the chemical solvent [13].

In this article, we studied the post-combustion integration of chemical absorption process using monoethanolamide (MEA) into a glass factory with recovery the heat of the waste gases from glass production, to supply the necessary heat for chemical solvent regeneration.

# 2 METHOD

The highest emissions potential in the atmospheric air of the raw and auxiliary materials is in the melting phase [3]. The pollution substances are generated due to the following processes: the melting of raw and auxiliary materials introduced in the furnace and the combustion of fossil fuels (oil or natural gas) to obtain the heat required for melting of the raw materials. The carbon dioxide is generated from the fossil fuels combustion process and from the decomposition of carbonates of the batch materials (sodium carbonate and limestone). In the real melting phase, the following processes take place [3]:

- The evacuation of the free water contained in the mixture (the moisture content of the mixture);
- The decomposition of carbonates, sulfates and nitrates (with forming the gaseous compounds such as CO<sub>2</sub>, SO<sub>3</sub>, SO<sub>2</sub>, N<sub>2</sub> and O<sub>2</sub>) as well the elimination of crystallization water from raw materials containing it;
- The formation of liquid phases by melting some components, including the eutectics that can result from the initial mixture or from certain local separations of its components;
- The mutual dissolution of the liquid phases formed with obtaining a glass melting without crystals but still containing gases;
- The volatilization of some components of the glass and mixture;
- The dissolution of gases in liquid glass (fuel, air, flue gases, CO<sub>2</sub>, SO<sub>2</sub>, etc. from raw materials).

As it is observed, the carbon dioxide emissions are generated only in two processes.

The determination of the emissions resulted of technological processes from glass industry can perform in two methods: the first method by direct determination, continuous measurement and monitoring, and the second method is by calculation using the emission factors. In this study, we used the second method by calculation the amount of the carbon dioxide generated in the manufacturing glass process.

#### 2.1 Determination of the CO<sub>2</sub> emissions by mass balance

In this section, we calculated the  $CO_2$  emissions produced at household glass manufacturing. In Tables 1 and 2 there are presented the compositions of the raw materials used to produce glass and the oxide composition of the glass resulted. The amount of raw materials considered was of 100 kg. In addition, the shards (30 kg) and the water, as moisture (1.28 kg) was added at raw materials presented in Table 1. Thus, the total losses with the water and the decomposition gases are of 16.7 kg/100 kg<sub>glass</sub>.

#### Table 1: Raw materials

Raw materials	Gravel	Al(OH) <sub>3</sub>	CaMg(CO <sub>3</sub> ) <sub>2</sub>	CaCO <sub>3</sub>	Na <sub>2</sub> CO <sub>3</sub>	NaNO <sub>3</sub>	K <sub>2</sub> CO <sub>3</sub>	Sb <sub>2</sub> O <sub>3</sub>	Total
Amount, [kg]	51.17	1.39	12.96	1.09	16.04	2.3	0.51	0.14	85.6
Losses, [kg]	0	0.48	6.19	0.48	6.66	1.46	0.16	0	15.43

#### Table 2: Glass oxide composition

SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	CaO	MgO	Na <sub>2</sub> O	K <sub>2</sub> O	$Sb_2O_3$	Shards	Total
[%]	[%]	[%]	[%]	[%]	[%]	[%]	[%]	[%]
51.17	0.91	4.55	2.8	10.08	0.35	0.14	30	100

The reactions that take place in the manufacturing of glass are shown in Table 3. As can be seen the  $CO_2$  emissions are produced at the raw materials decomposition. Based on the chemical reactions listed in Table 3 and the amounts of the raw materials listed in Table 1, the amounts and volumes of the gases and water vapors that are produced by melting can

be calculated using the stoichiometric calculation. The results obtained for the household glass defined are presented in the Table 4.

**Table 3:** Reactions of glass production [14]

Raw materials decomposition	Glass formation	Clearing glass
$\begin{split} & \text{Na}_2\text{CO}_3 \rightarrow \text{Na}_2\text{O} + \text{CO}_2 \\ & 2\text{Na}\text{NO}_3 \rightarrow \text{Na}_2\text{O} + \text{NO}_2 + \text{NO} + \text{O}_2 \\ & \text{K}_2\text{CO}_3 \rightarrow \text{K}_2\text{O} + \text{CO}_2 \\ & \text{Ca}\text{CO}_3 \rightarrow \text{Ca}\text{O} + \text{CO}_2 \\ & \text{Ca}\text{Mg}(\text{CO}_3)_2 \rightarrow \text{Ca}\text{O} + \text{Mg}\text{O} + 2\text{CO}_2 \\ & 2\text{Al}(\text{OH})_3 \rightarrow \text{Al}_2\text{O}_3 + 3\text{H}_2\text{O} \end{split}$	$\begin{array}{l} 2Na_2O + CaO + 3SiO_2 \rightarrow \\ 2Na_2O.CaO.3SiO_2 \\ Na_2O + 2SiO_2 \rightarrow Na_2O.2SiO_2 \\ Na_2O + 2CaO + 3SiO_2 \rightarrow \\ Na_2O.2CaO.3SiO_2 \\ Na_2O + 3CaO + 6SiO_2 \rightarrow \\ Na_2O.3CaO.6SiO_2 \end{array}$	$Sb_2O_3 + O_2 \rightarrow Sb_2O_5$ - which takes place at the nitrates decomposition with the oxygen released by them, and at temperatures above 1200 °C, the reaction takes place: $Sb_2O_5 \rightarrow Sb_2O_3 + O_2$ - releasing oxygen, which helps refining glass.

Table 4: Gases resulting by melting stages

Component	$CO_2$	NO <sub>2</sub>	NO	O <sub>2</sub>	H <sub>2</sub> O
Amount, [kg]	13.49	0.62	0.41	0.43	1.28
Volume, [m <sup>3</sup> <sub>N</sub> ]	6.9	0.3	0.3	0.3	2.2

The fuel used for generated the heat required in the melting furnace contains  $CH_4$  in volumetric concentration of 96.96 %. The low heating value of fuel is of 8813.72 kcal/m<sup>3</sup> and the  $CO_2$  emission factor is of 55.75 t<sub>CO2</sub>/TJ. The results for household glass melting furnace are presented in Table 5.

Table 5: Results melting furnace

Parameter	Value				
Time cycle, [h]	0.8				
Number batches, [batch/24 h]	72				
Combustion air flow in melting furnace, [m <sup>3</sup> <sub>N</sub> /h]	4100				
Fuel consumption in melting furnace, $[m_N^3/h]$	285				
Flue gases composition from melting furnace, [%]	$CO_2$	$O_2$	СО		
File gases composition from menting furnace, [%]	8	6	0.55		
Glass flow extracted, [kg/h]	972				
Flue gases flow from melting furnace, [m <sup>3</sup> /h]	28000				
Flue gases temperature from melting furnace, [°C]	1470				

The calculation of the process emissions [2]:

$$m_{CO2}^{mp} = FE_{mp} \times m_{gl} \tag{1}$$

where:  $m_{CO2}^{mp}$  - is the CO<sub>2</sub> emissions from the melting process, [kg<sub>CO2</sub>/h];  $FE_{mp}$  - is the CO<sub>2</sub> emission factor (determined based on stoichiometric chemical reactions), [kg<sub>CO2</sub>/t<sub>glass</sub>];  $m_{st}$  - is the product of glass, [t<sub>glass</sub>/h].

The calculation of the combustion emissions [2]:

$$m_{CO2}^{fuel} = FE_{fuel} \times LHV_{fuel} \times m_{fuel}$$
(2)

where:  $m_{CO2}^{fuel}$  - is the CO<sub>2</sub> emissions from fuel combustion, [kg<sub>CO2</sub>/h];  $FE_{fuel}$  - is the CO<sub>2</sub> emission factor of fuel (determined on the basis of elementary/chromatographic analysis), [kg<sub>CO2</sub>/ MJ];  $LHV_{fuel}$  - is the low heating value of fuel, [MJ/m<sup>3</sup><sub>N</sub>];  $m_{fuel}$  - is the fuel consumption, [m<sup>3</sup><sub>N</sub>/h].

The CO<sub>2</sub> total emissions are obtained by summing up the process emissions and the combustion emissions:

$$m_{CO2}^{total} = m_{CO2}^{mp} + m_{CO2}^{fuel} \tag{3}$$

The CO<sub>2</sub> total emissions was calculated using Eq. 1-3 and was obtained the following results: the amount of CO<sub>2</sub> from melting stage is of 134.9 kg<sub>CO2</sub>/h, the amount of CO<sub>2</sub> from fuel combustion is of 586 kg<sub>CO2</sub>/h, and the total amount of CO<sub>2</sub> is of 720.9 kg<sub>CO2</sub>/t<sub>glass</sub>, the all results obtained are for one tone of glass produced.

## 2.2 Chemical absorption process description

In this section, we studied the integration of chemical absorption process in the glass production. The chemical absorption processes consists by the passing the gases that contains  $CO_2$  by an absorption unit. The waste gases from the glass production are introduced in the absorption unit at the bottom and the chemical solvent at the top. The chemical solvent consists of water and one type of amine. In this study, we used monoethanolamide in different concentration in the chemical solvent. After the chemical absorption process takes place, the chemical solvent rich in  $CO_2$  get out at the bottom of the absorption unit. The gases treated leaves the absorption unit at the top. The regeneration of the chemical solvent occurs in the desorption unit at of temperature of 120 °C [15]. In this work, the thermal heat required for solvent regeneration is taken from the heat of waste gases. After the desorption process, the pure carbon dioxide is evacuated at the top of desorption unit to be compressed and storage. The chemical solvent lean in  $CO_2$  is recirculated, but not before to transfer a part of its heat to the chemical solvent rich in  $CO_2$ . The pressure of the chemical solvent is maintained by using pumps. The process was simulated in the software Chemcad 6.01. The thermodynamic packages used for simulation the processes (glass production and chemical absorption process) were SRK (Soave-Redlich-Kwong) and Amine packed. In figure 2 is presented the scheme used in Chemcad program.

The gases from the glass manufacturing and the combustion fuel have a high temperature, approximately 1700 °C. The optimal temperature for the chemical absorption is of 50 °C [16], therefore the gases was cooled up to this temperature. It can see that the heat of the gases resulted from process was used to produce power, by two steam turbine, one of medium pressure and one of low pressure. A part of the steam produced was taken from the low pressure steam turbine at five bar for regeneration of the chemical solvent. The  $CO_2$  capture efficiency was maintained of 90 % and the weight concentration of MEA was varied, also it was varied and the lean loading solvent to determine the effect on the power produced.

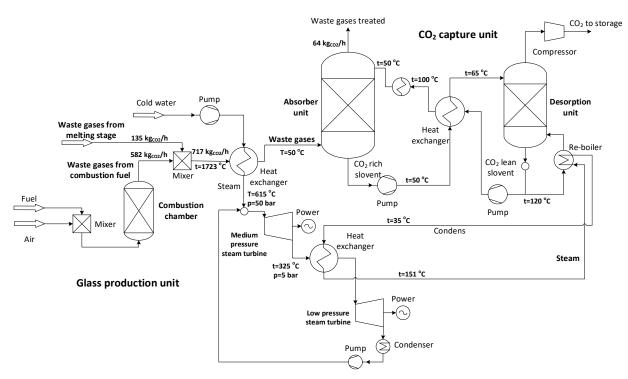


Figure 2: Simulation of the glass process and the chemical absorption process in Chemcad [17]

The thermodynamic efficiency of the cycle before and after the integration of the chemical absorption process was calculated using the following equation:

$$\eta_t = \frac{P_t \cdot \eta_m \cdot \eta_g}{Q_{fuel} \cdot Q_{mp}} \cdot 100 \tag{4}$$

where:  $\eta_t$  - is the thermodynamic efficiency of the cycle, [%];  $P_t$  - is the power produced of the steam turbines, [kW];  $\eta_m$  - is the mechanical efficiency, 98.5 %;  $\eta_g$  - is the steam generator efficiency, 99.1 %;  $Q_{fuel}$  - is the heat of the fuel, [kJ/s];  $Q_{mn}$  - is the heat of the gases from melting process, [kJ/s];

Also, it was calculated the  $CO_2$  emission factor before and after integration of the separation technology like as rapport between the amount of the  $CO_2$  released in the atmosphere and the power produced of the cycle.

#### **3 SIMULATION RESULTS AND DISCUSSION**

In this study, we analyzed the integration of the chemical absorption process in the glass production. The amount of the carbon dioxide generated from the glass process resulted by simulation of the process was validated with the results obtained based on the calculation of  $CO_2$  emissions by mass balance. The amount of carbon dioxide from fuel combustion is higher than the amount of carbon dioxide from raw materials decomposition. The molar concentration of  $CO_2$  in the gases from fuel combustion is of 7.8 %, after the gases are mixed with the waste gases from melting boiler, the molar concentration increases up to 9.4 %.

The performances of the chemical absorption process depend on the established value of more parameters. In this study, for the CO<sub>2</sub> capture efficiency of 90 %, and for all absorption process parameters established all cases studied have been compared. The weight concentration of MEA in the chemical solvent was varied, respectively we considered three cases: MEA in weight concentration of 10, 20 and 30 %. The lean loading solvent is an important parameter of the chemical absorption process, the value of these influences the amount of heat required for solvent regeneration. In the specialty literature, the value of the lean loading solvent is of 0.21 kmol<sub>CO2</sub>/kmol<sub>MEA</sub> [11]. In our study, the lean loading solvent was varied between 0.16-0.24 kmol<sub>CO2</sub>/kmol<sub>MEA</sub>. The heat duty for chemical solvent regeneration was between 1400-4000 MJ/h. In Table 6 there are presented the main results obtained for the chemical absorption process.

Glass production			
Flow fuel, [kg/h]	215		
Flow air, [kg/h]	4500		
Excess air, $\lambda$ , [-]	1.32		
Glass produced, [kg/h]	972		
Temperature of waste gases, [°C]	1723		
CO <sub>2</sub> from melting stage, [kg/h]	135		
CO <sub>2</sub> from fuel combustion, [kg/h]	582		
$CO_2$ total, [kg/h]	717		
Power, [kW]	1027		
CO <sub>2</sub> emission factor, [kg/MWh]	700		
Thermodynamic efficiency, [%]	31		
Chemical absorption process			
Temperature of chemical absorption process, [°C]	50		
Temperature of desorption process, [°C]	120		
Waste gases flow, G, [kg/h]	4878		
Chemical solvent flow, L, [kg/h]	6000-17000		
$L/G$ ratio, $[kg_{w.g}/kg_{c.s}]$	1.23-3.32		
MEA wt. concentration, [%]	10-30		
CO <sub>2</sub> lean loading solvent, [mol <sub>CO2</sub> /mol <sub>MEA</sub> ]	0.16-0.24		
Heat duty for solvent regeneration, [MJ/h]	1400-4000		
Specific heat duty for solvent regeneration, [GJ/t <sub>CO2</sub> ]	2-6		
Power, [kW]	750-910		
CO <sub>2</sub> emission factor, [kg <sub>CO2</sub> /MWh]	70-85		
Thermodynamic efficiency, [%]	22-27		
Efficiency penalty, [%]	11-26		

**Table 6:** The main results for chemical absorption process

The ratio between the chemical solvent flow and the waste gases flow was calculated for the three cases studied. The lowest value of the L/G ratio was obtained for the 30 % wt. MEA (Figure 3) of 1.23. With the decreasing of the weight concentration of MEA in the chemical solvent, the L/G ratio increased. However, the weight concentration of MEA is limited, since in higher concentration of 30 % the monoethanolamine is corrosive [18]. In Figures 3 and 4 there are represented the heat duty and the specific heat duty according to the  $CO_2$  lean loading solvent. The heat duty for solvent regeneration increases with the  $CO_2$  lean loading solvent decreases due to the amount of chemical solvent used for to maintain the CO2 capture efficiency of 90 %. The lowest value for the heat duty was obtained for a MEA 30 wt. % and

for a CO<sub>2</sub> lean loading solvent of 0.24 kmol<sub>CO2</sub>/kmol<sub>MEA</sub>. The specific heat duty represents the thermal energy required for capturing one ton of CO<sub>2</sub>. The results obtained of the specific heat duty for the cases studied are between 2-6 GJ/t<sub>CO2</sub>. The value of the specific heat duty for the case MEA wt. 30 % and CO<sub>2</sub> lean loading solvent of 0.22 mol<sub>CO2</sub>/mol<sub>MEA</sub> is of 2.21 GJ/t<sub>CO2</sub>.

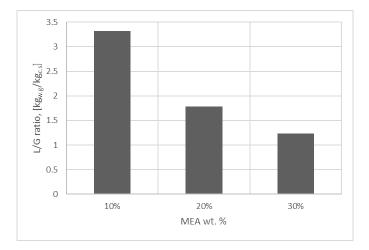


Figure 3: The L/G ratio according to the weight concentration of MEA

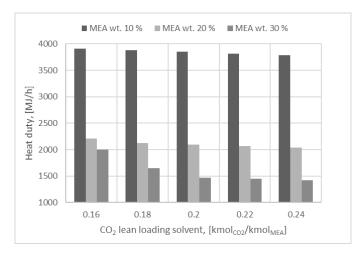


Figure 4: The heat duty according to the CO<sub>2</sub> lean loading solvent and the weight concentration of MEA

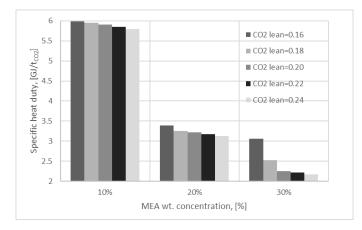


Figure 5: The specific heat duty according to the weight concentration of MEA and the CO<sub>2</sub> lean loading solvent

The thermal heat required for chemical solvent regeneration is provided by the low pressure steam turbine. The steam is extracted at pressure of 5 bar and at temperature of 151  $^{\circ}$ C. In all cases studied the heat produce of turbines covers the demand of heat for desorption process. In Figure 6 is represented the thermodynamic efficiency after the chemical absorption process was integrated. Without the CO<sub>2</sub> capture technology the power produced is of 1 MW after the integration of chemical absorption process the thermodynamic efficiency of cycle decreases with 11-26 percentage points

according to the amount of steam extracted for chemical solvent regeneration. The total  $CO_2$  emissions factor without carbon dioxide capture for glass process is of 700 kgCO<sub>2</sub>/MWh of which 80 % is from fuel combustion. For a high concentration of MEA in the chemical solvent the  $CO_2$  emission factor is lower (Figure 7) because the amount of the  $CO_2$  released in atmosphere is reported of the total power produced.

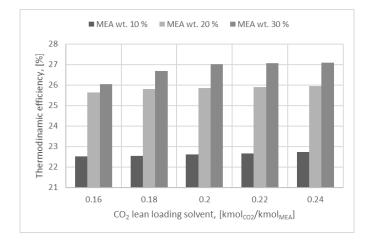


Figure 6: The thermodynamic efficiency according to the concentration of MEA and the CO<sub>2</sub> lean loading solvent

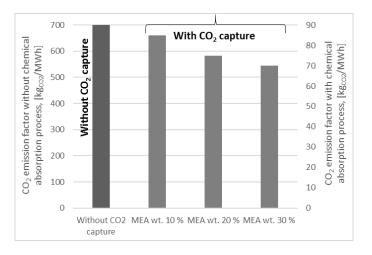


Figure 7: The CO<sub>2</sub> emission factor according to the weight concentration of MEA

# **4 CONCLUSONS**

In this study, we analyzed from a technical point of view the integration of chemical absorption process in glass production for reducing the carbon dioxide emissions. The glass manufacturing is a consuming energy process. For produced the glass in the melting furnace is required a high temperature that is provided by fossil fuel combustion. The carbon dioxide emissions are generated of fuel combustion and raw materials decomposition in the melting phase. The chemical absorption process was analyzed according to the weight concentration of monoethanolamine in the solvent and the  $CO_2$  lean loading solvent. The L/G ratio is higher for a smaller concentration of MEA in the solvent. For a 30 % weight concentration of MEA the L/G ratio obtained was 1.23 kgw.g/kgc.s. The heat duty was lower for a higher concentration of MEA due to the amount of the chemical solvent. The heat of waste gases was used to provide the heat duty for solvent regeneration thus reducing the integration impact of capture technology. The optimal case in terms of the thermal energy consumption was obtained for a wt. 30 % MEA and for a CO<sub>2</sub> lean loading solvent of 0.24 mol<sub>CO2</sub>/mol<sub>MEA</sub>.

The  $CO_2$  capture technology by chemical absorption process is a promising method for separating the  $CO_2$  emissions from glass production if the heat of waste gases is recovered and used in a steam turbine cycle. The thermal heat produced of the steam turbine cycle covers the necessary heat in desorption unit. In addition, the remaining energy surplus after the solvent regeneration can be sold to cover a part of the costs with integration of the  $CO_2$  capture technology and the steam turbines. From this point of view, an economic analysis is needed that we propose to do in future works.

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