

POST COMBUSTION REMOVAL OF CARBON DIOXIDE FROM FLUE GASES

Ioana Ionel¹, Dumitru Cebrucean^{2*}, Francisc Popescu³, Viorica Cebrucean Harea⁴, Luisa Izabel Dungan⁵, XXXPadureanu XXX⁶

¹Prof.dr.ing., Faculty of Mechanical Engineering, Politehnica University of Timisoara, Ionel_Monica@hotmail.com, ioana.ionel@mec.upt.ro

Tel +40 256 403670 Fax +40 256 403669, Bv M Viteazu Nr 1, 300222, Timisoara, Romania

^{2*}PhD student.Eng., Faculty of Mechanical Engineering, Politehnica University of Timisoara, Bv. M. Viteazu, Nr. 1, 300222, Timisoara, Romania, tel/fax: +40256403669/403670, e-mail:

dumitru_cebrucean@yahoo.com

³Assoc. professor, PhD Eng., Faculty of Mechanical Engineering, Politehnica University of Timisoara

⁴MSc.Eng., Faculty of Mechanical Engineering, Politehnica University of Timisoara

⁵PhD, Eng. Assist., Faculty of Mechanical Engineering, Politehnica University of Timisoara

⁶Assoc. professor, PhD Eng. Faculty of Mechanical Engineering, Politehnica University of Timisoara

Abstract: *The primary aim of this work is to analyze and describe the process of capturing carbon dioxide (CO₂) from the flue gases by means of chemical absorption. Aqueous monoethanolamine, 30 and 40 wt % MEA, has been selected for removing CO₂ from the combustion gases. The concentration of CO₂ in the flue gases ranges from 15 to 18 % after combustion and 2 to 4 % after the CO₂ absorber. Secondly, the process of coal co-combustion with biomass waste in a fluidised bed reactor was investigated. Experimental test results have shown that the emissions of CO₂, SO₂ and NO_x have been significantly reduced during the removal processes and have proved the viability of the proposed systems.*

Keywords: CO₂ capture / monoethanolamine (MEA) / fluidised bed combustion / efficiency / co-combustion fossil fuel biomass

1. PURPOSE

Emissions of greenhouse gases are charged to cause climate change. The main greenhouse gas is carbon dioxide (CO₂) and the major source of it is the combustion of fossil fuels (coal, oil and gas) to supply energy under different forms, such as heat, electricity, mechanical work, etc. Global emissions of CO₂ from fossil fuel combustion increased from 20.7 billion tones (Gt) in 1990 to 24.1 Gt in 2002 (i.e. an increase of 16 % in comparison to 1990), as Metz et al. (2005), OECD (2004) and Beising (2007) are indicating.

At the Bangkok reunion of the UN climate report in May 2007 one concluded that until 2015 the emission of greenhouse gases should be stabilized, in order to hope not to exceed with more than two degrees Celsius the average temperature. Available technologies exist; their costs are not unrealistic as they are in the range of 0.12 % of the world global economic potential. Not doing anything might be to expensive and late solution or postponed action will cost much more in comparison to prompt present action. The clock for the traditional energy system keeps ticking louder notifies Scheer (2007).

Also it is known that only by the integration of all new admitted countries in the biomass co-firing pathway, the European Union will be able to reach its own targets of reduction of CO₂ and increasing of renewable source share by the year 2010.

CO₂ capture is already an industrial technology, used today notably to process natural gas. It is commonly called on in the manufacture of fertilizers, in the food-processing industry and in the energy sector (the oil and gas industry). The main problem is generally the low concentration of CO₂ in the flue gas.

It would be out of the question to seek to compress the CO₂ for storage, from the stand point of both energy and storage capacity. Separation methods are thus required so as to trap CO₂ preferentially.

Three main categories are recognized (Figures 1, 2 and 3, according <https://www.co2castor.com/>:

- post-combustion capture,
- oxy-fuel combustion capture, and
- pre-combustion capture.

Post-combustion capture is designed to extract the CO₂ that is diluted in the combustion flue gas. It can be integrated into existing facilities without demanding any major modifications. The most common process is CO₂ capture by solvents, generally amines.

Other processes are under consideration involving the calcium cycle and cryogenic separation. The former consists in quicklime-based capture that yields limestone; this is then heated, thereby releasing CO₂ and producing quicklime again for recycling. The cryogenic process is based on solidifying CO₂ by frosting to separate it out.



Fig. 1: Post-combustion capture.

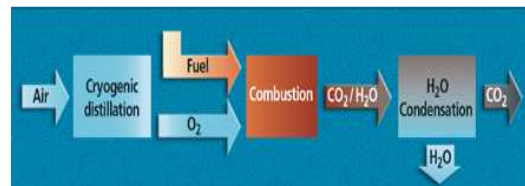


Fig. 2: Oxy-fuel combustion capture.

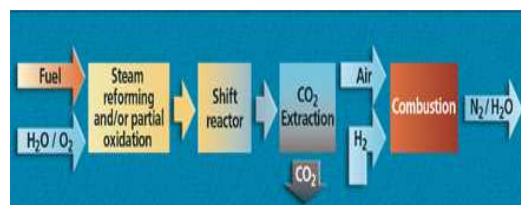


Fig. 3: Pre-combustion capture.

The **technology of oxy-fuel combustion capture** is not CO₂ capture in the true sense of the term. Here, the process is applied at the input as opposed to the output stage: the objective is to obtain flue gas with a 90 % CO₂ content by performing combustion in the presence of pure oxygen. Because it recycles part of the CO₂ as a substitute for the nitrogen in air, oxy-combustion is particularly well suited when an existing facility is being retrofitted. However, separating out the oxygen from air, performed mainly using the cryogenic principle, is both costly and energy-consuming. To give an idea, the energy consumption involved in supplying pure oxygen to a 500 MW coal-fired power station that operates 8000 hours a year

would represent 15 % of the electricity it generates annually. To avoid the cost of separating out the oxygen from air, a promising technology is under consideration: **chemical looping combustion**. It consists in bringing the oxygen in the air into contact with a metallic medium that, when it circulates, transfers the oxygen

With the **pre-combustion capture**, the goal is to trap the carbon prior to combustion: the fuel is converted on entering the installation into synthesis gas – a mixture of carbon monoxide (CO) and hydrogen. The two main techniques are steam reforming of natural gas in the presence of water and partial oxidation in the presence of oxygen. The CO present in the mixture reacts with the water during the shift conversion stage to form CO₂ and hydrogen. The CO₂ is separated from the hydrogen, which can then be used to produce energy (electricity or heat) without giving off CO₂.

The gases exhausted as well the possibility of free CO₂ energy generation is resumed by Figure 4.

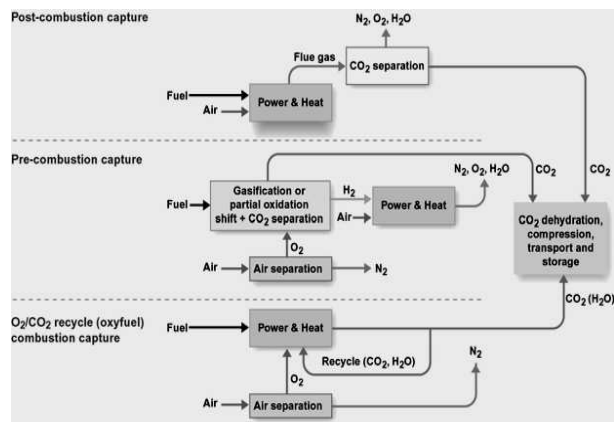


Fig. 4: General overview concerning CO₂ capturing technologies.

2. SCOPE OF THE RESEARCH

Presently, power generation represents the largest contributor to world manmade emissions, accounting for about one-third of the world's total exhausted amount of CO₂. These emissions can be reduced by a variety of measures, such as (i) improving combustion efficiency of the power plant, (ii) fuel substitution to lower or zero-carbon fuels, (iii) CO₂ capture technologies and (iv) increasing the use of renewable energy sources. If no actions will be taken, the global manmade CO₂ emissions will dramatically increase and support the ever known green gas effect, meaning irreversible the global warming process, in principal.

Among the types of fossil fuel used, coal has the highest carbon content, resulting in coal-fired power plants having the highest output rate of CO₂ per kilowatt-hour produced. Mitigation of the emissions of carbon dioxide from coal-fired power plants is the target of this work-project.

Co-combustion of biomass with coal seems to be a solution for solving this problem. The explanation is based on the fact that the use of biomass for energy production does not supplementary increase the CO₂ content of the atmosphere trough combustion in comparison to the CO₂ destroyed by photosynthesis during the lifetime, and so, it is considered CO₂ neutral. Moreover, biomass has low concentration of sulfur, which is also very important.

Substantial research & development efforts are now in progress aimed at improving capture technologies so as to render them more efficient and more cost-effective. Under these efforts also the presently reported research is included.

The primary aim of this study is to analyze and describe the process of capturing carbon dioxide from the flue gas by means of chemical absorption. Aqueous monoethanolamine has been selected for removing CO₂ from the combustion gases. Secondly, co-firing process of biomass with coal in fluidised bed combustion is also investigated. Fluidized bed has been chosen due to its uniform temperature distribution at lower level in the combustion zone during operation and longer residence time in such regions, and thus, one is expecting to higher the combustion efficiency and to enhance the NO_x production by thermal mechanism.

Flue gases from biomass-coal co-combustion are containing not only emissions of carbon dioxide, but also other air pollutants such as oxides of nitrogen and sulfur which are caused from the fossil fuel share, and, in addition, also special pollutants such as chloral and flour acids, that are also very dangerous. Therefore, it was highly desirable to remove all impurities, particles and air pollutants prior to the CO₂ capture, as they were supposed to inhibit the ability of the solvent to absorb the CO₂. In order to retain these contaminants, the experimental installation was designed and equipped with upstream de-NO_x and de-SO_x facilities, as well as with fly ash and particles removal systems. Especially a flue gas desulphurization method is described and presented. For the removal of sulphur dioxide a "wet" desulphurization unit has been designed, and, as solvent has been used aqueous sodium or calcium hydroxides.

Experimental results have shown that the emissions of CO₂, SO₂ and NO_x have been significantly reduced during the removal processes. The combustion process of biomass with coal in fluidised bed has been effective.

3. EQUIPMENT, FUEL CHARACTERISTICS AND OPERATION

Figure 4 shows the experimental lab facility for biomass co-firing with coal (Lignite of Romanian origin with low calorific value) in a fluidized bed combustion system, as well as the flue gas post-treatment processes. Emissions of sulfur dioxide are removed from the flue gases before the process of CO₂ capture. The flue gases to be treated are directed to an absorber, where they are mixed with a solvent. Having more affinity with the CO₂ molecules than with the other components of the flue gases (in particular nitrogen), the solvent captures the CO₂ (the solvent is "enriched") and the other molecules are discharged from the absorber (treated flue gases).

Details about the facility are specified by Cebrucean et al. (2007), Ionel et al. (2007), Savu Alex. and al (2007).

Table 1 gives the main characteristics of the used fuels in co-combustion. The figures are partially verified with data offered by Alie and al. (2005). The coal is of local origin and is low calorific Lignite, with sulfur and considerable amount on ash and humidity. From the large pallet of biomass tested, in the present article one presented only results concerning sawdust, as it is most available and representative.

Table 1: Main characteristics of used fossil coal and waste biomass.

Characteristics and IS units on given basis	Lignite	Wood sawdust
LHV (raw) [MJ/kg]	9	12.4
Moisture (raw) [%]	50.4	33
Volatile matter (dry) [%]	52.11	83.2
Ash (dry) [%]	15.1	0.34
Fixed C (dry) [%]	32.83	16.5
H (dry) [%]	4.9	5.7
N (dry) [%]	0.69	0.13
S (dry) [%]	0.39	0.05
Cl (dry) [%]	< 0.1	< 0.1
O (dry) [%]	13	45
ash melting temperature [°C]	1050	1200

Main concern has been paid to the deposit formation. As Junker and Folmer (1998), indicate co-firing of biomass with coal decreases the ash deposition rate relatively when interpolating with the amounts of dry ash flow from each fuel. One used as biomass participation rates of 5, 15 and 20 % (by mass), but the results presented in the frame of this paper are resumed only at the last case. Analysis of the chemical composition of the deposit indicates that the inorganic from the coal and bio fuels interact, and by co-firing combinations lead to sulphating as dominant.

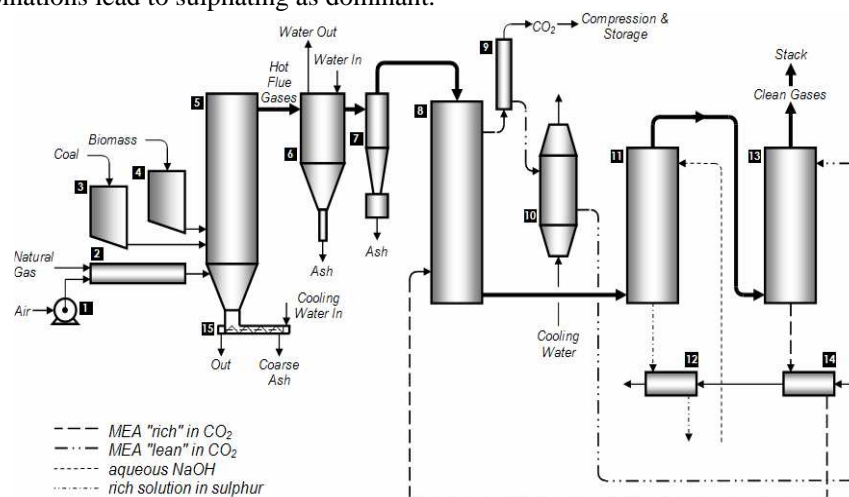


Fig. 5: Co-combustion of biomass with coal in fluidized bed with flue gas post-treatment, including flue gas desulphurization and carbon dioxide capture: 1-air fan, 2-combustion air pre-heater, 3-coal feed, 4-biomass feed, 5-combustion chamber, 6-heat exchanger flue gas-to-water, 7-cyclone, 8-desorber, 9-CO₂ separator, 10-heat exchanger MEA-to-water, 11-FGD (flue gas desulphurization unit), 12-cooler, 13-CO₂ absorber, 14-cooler, 15-bottom ash cooling screw.

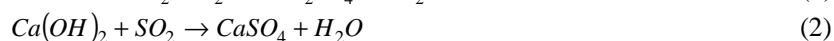
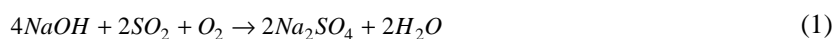
The chamber of combustion (5) consists of a vessel with a grate at the bottom through which air is introduced. In order to support the ignition of the mixture, the combustion air is heated up in an air pre-heater (2). When the inlet-bed temperature in the combustor reaches almost 400 °C, the fuel mixture of biomass and coal starts the ignition progression and burning.

During the process of combustion, the coarse ash is removed from the combustor by means of a screw (15), located at the bottom. From the top of the combustion chamber, the hot combustion gases with temperatures until 1000 °C (due to a separate post combustor running gas situated at the end of the furnace) are passed through a heat exchanger (6), where they are cooled down to the temperature levels required for the removal processes. After transferring the specific heat, the gas enters tangentially a cyclone (7), where particles of the fly ash are separated and removed from the bottom of the unit, while the cleaned flue gas leaves at the top. The amount of fly ash depends mainly on the ash content of the fuel,

which is in case of Romanian Lignite considerable (at least 35 - 40 % by mass on humid basis). After leaving the cyclone, the flue gas with low ash content flows through a desorbing device (8). Here, the flue gas having appropriate temperature gets in counter heat exchange with the solution of amine "rich" in carbon dioxide. The flue gas temperatures measured during the test are around 255 °C before entering the desorbing unit. After the flue gas pre-treatment, a flue gas desulphurization process follows.

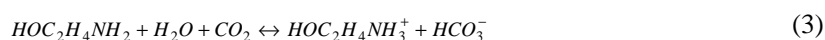
The primary aim of this research was the mitigation of carbon dioxide. As this complex process cannot be effectively achieved without capturing previously other gaseous pollutants such as sulfur dioxide and nitrogen oxides. Sulfur dioxide can be removed using a variety of methods one of them is classified as flue gas desulphurization (FGD).

The co-combustion test installation has been equipped with a "wet" scrubber (11), in which appropriate aqueous solutions of sodium hydroxide (NaOH) or Ca(OH)₂ were selected as active washing fluids [4]. Prior to be introduced, sodium hydroxide or calcium oxide were dissolved in a solvent - water (H₂O) - forming a strongly alkaline solution, the mass ratio of NaOH to H₂O was 1:100, respectively the molar ratio Ca:S was 1.5:1. In the FGD unit, the flue gases get into direct contact with the selected aqueous solutions which are pumped to the scrubber with 2 l/h. Sulphur dioxide reacts with sodium hydroxide and forms a mixture of sodium sulfite (Na₂SO₃) and water. After that, sodium sulfite absorbs additionally sulphur dioxide, resulting in a chemical compound of sodium hydrogen sulfite (NaHSO₃). It is important to note that, the solution of sodium hydroxide also reacts with other acid gases. For example, carbon dioxide reacts with aqueous sodium hydroxide to yield sodium carbonate (Na₂CO₃). In case of injecting calcium hydroxide as additive in the FGD, the solid calcium carbonate is formed finally. The absorption processes occur as follows:



After the removal of sulphur dioxide, the flue gas is sent to the CO₂ absorption unit (13). The CO₂ is removed from the flue gas by means of a chemical solvent. As a solvent, there was selected an aqueous solution of monoethanolamine (MEA), with concentration ranging between 35 and 45 %. Flue gases with low sulphur content enter the CO₂ absorption tower and come into counter-flow contact with "lean" solution of MEA, which chemically absorbs the CO₂-gas. The fundamental reaction between carbon dioxide, a weak acid, and monoethanolamine, a weak base, is reversible under specific thermal conditions. Under these circumstances, if aqueous MEA is cooled to the temperature levels of 40-60 °C, then the chemical solvent retains the CO₂. Opposite to this procedure, when the MEA is heated up to 120-140 °C, it releases the CO₂-gas and the regeneration of the chemical solvent takes place.

The principal chemical reaction is:



During the absorption process, the reaction proceeds from left to right.

From the bottom of the column, the "rich" MEA solution, which contains the chemically bound CO₂, is passed through a cooling phase (14). Afterward, it is pumped to the de-sorption tower where and it is heated in counter flow up to 120-140 °C by the flue gas stream, in order to be able to release almost pure CO₂.

During regeneration, the reaction proceeds from right to left, thus CO₂ and H₂O evolve separately from the amine solution. The captured CO₂ leaves through the top of the separation unit (9). It is then compressed and stored. The "lean" solution of MEA, containing far less CO₂, is cooled down to 40 °C in a cooler (10), and recycled back to the absorber, for further additional CO₂ capture and continuity of the global process.

4. RESULTS

Temperature behavior inside the combustor is shown in Figure 6. During operation the highest temperature received was 980 °C (for few seconds expecting higher NO_x). No other major operational

difficulties were observed during temperature variation. The decrease of the gas temperature was caused by the fuel feeding interruption.

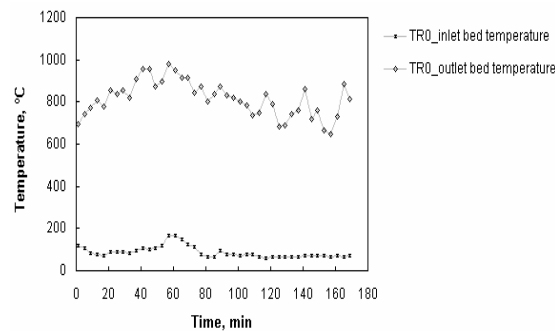


Fig. 6: Temperature profile inside the combustor.

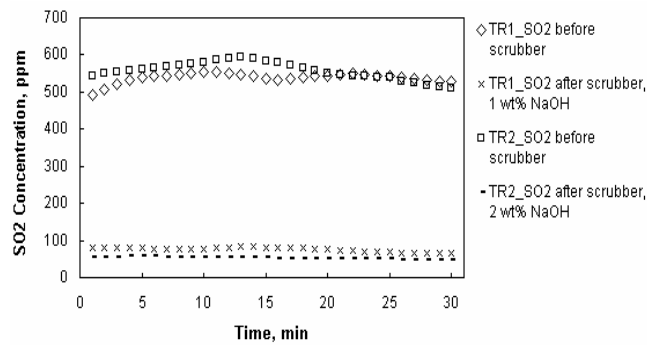


Fig. 7: SO₂ concentration profiles (TR1: 1 wt% NaOH, pH=13; 2 wt% NaOH, pH=13.3).

Figure 7 shows the concentration of SO₂ in the flue gas before and after the scrubber, using a strong alkaline solution of 1 wt % and 2 wt % NaOH. After leaving the scrubber the concentration of SO₂ was in the range of 50-75 ppm.

CO₂ absorption into 35 wt % MEA was relatively good with an overall absorption efficiency of 66 %.

It should be taken into account that the acceptable concentrations' levels of SO₂ and NO_x in the flue gas before the absorber are recommended to be in the range from 10 to 50 ppm, as described by Metz and al. (2002).

Using a 2 wt % NaOH concentrated solution it was possible to achieve 50 ppm of SO₂. But in case if NO_x are not controlled the ability of the MEA solvent to capture more CO₂ decreases. During operation we measured high NO_x, 280-340 ppm.

Figure 8 shows CO₂ emission profiles and Figure 9 presents the removal efficiency during test runs.

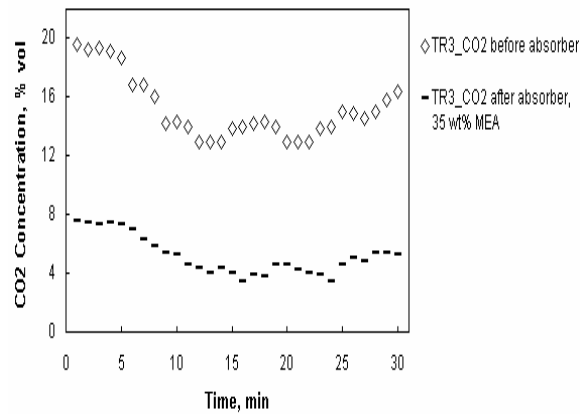


Fig. 8: CO₂ concentration profiles (TR3: 35 wt% MEA, pH=10.7).

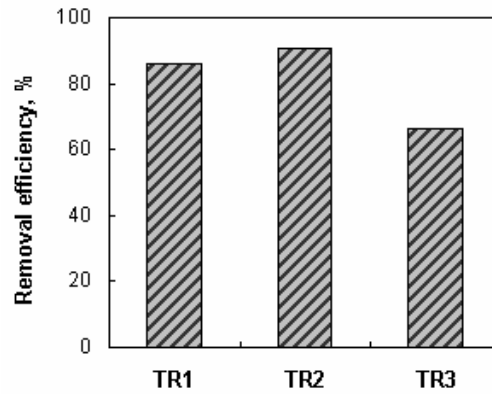


Fig. 9: SO₂ and CO₂ removal efficiency (TR1: 86% 1 wt% NaOH; TR2: 91% 2 wt% NaOH; TR3: 66% 35 wt% MEA).

5. CONCLUSIONS

The study presented is concerning the carbon dioxide capture and separation from the flue gas by means of aqueous solution of monoethanolamine as well as, the SO₂ removal procedure using sodium hydroxide. The main conclusions of the study are summarized as follows:

The higher the concentration of CO₂ in the flue gas, the faster it is absorbed by MEA. There are several compounds, typically present in flue gas, to which MEA absorption is particularly sensitive (e.g., SO₂, H₂S, NO_x, etc).

Careful attention must also be paid to the fly ash and soot present in the flue gas, as they might plug the absorber if contaminants levels are too high. To a lesser or greater extent, the abundance of these molecules in the flue gas depends upon the composition of the fuel mixture between coal and biomass. The target was to reduce as much as possible their concentration in the flue gas, since they can inhibit the ability of the solvent to absorb CO₂.

Of all experiments performed, gaseous emissions of SO₂ and NO_x were reduced, as expected when using biomass with low sulphur and nitrogen content. Also, depending on the firing conditions NO_x emissions were found to decrease or remain at the same level.

Using a wet scrubber is one of the options for removing sulphur dioxide, and the technique should be applied before flue gases enter the CO₂ absorber. Using sodium hydroxide, one achieved a reduction of SO₂ by 90 % and greater. Carbon dioxide concentration in the flue gas has been decreased by 60 %,

representing an average of all data mapped. However, the low content of sulphur, of oxides of nitrogen and some particles of ash and dust, which were in the flue gas before the CO₂ absorber, has determined the degradation of MEA.

No major operational difficulties were observed adding biomass to the combustion process, concluding that small quantities of biomass, up to 10 % by heat input, can be easily added to conventional systems based on fossil solid fuel (coal), without major investments. Since biomass fuels are more volatile, the furnace volume must be large enough to accomplish complete combustion of the gases, as a requirement.

Large quantities of heat are required by the desorption unit to regenerate the MEA solvent. Deciding where this heat is to come from is a fundamental part of the design of an MEA absorption plant. One approach is to extract the required heat from the flue gas that leaves combustor, as it was shown. As consequence, the power plant is more difficult to design, more costly during the function and the power output of the station decreases. For instance, power plants with CO₂ capture lose about 10 % in efficiency, in comparison with those without CO₂ capture. This means that the consumption of fossil fuels will increase dramatically, the cost of energy production will increase too. One has still to optimize the global process, and there is only matter of time to succeed. Nevertheless the first developments in Europe demonstrated the technical possibilities for CO₂ retention, in order to its sequestration. Also major plans are drawn for the next future, in international co-operation. Key targets of CASTOR are pointed out in <http://www.cachetco2.eu/c2ws/projects.html>.

is a major reduction in post-combustion capture costs, from 50-60 € down to 20-30 € per ton of CO₂ and the validation of the new processes in a capture pilot plant installed in a coal-fired power station in Denmark, to advance general acceptance of the overall concept in terms of storage performance (such as capacity, CO₂ residence time, etc.), storage security and environmental acceptability and to start the development of an integrated strategy connecting capture, transport and storage options for Europe' according to <http://www.encapco2.org/>.

Intense research, development and demonstration efforts have to be directed towards the optimising development of the new energy technologies, and improve the existing one, as promising results are already achieved.

The proposed technology of co-firing biomass with coal is a viable option that promises reduction in the emissions of CO₂, SO₂ and NO_x, allowing in addition to the clean combustion, also the CO₂ capturing.

The novelty consists also of the co-combustion process of a fossil fuel with a renewable, CO₂ neutral energy resource, such as biomass waste. Globally the CO₂ reduction might become more attractive as price, taking into account the positive involvement of the CO₂ credits that are achieved, by that way, and that are adding financial input to the economy of the technology, in its whole.

NOMENCLATURE

MEA – Monoethanolamine,
FGD - flue gas desulphurization unit,
TR_i - Registration point/range for temperature or other parameters.

ACKNOWLEDGEMENT

The national founded excellence research project EmSHIPS (2006-2008) NB 108/2005 and OVAPED (2006-2008) NB 282 and EPOC Nb 22131 are mentioned as financer of the present reported datal. Special thanks addressed to SC SAVPROD SRL Bucuresti, as partner in the Romanian patent dealing with the presented matter.

REZUMAT

Principalul scop al acestei lucrări este de a analiza și descrie procesul de captare a dioxidului de carbon (CO₂) din gazele arse prin intermediul absorbției chimice. Soluția apoasă de monoetanolamină, precum și

cea cu 30 – 40 % conținut de umiditate a fost selectată pentru începătarea CO₂ din gazele arse. Concentrația de CO₂ din gazele de evacuare variază între 15 și 18% după ardere și 2 – 4% după absorbția de CO₂. În al doilea rând a fost investigat procesul de co-ardere a cărbunelui și biomasei în pat fluidizat. Rezultatele experimentale au arătat că emisiile de CO₂, SO₂ și NO_x s-au redus semnificativ în timpul procesului de îndepărtare și au demonstrat viabilitatea sistemelor propuse.

Cuvinte cheie : captare de CO₂, monoetanolamină, arderea în pat fluidizat, eficiență, co-arderea dintre biomasă și combustibil fosil.

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