

OPTIMIZAREA UTILIZĂRII MATERIALELOR ABSORBANTE ÎN TEHNOLOGIA DE CAPTARE A CO₂ POST-COMBUSTIE AFERENTE DIFERITELOR PROCESE TEHNOLOGICE (CIMENT, STICLĂ)

OPTIMISATION OF UTILISATION OF ABSORBENT MATERIALS FOR CO₂ POST-COMBUSTION CAPTURE TECHNOLOGY FOR DIFFERENT INDUSTRIAL PROCESSES (CEMENT, GLASS)

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The aim of the paper consists in optimising the integration of CO₂ post-combustion capture technology using chemical absorption within the fluidised circulating coal combustion (FCCC) with direct application in glass and cement industries. The authors have performed some comparison of the effects of using primary amines (MEA) and tertiary amines (TEA) in order to reduce energy consumption of regeneration process, and, at the same time, keeping the efficiency of CO₂ capture over 85 %. The paper presents results of an experimental study using laboratory equipment for fluidised circulating coal combustion, and also simulation results of CO₂ post-combustion capture using specialised software HYSYS 3.2.

Obiectivul lucrării constă în optimizarea integrării proceselor de captare a CO₂ post-combustie prin absorbție chimică în cadrul tehnologiei de ardere a cărbunelui în strat fluidizat circulant (ASFC) cu aplicare directă în industriile sticlei și cimentului. Se compară efectele utilizării aminelor primare (MEA) și terțiare (TEA), pentru reducerea consumului energetic aferent procesului de regenerare, menținând o eficiență de peste 85% a procesului de captare a CO₂. Lucrarea se bazează atât pe realizarea unui studiu experimental utilizând instalația de ardere în strat fluidizat circulant cât și pe rezultatele obținute în urma simulării procesului de captare a CO₂ post-combustie utilizând programul HYSYS 3.2.

Keywords: solid electrolytes, LSGM, microwave technique, IT-SOFC.

1. Introducere

Cement industry is responsible for about 5 % of total global CO₂ emissions resulted from production activities and for about 3 % of global emissions generated by human activities. The cement production process leads to generating about 0.73-0.99 kg CO₂ per kg of produced cement. CO₂ emissions of cement industry come from: fabrication process (40 %), transport (5 %) and combustion of fossil fuels for electricity used within the process (5 %). The rest of emissions (about 50 %) come from decarbonation of limestone [1 - 2].

In perspective, up to 2020 there is intended to reduce CO₂ emissions per tonne of cement with about 30 %, as shown in Table 1.

Another industrial process directly responsible for great quantities of CO₂ emissions is glass manufacture. The greatest air pollutant is represented by the melting phase of raw and auxiliary materials in glass furnaces. The pollutant emissions, generally, appear due to the following

processes: a. combustion of fossil fuels in furnaces; b. melting phase of raw and auxiliary materials in glass furnaces.

The phenomena directly responsible for CO₂ generation during the melting phase include decomposition of carbonates, such as soda and limestone [3 - 4].

From data presented in Table 2 there can be shown the importance of CO₂ emissions resulted from the combustion process used within production of two types of glass [5].

Nowadays, there are three types of solutions for reducing emissions of carbon dioxide, thus reducing Green House Gasses (GHC): partial and progressive replacement of fossil fuels with renewable energy sources and raw materials, partially or totally, with some with low or zero greenhouse gas ; increasing the efficiency of utilisation of energy resources; integration of carbon capture, transport and storage technologies within energy processes. Unfortunately, none of these solutions can entirely solve the problem of

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Tabelul 1

Technical potential for CO₂ emissions reduction per tonne of cement up to 2020 [2]
Potențialul tehnic de reducere a emisiilor de CO₂ pe tona de ciment, până în anul 2020 [2]

Area / Domeniul îmbunătățirii	Measure / Denumire măsura	Weight Pondere
Process emissions / Emisii de proces	Adding cements / Cimenturi cu adaos	7%
Emissions from fossil fuels combustion Emisii rezultate din arderea combustibilului	Efficiency of combustion Changing the fuel type Eficiența arderii	11%
	Schimbarea tipului de combustibil	3%
Transport /Transport	Transport efficiency and transport using biomass Eficiența transportului și transport cu biomasă	sub 1% under
Power generation Generare de electricitate	Energy efficiency and power generation with low carbon emissions Eficiență energetică și generarea de energie electrică cu emisii scăzute de carbon	sub 1% under
Other / Alte reduceri	Alternative fuels / Combustibili alternativi	12%
Total	All measures / Toate măsurile	cca 30% about

Tabelul 2

CO₂ emissions for glass production / Emisii CO₂ la fabricarea sticlei

CO ₂ emission type Tip emisie CO ₂	Quantity (kg/t glass) / Cantitate (kg/t sticlă)	
	Household glass Sticlă de menaj	Optical glass Sticlă optică
Process emissions/ Emisii de proces	140	87
Combustion emissions/ Emisii de ardere	590	8276.6
Total emissions / Emisii totale	730	8363.6

reducing CO₂ emissions, and, thus, allowing further use fossil fuels within the sustainable development.

In both industrial process (cement and glass production) a significant quantity CO₂ emissions come from fossil fuel combustion used within technological processes.

In the present paper the authors have concentrated upon CO₂ separation stage from flue gasses resulted from coal combustion. The main objective of the paper consists in reducing energy consumption needed for regeneration process of different types of primary and tertiary amines solvents, keeping at the same time high level of efficiency of CO₂ capture. The energy demand of regeneration process of solvent is influenced by three factors, each of them corresponding to an endothermic process: heat demand for increasing the temperature of solvent in the desorption unit, heat demand for breaking the link between CO₂ and nitrogenous compounds and heat demand for water vaporisation [6].

Carbon capture and storage, (CCS) technology includes the following stages: CO₂ separation from flue gasses evacuated from combustion equipment and further capturing in a tank; compressing CO₂ up to supercritical stage so it can be transported to the storage site; and finally, CO₂ storage in different sites (depleted hydrocarbon deposits, artificial deposits, etc.).

It is well known that the major technical disadvantage of CO₂ separation process is due to high energy demand, as shown in literature references [7-10]. A feasible solution to solve this problem represents utilisation of solvents with high capacity of CO₂ retention from flue gasses and with low energy consumption for regeneration process;

such experiments have been successfully performed in different countries [11-13].

In [14-16] there have been shown that use of primary, secondary and tertiary amine solvents leads to significant decrease of energy consumption for regeneration process; the results of studies show that for efficiencies of CO₂ capture process of 80 %, 85 %, 90 % and 95 % the minimum energy consumption for regeneration process is obtained for equal mixture of methyl diethanolamine (20% MDEA) and diethanolamine (20% DEA) and water (60% H₂O), these being 2.3, 2.4, 2.7; and respectively 2.95 GJ/tonne CO₂ [17 - 18].

Establishment of optimal conditions for CO₂ capture using chemical absorption with amines consists in mixture process and in choosing the right proportion for combining different types of amines. Thus, the optimal proportions for the maximal absorber efficiency for MDEA and DEA are: 40% MDEA with 10% DEA, 30% MDEA with 10% DEA and 40% MDEA with 5% DEA; this has also been studied in [19].

The results presented by authors have been obtained through an experimental study within fluidised circulating coal combustion (FCCC) equipment and also through a simulation study using specialised software Hysys 3.2. Utilisation of specialised simulation software has the advantage of analysing different cases that can be hardly reproduced in experimental study, as have been shown in papers, ASPEN Plus [20-22], Aspen HYSYS [23 - 24] Hysys [25], UniSim Design Suite of Honeywell [26], HYSYS 7.1 [27], ProTrea (Optimized Gas Treating) [16].

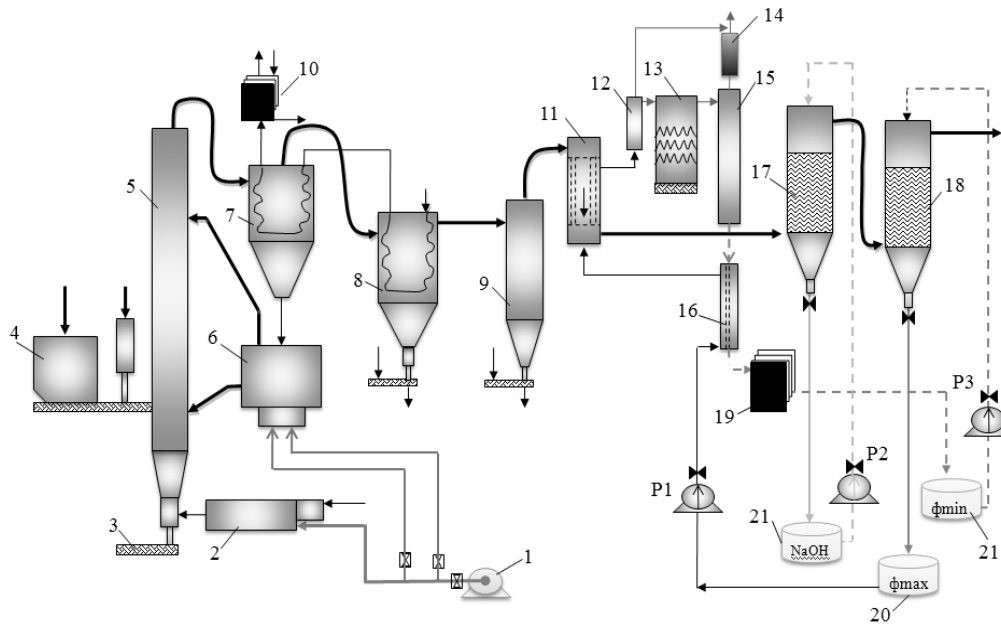


Fig.1 - Scheme of the fluidised circulating combustion unit / Schema instalației de ardere în strat fluidizat circulant

1. Blower; 2. Pre-firing using natural gas; 3. Ash evacuation screw conveyor; 4. Fuel feeding system; 5. Combustion chamber; 6. Recirculation system; 7. Cyclone; 8. Convective heat exchanger; 9. Cyclone; 10. Heat exchanger; 11. Flue gases –MEA heat exchanger; 12. CO₂ expansion tank; 13.Reboiler; 14. Condenser; 15. Desorption column; 16. Economisers; 17. Desulphurization unit; 18. Absorbing column; 19. H₂O-MEA heat exchanger; 20. Rich MEA solution tank; 21. NaOH solution tank; 22. Lean MEA solution tank; P1.Pump φ max; P2.Circulation pump NaOH &H₂O; P3.Pump φ min.

1.Ventilator; 2.Antefocar; 3.Șnec evacuare cenușă; 4.Buncăr cărbune/biomasă; 5.Focar; 6.Dispozitiv recirculare cenușă; 7.Ciclon recirculare cenușă; 8.Corp convectiv; 9.Ciclon; 10.Schimbător de căldură; 11.Schimbător de căldură MEA-gaze de ardere; 12.CO₂ Vas de detentă; 13.Reboiler; 14.Condensator; 15.Coloana desorbție; 16.Economizor; 17.Coloană desulfurare gaze; 18.Coloană de absorbție; 19.Răcitor; 20.Rezervor soluție MEA bogată; 21.Rezervor NaOH;22.Rezervor soluție MEA regenerată; P1.Pompă φ max; P2.Pompă circulație NaOH &H₂O; P3.Pompă φ min .

2. Description of the experimental unit

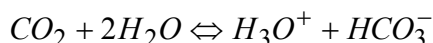
Figure 1 shows the experimental unit for fluidised circulating coal combustion (FCCC) used for the study. The experimental unit installed at the laboratory of Renewable Energy Sources at the Power Engineering Faculty of University Politehnica of Bucharest allows performing experiments for fossil fuel combustion, renewable fuels combustion and co-combustion.

The experimental unit is equipped with a desulphuring equipment (12) used for reducing the concentration of sulphur oxides in flue gasses before entering the absorption equipment (13). Solution with lean CO₂ (15 – MEA, lean) is introduced in the absorption unit at upper part using a pump in a counter-current allowing complete CO₂ separation from flue gasses corresponding to chemical reactions of equilibrium CO₂-MEA-H₂O:

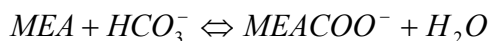
Dissociation of MEA:



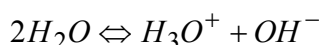
Bi-carbonate formation:



Carbamate formation:



Water hydrolysis:



3. Methodology used within the experimental and simulation study

There has been used local lignite for experimental study having the following characteristics, shown in Table 3.

Table 3

Fossil fuel characteristics
Compoziția combustibilului folosit

Lignite flow / Debit Lignit	10 kg/h
<i>Lignite composition / Compoziția lignitului</i>	
Hydrogen / Hidrogen	1.25%
Humidity / Umiditate	36%
Sulphur / Sulf	1%
Nitrogen / Azot	0.65%
Oxygen / Oxigen	2.55%
Carbon / Carbon	21.55%
Lower calorific value Puterea calorifică inferioară	7543 kJ/kg

Tabelul 4

Flue gasses composition
Compoziția gazelor de ardere rezultate în urma arderii combustibilului

CO ₂	11%
N ₂	72%
O ₂	7%
H ₂ O.	10%

There have been used two gas analysers TESTO 350 XL for determining flue gasses composition.

Regeneration of solution using amines has been performed in two stages: in the first stage there have been used flue gasses from the lignite combustion process for partial regeneration of the solvent through increasing the temperature of the solvent; in the second stage there have been used electric resistances for supplementary heating of amine solution up to the temperature corresponding to total regeneration.

Even though that it is known that primary amines, monoethanolamine (MEA) react very well with acid gasses and offer a very good CO₂ separation from flue gasses they have a limited capacity of absorption: for absorption of 1 mole of CO₂ there is needed two moles of amines, and they also have a high energy demand for regeneration between 3 and 5 GJ/tCO₂ [23]. Due to amine's high corrosive nature they are used in different mixtures with secondary and tertiary amines.

The utilisation of tri-ethanolamine for the absorption process has the advantage that regenerates very easy in proportion of 95 %, having also low energy consumption for regeneration. But, it is well known that this type of amine reacts with more difficulty with CO₂ needing different activators to mixture with such as primary amines, monoethanolamine and diglycolamine (MES, DGA) or piperazine (PZ) in order to increase speed reaction. Taking into consideration these characteristics of primary and secondary amines they have been used in different proportion in the present experimental and simulation research looking at variation of the following parameters:

- The effect of variation of regeneration energy as function of efficiency of CO₂ capture for different ratio of flows of liquid and flue gasses (L/G) ratio, [GJ/tCO₂].
- The influence of concentration and type of amine on regeneration energy of the solvent.
- Variation of regeneration energy of solution as function of efficiency of CO₂ capture process and lean CO₂ loading solvent, keeping constant concentration of TEA+MEA.

HYSYS 3.2 is a software application that allows simulation in dynamic regime and for equilibrium reactions of thermo-chemical processes. The software includes procedures for determination of chemical and physical properties of substances, elaboration of mass and energy balances, design of different equipment and optimisation of thermo-chemical processes.

For simulation with amines there have been chosen the Amine fluid package and Kent-Eisenberg, (non-ideal vapour phase), as a thermodynamic calculation model since they offer the best model accuracy. The software application requires a limited number of input values such as:

temperature, pressure and flow.

The efficiency of CO₂ capture process has been calculated as a ratio between the value of carbon dioxide from flue gasses at the input of the absorber and the value of the carbon dioxide from flue gasses at the output of the desorber.

$$\varepsilon_{CO_2} = \frac{CO_2^i(\text{vol}\%) - CO_2^e(\text{vol}\%)}{CO_2^i(\text{vol}\%)} \cdot 100 [\%] \quad (1)$$

Where: ε_{CO_2} - efficiency of CO₂ capture process

CO_2^i - carbon dioxide from flue gasses at the input of the absorber

CO_2^e - carbon dioxide from flue gasses at the output of the desorber

Lean CO₂ loading solvent represents a very important parameter for the capture process that through determination of the optimal value helps to determination of the maximum regeneration degree with minimal heat consumption. In order to determine the optimal value of lean CO₂ loading solvent there have been varied the solvent flow, the efficiency of CO₂ capture process and the temperature in the desorber through change the energy demand of the desorber, other parameters (solvent concentration, pressure in absorber/desorber) being constant.

$$\gamma = \frac{\text{mol } CO_2}{\text{mol } MEA + \text{mol } TEA} \quad (2)$$

Where: γ - Loading degree of lean/rich solution molCO₂/mol TEA+MEA

4. Results and discussions

4.1. Interpretation of experimental results

Figure 2 shows the variation of lean CO₂ loading solution having a mixture of tertiary and primary amines that had the role to increase the speed reaction. Adding the primary amine in proportion of 4 % MEA leads to increasing energy demand for regeneration of the solvent, since this type of amine needs more energy for regeneration process. For the case of efficiency of 85 % it has been observed that for a lean CO₂ loading solution with 0.16 molCO₂/mol TEA+MEA higher energy consumption with 46 % than for a concentration of 50 % TEA up to 2.15 GJ/tCO₂ [28].

Increasing the weight of MEA to 6 % in the solution mixture (30 %TEA+6 %MEA) led to increasing the energy demand for regeneration with 5.6 % compared to the previous case keeping the same value for lean CO₂ loading solution at 0.16 and for an efficiency of 85 %, (Figure 3). No matter the value of the efficiency of capture

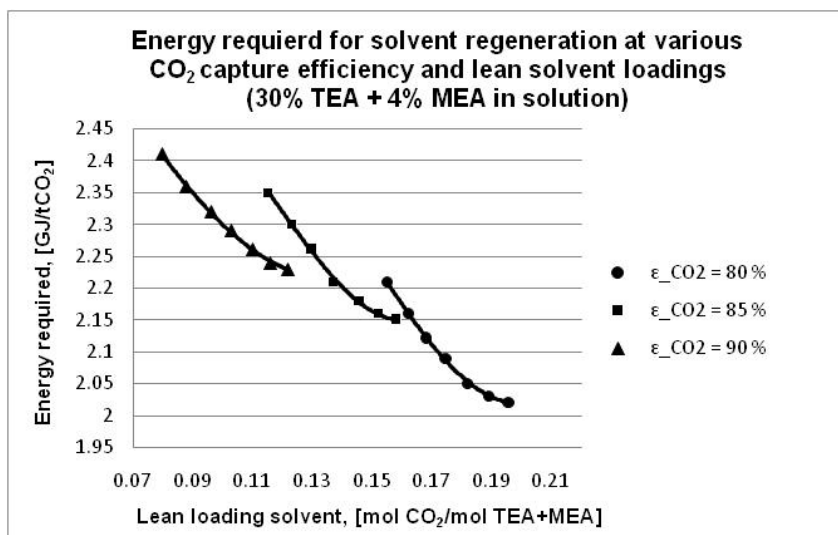


Fig.2 - Regenerative energy variation depending on efficiency CO₂ capture process and lean CO₂ loading solvent, keeping constant concentration of 30% TEA+4% MEA. / Variația energiei de regenerare a soluției în funcție de eficiența procesului de captare a CO₂ și de gradul de încărcare al soluției sărace în CO₂, menținând constantă concentrația de 30% TEA+4% MEA.

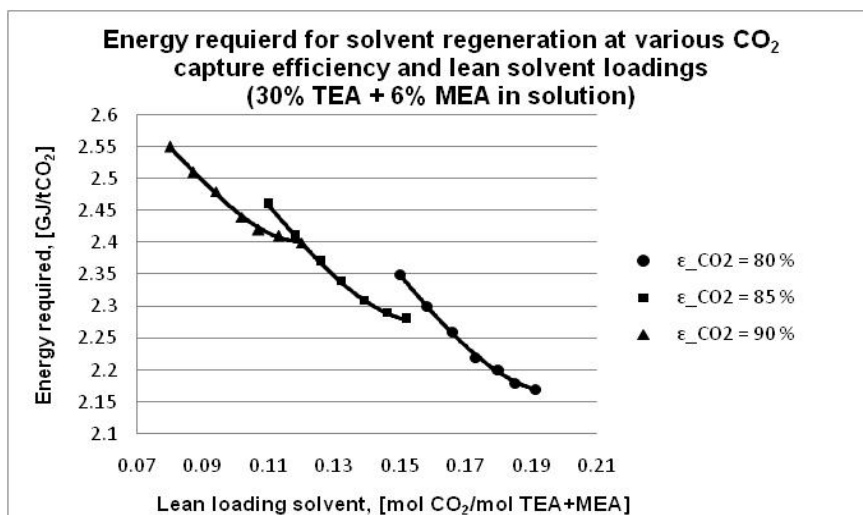


Fig.3 - Regenerative energy variation depending on efficiency CO₂ capture process and lean CO₂ loading solvent, keeping constant concentration of 30% TEA+6% MEA. / Variația energiei de regenerare a soluției în funcție de eficiența procesului de captare a CO₂ și de gradul de încărcare al soluției sărace în CO₂, menținând constantă concentrația de 30% TEA+6% MEA.

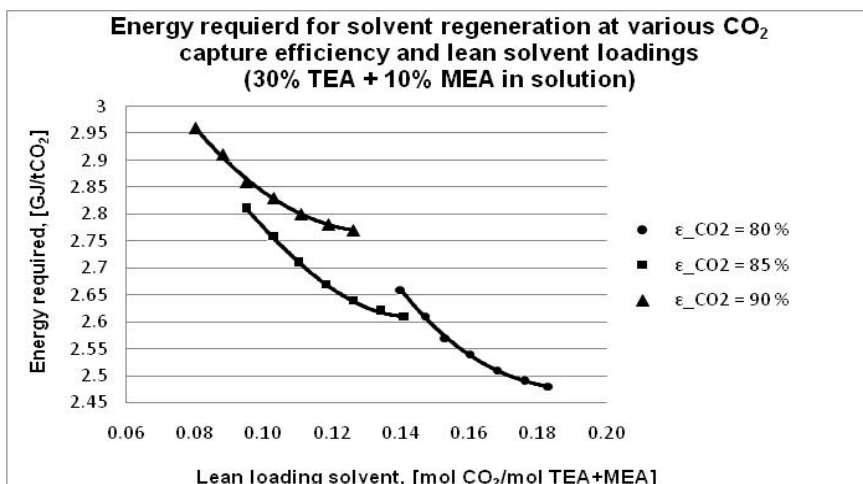


Fig.4 - Regenerative energy variation depending on efficiency CO₂ capture process and lean CO₂ loading solvent, keeping constant concentration of 30% TEA+10% MEA. / Variația energiei de regenerare a soluției în funcție de eficiența procesului de captare a CO₂ și de gradul de încărcare al soluției sărace în CO₂, menținând constantă concentrația de 30% TEA+10% MEA.

process for this case there can be observed an increase of energy consumption for solvent regeneration having values between 5.6 and 6.8 % compared to the case of concentration of 30 % TEA+4 % MEA.

For the case of increasing the proportion up to 10 % MEA (Figure 4) in the solution mixture

(30 %TEA+10 %MEA) in order to increase speed reaction there has been observed an increase with 13-15 % of energy demand for solvent regeneration, no matter the value of efficiency, for the same values of lean CO₂ loading solution compared to the previous case.

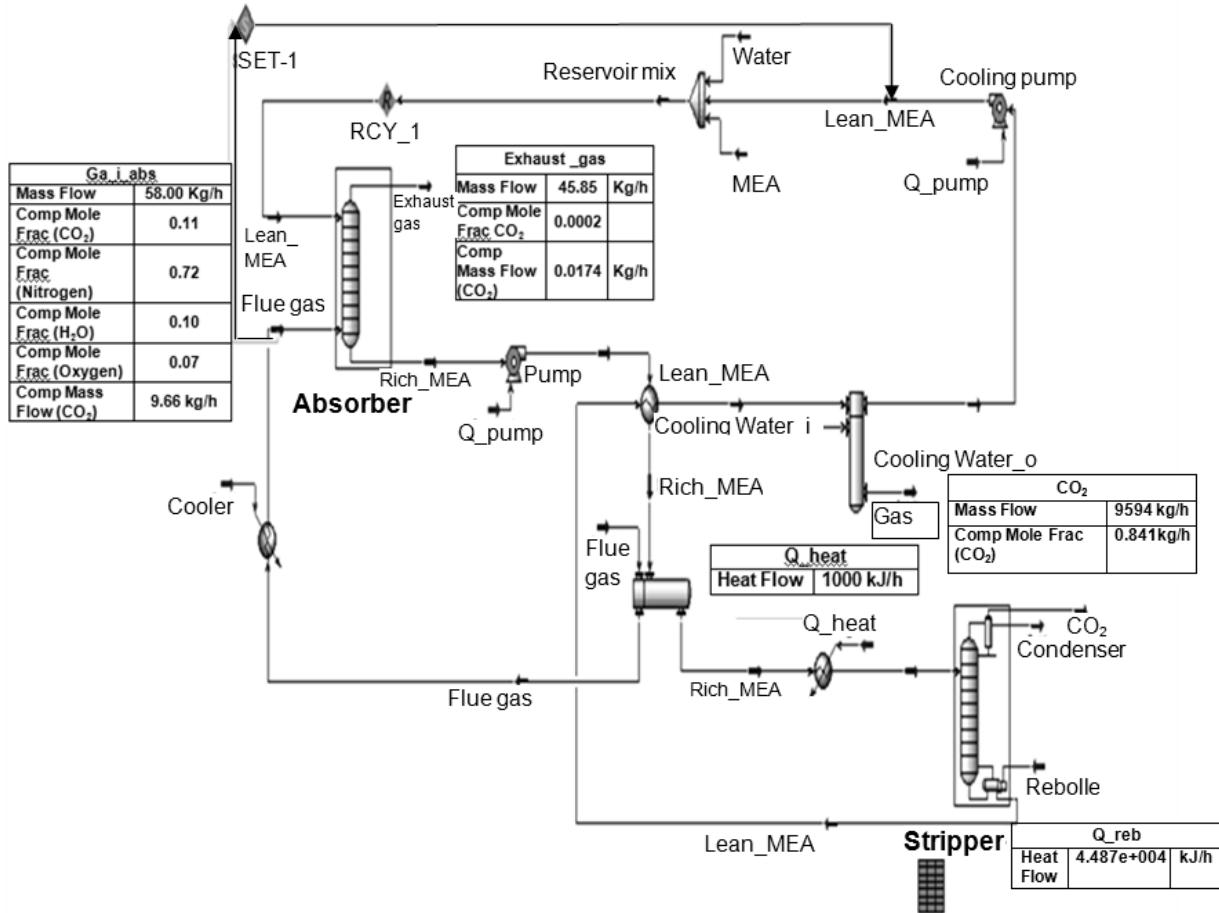


Fig.5 - Simplified scheme of CO₂ capture process elaborated in HYSYS 3.2./ Schema procesului de captare a CO₂ realizată în HYSYS 3.2.

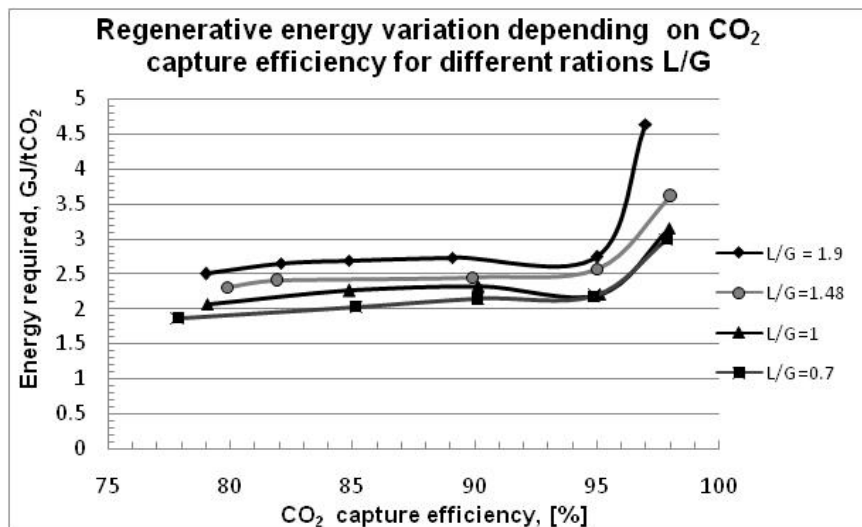


Fig. 6 - Regenerative energy variation depending on efficiency of CO₂ capture for different L/G ratios and keeping constant concentration of TEA30%+MEA5% / Variația energiei de regenerare în funcție de eficiența de captare a CO₂ pentru diferite rapoarte L/G și menținând constant un amestec de TEA30%+MEA5% .

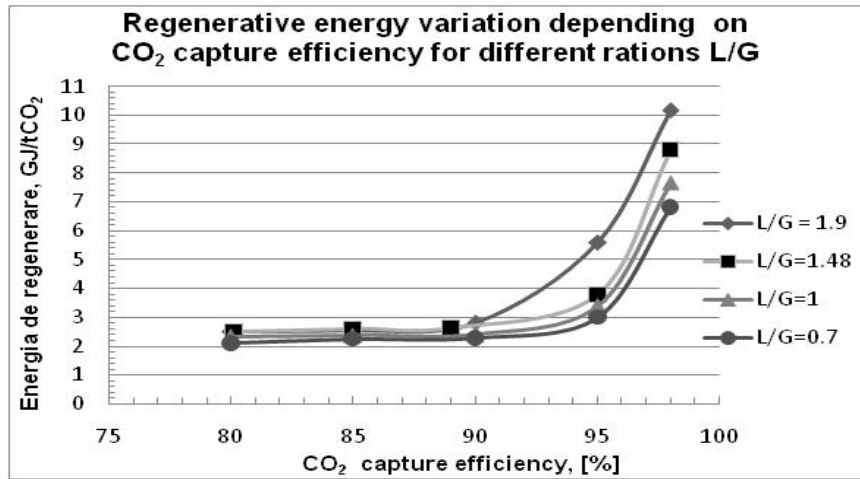


Fig.7 - Regenerative energy variation depending on efficiency of CO₂ capture for different L/G ratios and keeping constant concentration of TEA30%+MEA10%. / Variația energiei de regenerare în funcție de eficiența de captare a CO₂ pentru diferite rapoarte L/G și menținând constant un amestec de TEA30%+MEA10% .

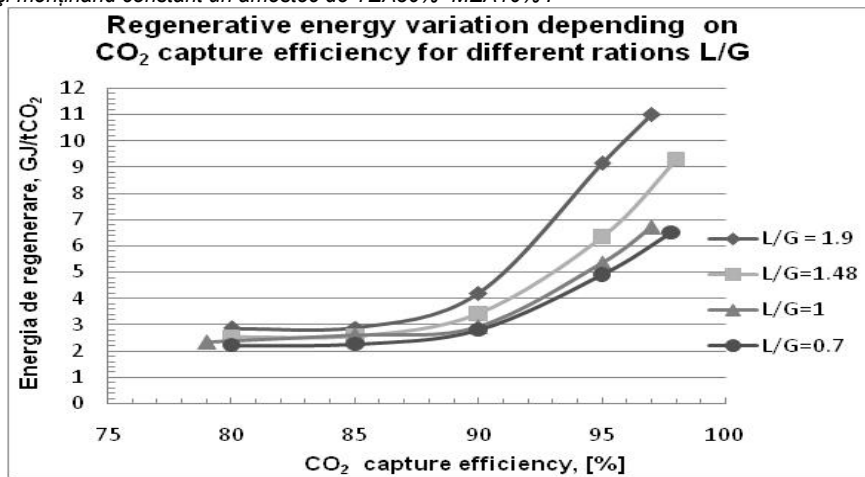


Fig.8 - Regenerative energy variation depending on efficiency of CO₂ capture for different L/G ratios and keeping constant concentration of TEA30%+MEA15%. / Variația energiei de regenerare în funcție de eficiența de captare a CO₂ pentru diferite rapoarte L/G și menținând constant un amestec de TEA30%+MEA15% .

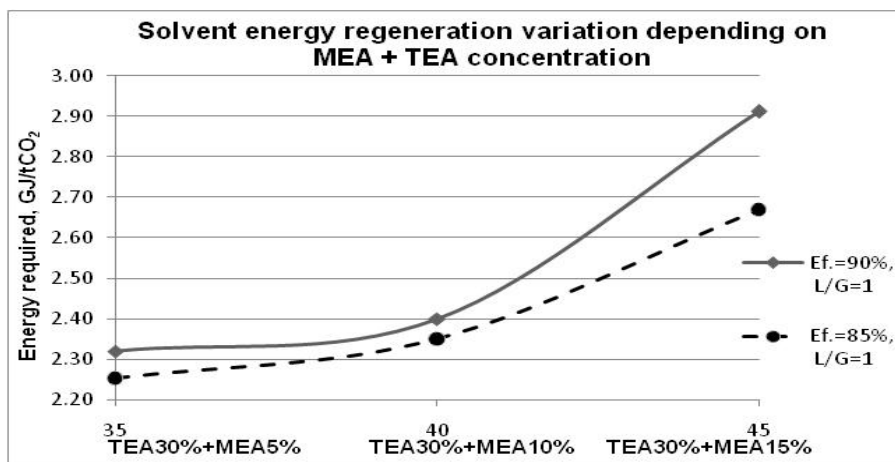


Fig.9 - Solvent energy regeneration variation depending on MEA+TEA concentration keeping constant L/G=1. / Variația necesarului de energie de regenerarea pentru diferite concentrații de TEA+MEA, menținând constant raportul L/G=1.

4.2 Interpretation of simulation results in HYSYS 3.2

The advantage of a simulation process is that it allows analysis of different cases difficult to obtain using experimental unit. The simulation of the entire CO₂ capture process has been performed through optimisation of absorption and desorption units modifying the number of stages and differential pressure. The elaborated scheme within the simulating software is presented in Figure 5.

For the case of mixture of 30 % TEA+5 % MEA, (Figure 6) there has been observed minimal energy consumption for a ratio L/G of 0.7 no matter the CO₂ capture process efficiency. For a capture efficiency of 85 % there has been observed minimal regeneration energy demand of 2 GJ/t CO₂ for the ratio L/G of 0.7. When increasing the ratio L/G to 1.9 it has been observed an increase of energy consumption for solvent regeneration with 40 % reaching 2.8 GJ/t CO₂.

For the case of increasing the weight of MEA 30 % TEA+10 % MEA (Figure 7) it has been observed a light tendency of increasing the energy demand for regeneration, thus the minimum energy demand for regeneration for the CO₂ capture process efficiency of 85 % is 2.3 GJ/t CO₂ for a ratio L/G of 0.7. For the case of efficiency higher than 93 % there has been observed a more significant increase of energy demand for regeneration.

Increasing the weight of MEA in the mixture 30 % TEA+15 % MEA (Figure 8) has shown a significant increase of energy demand for regeneration of solvent for efficiencies higher than 92 %. This tendency can be explained by high energy consumption for regeneration needed by MEA. L/G ratio for which energy demand for regeneration is minimal is between 0.7 and 1. In case of increasing CO₂ capture efficiency from 90 to 95 % the energy demand for regeneration increases with 66 % for the ration L/G of 1.

Figure 9 shows the influence of concentration and type of amine on the energy demand for regeneration of solvent, keeping constant L/G ratio to 1 and varying the efficiency of CO₂ capture process from 85 % to 90 %. As solvents there have been used mixtures of primary and tertiary amines (MEA+TEA).

There has been observed an increase of energy demand for regeneration with increasing the weight of primary amine in the mixture, which needs more energy for regeneration, with 5, 10 and 15 %. The increase of energy demand with 20% is observed for the case of CO₂ capture efficiency of 85 %, and with 25 % for the efficiency of 90 %, reaching energy consumption of 2.3 and respectively of 2.9 GJ/tCO₂. For the case of mixture 30 % TEA + 10 % MEA it has been observed an insignificant increase of energy demand for regeneration resulting the optimal solution.

5. Conclusions

The CO₂ post-combustion capture technology using chemical absorption based on solvents has as a main problem high energy demand for the regeneration process of solvents. Thus, the present paper tries to identify the optimal combination of two types of amine solvents, primary amines (MEA) and tertiary amines (TEA) in different proportions so the energy demand can be kept at minimal values and the efficiency of CO₂ capture process can be kept over 85 %.

For an efficiency of CO₂ capture process of 85 %, from the point of view of energy demand for regeneration, the optimal solution is a mixture of primary and tertiary amines 5 % MEA+30 % TEA, ratio L/G=0.75 [kg_l/kg_{gas}]. For this solution there has been obtained the minimal energy demand for regeneration of solvent of 2.15-2 GJ/tCO₂. The results have been obtained in the experimental study and simulation process using the software HYSYS 3.2.

Regarding the lean CO₂ loading solution, there can be said that more quantity of solution is loaded into the desorption unit helps to decreasing energy demand for regeneration through the supplementary heat generated due to water vaporisation from the solution, helping to breaking the bonds between CO₂ and amine. At the same time there should be mentioned that a greater quantity of solution needs greater energy quantity for heating up the solution. The experimental study led to the optimal value of lean CO₂ loading solution of de 0.16 molCO₂/mol TEA+MEA, with a minimal energy consumption of 2.15 GJ/tCO₂ and an efficiency of 85 % of the CO₂ capture process. L/G ratio represents another very important parameter for analysing variation of energy demand for regeneration of CO₂ capture process. There has been determined the optimal value of this ratio for which the energy demand is minimal and efficiency of CO₂ capture is maximal. The variation of L/G ratio has been performed for constant flue gasses flow and modifying the flow of the solution, determining the optimal value for L/G ratio as function of the type and concentration of solvent used. The results show that the optimal value of L/G ratio for the minimal energy demand for regeneration (2 and 2.5 GJ/tCO₂) has been established between 0.7 and 1 [kg_l/kg_{gas}], keeping the efficiency of CO₂ capture process over 85 %.

As a general conclusion the mixture of primary and tertiary amines represents an advantageous solution that keeps the energy demand low for 30 % TEA+5 % MEA and the efficiency of CO₂ capture process can be increased up to 95 %.

The analysed process can be directly used in cement and glass industries, being an advanced measure of CO₂ management through efficient

capture and isolation of carbon. This method can be also used for large flows of flue gasses resulted from technological processes from cement and glass industries with condition to keep the L/G ratio within the recommended domain between 1 and 2.5 [kg_L/kg_{gas}]. Eventual restrictions regarding the flue gasses flow can be of economic nature due to the increase of costs of absorbant solution with increasing its quantity.

The described method, being a post-combustion CO₂ capture method, imposes a certain domain of temperature for flue gasses cooling (30 - 45°C), in order to achieve optimal efficiency of carbon absorption.

Heat from flue gasses, resulted at high temperatures (over 1000°C), from technological processes from glass and cement industries and also from other industries with combustion processes, can be recovered within the analysed contour, e.g. pre-heating combustion air, or using an external recuperation for heat generation. This can lead to increasing energy efficiency of the process through heat recuperation from flue gasses as secondary energy resources, which also leads to decreasing environmental impact, and flue gasses are cooled down to temperatures required by CO₂ capture technology [29].

In the cement industry, for substantial reduction of CO₂ emissions (about 30 % and even more), it is opportunistic to combine this technology with other measures such as: utilisation of different technologies for CO₂ capture at stages before combustion or during combustion process, cement production with low clincher contain, e.g. composite cement from fly ash or slag, intensive utilisation of alternative fuels (biofuels, with low carbon content, with reduced CO₂ emissions), increasing the energy efficiency of cement production processes through improving the equipment technology [30-34].

Increasing the efficiency of CO₂ capture by using the proposed method can lead to increasing the economic benefit of the cement, glass and other construction materials companies through commercialisation of CO₂ emissions.

Increasing the price of the carbon certificate can be a measure to support different CO₂ capture technologies. In 2012 the price for carbon certificate has varied between 4 and 12 Euro/tCO₂. Some studies anticipate an increase of price of carbon certificate up to 60 Euro/tCO₂ [35].

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