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REMOVAL OF CO_2 FROM GAS STREAMS IN AIRLIFT REACTORS

BY

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Abstract. Emissions of carbon dioxide (CO_2) and other greenhouse gases have become a major environmental issue due to their intensive contribution on the global warming. In the last decades the removal of CO_2 from gas mixtures has become an important industrial operation due to the necessity of different gases purification and from recovery reasons taking into accounts the large applications of CO_2 in chemical and food industry. Various techniques have applied for CO_2 removal. These technologies are based on different physical, chemical or biological processes. In addition several types of contactors have been reported for CO_2 removal such as stirred tanks, packed columns, bubble columns and membrane contactors. This paper discusses the most recent application of airlift reactors in flue gas treatment, in particular the potential of application for the effective CO_2 removal from contaminated flue gases taking into account the most important advantages of these devices: simple design and construction, no moving parts, high mass and heat transfer, intensive mixing, low shear stress to cells, low energy consumption, low operational costs. The most relevant characteristics and properties of CO_2 as well as the most applied technologies for CO_2 removal are reviewed in this paper, based on the information from literature data.

Key words: airlift reactor, absorption, CO_2 removal.

1. Introduction

In the last decades, the emissions of pollutants in the atmosphere have increased considerably with the intensive development of industry processes, transport, electricity, heat production etc. Carbon dioxide (CO_2) is one of the most important greenhouse gases responsible for the intensification of global warming potential and climate change due to its ability to absorb infrared radiation. The increase in atmospheric CO_2 emissions from different industrial emission sources such as combustion of fossil fuels, biological processes (decomposition of organic material, fermentation, digestion), manufacture of cement, metallurgical coke, caustic soda etc. lead on the investigation and development of different methods and devices for removing of CO_2 from flue gas streams. In this context airlift reactors are promising contactors that can be successfully applied for removal of CO_2 from contaminated gas streams taking into account the most important advantages of them: simple design and construction, no moving parts, high mass and heat transfer, intensive mixing, low shear stress to cells, low energy consumption, low operational costs [1], [2].

The main objective of this work is to analyze and evaluate the performances of airlift reactors applications in flue gas treatment, in particular the potential for the effective CO_2 removal from contaminated flue gases based on the information from literature data.

2. CO_2 - General Characterization, Properties, Sources

Carbon dioxide is an inorganic compound, colorless and odorless gas, which in solid state is commonly called dry ice. The main chemical and physical properties of CO_2 [3] are related in Table 1. The toxicity and effects of CO_2 on human health can be expressed taking into account its concentration in the air, therefore air with [4], [5]:

− 1% CO_2 can cause drowsiness;

− 5% CO_2 causes dizziness, headaches, confusion and difficulty in breathing;

− 6-10% CO_2 causes headaches, dizziness, sweating and shortness of breath;

− 10-15% CO_2 causes impaired coordination and abrupt muscle contractions;

− 20-30% CO_2 causes loss of consciousness, convulsions and coma;

− > 30% CO_2 can cause death.

Environmental Protection Agency (EPA) of USA classifies the carbon dioxide as a dangerous pollutant that affects human health. Signed in 1997 by 161 countries, the Kyoto Protocol aims to reduce the average amount of greenhouse gases by 5.2% in 2012 compared to 1990. Romania was the first country that ratified the Kyoto Protocol, by the Law no.3/2001 engaged to an 8% reduction in greenhouse gas emissions in the period 2008-2012 compared to 1990.

CO_2 is an important component in the natural carbon cycle (is produced during respiration of animals and plants and in the exchange between the atmosphere and ocean, released in volcanic eruptions and used by plants in photosynthesis). A major source of anthropogenic CO_2 emissions in high concentrations is the biogas resulted from anaerobic digestion of organic waste from the fermentation tanks or landfills, which typically contains 50-80% CH_4 and 20-50% CO_2 and other trace compounds [6]. Also large quantities of CO_2 are released during combustion processes of fossil fuels (oil, gasoline, diesel, methane, coal). CO_2 could have large applications in chemical and food industry. The main applications of carbon dioxide [7], [8] are related in Fig. 1.

Table 1
CO_2 Properties [9]

Molecular formula	CO_2
Molar mass	44.01 g/mol
Density	Solid: 1600 g/L; Gas: 1.98 g/L
Flamability	Non-flammable
Melting point	−57°C (under pressure condition)
Boiling point	−78°C
Solubility in water	1.45 g/L
Viscosity	0.07 cP at −78°C
Appearance	Colorless, odorless gas

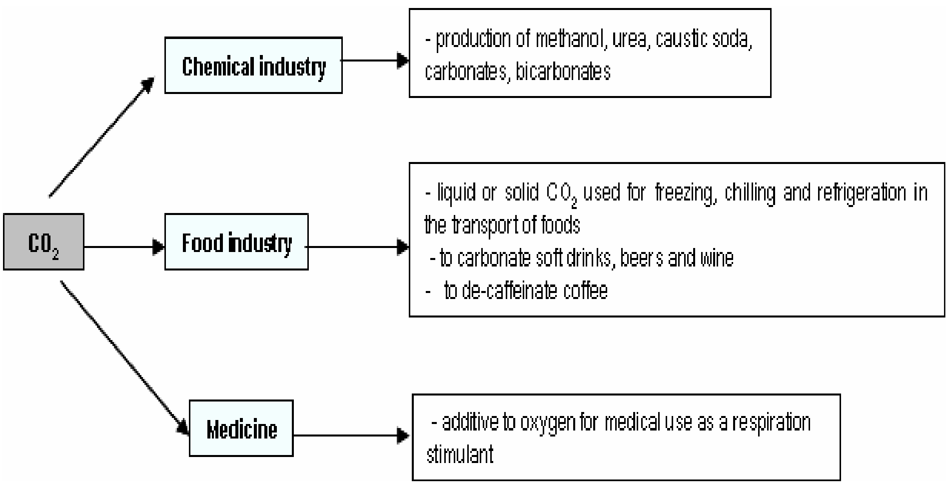

Fig. 1 − The main applications of CO_2.

3. Processes and Techniques Applied for Decontamination of CO_2 Gas Streams

In the last decades the increase in atmospheric CO_2 emissions leads on the investigation and development of different methods and devices for removing of CO_2 from flue gas streams. Various techniques have applied for CO_2 removal/capture such as [10]:

− physical absorption (scrubbing liquid);

− chemical absorption (chemical reaction with a liquid, such as absorption in aqueous amine);

− adsorption in solid sorbents such as activated carbon, zeolites, carbonates;

− membrane separation processes;

− cryogenic separation processes (high-pressure cooling);

− biological methods (biofixation of carbon dioxide by photosynthetic organism).

Among these methods, the *chemical absorption* using aqueous alkanolamine (monoethanoloamine, diethanolamine, methyl diethanol amine, 2-amino-2-methyl-1-propanol) or hot potassium carbonate solvents is currently the most promising industrial process for removal of CO_2 from different gas streams [11]. In addition several types of gas-liquid contactors have been reported for CO_2 removal such as venturi scrubbers, spray tower, stirred tanks, packed columns, bubble columns [10].

Petrov *et al*. [11] studied the chemisorptive removal of carbon dioxide from process streams using a slurry bubble column and a mixture of hexadecylamine or dodecylamine in various concentrations with methanol and others alcohols as the solvent. The conversion rate of CO_2 was significant (up to 99% for CO_2 absorption with methanol as the solvent). Álvarez *et al*. [12] carried out gas-liquid mass transfer studies of carbon dioxide in polluted aqueous solutions with surface active substances in a cylindrical bubble column. The authors noticed that the flow rate of gas and liquid phases produced an intensive increase of mass transfer rate due to the effect upon the interfacial area or upon driving force.

The conventional column absorbers used for CO_2 removal are energy-consuming and are difficult to operate due to the problems associated with foaming, flooding or entrainment [13]. In this context researches have been conducted in order to develop new economical solutions. Studies showed that *membrane gas absorption process* is a good alternative to conventional techniques [13]. For instance, Portugal *et al*. [14] developed a novel efficient technology based on the application of hollow fiber membrane contactors with regenerable liquid absorbents for carbon dioxide removal from anaesthetic closed breathing circuits. The performances of the contactor were analyzed by a 2D numerical model developed by authors.

Physical absorption of CO_2 involves gas absorption in an absorbing liquid (solvent) under high pressure without chemical reaction. The most physical solvents used for CO_2 removal are: water, methanol, selexol (a mixture of dimethyl ether and polyethylene glycol), propylene carbonate, N-formyl morpholine, N-metilpirolidonă, tributyl phosphate, metilcianoacetat. Lampert and Ziebik [15] analyzed the CO_2 removal process applied to metallurgical fuel gases: blast-furnace gas and Corex gas using physical absorption process with the Selexol solvent. They obtained a specific power consumption of 335 kJ/kg CO_2 and 505 kJ/kg CO_2 for Corex gas and Blast furnace gas, respectively. The specific amount of CO_2 removed for Corex gas and Blast furnace gas was 0.484

kg CO_2/kg_{fuel} and 0.215 kg CO_2/kg_{fuel}, respectively. Also they obtained a purified gas of 5.2% and 1.9% CO_2 for COREX and Blast furnace raw gas containing 35% CO_2 and 16% CO_2, respectively.

Adsorption of gases on solid adsorbents is an efficient technique due to the high adsorption capacity, high selectivity, good mechanical properties and ease regeneration of the adsorbent [16]. Yong *et al*. [17] investigated the adsorption capacity of carbon dioxide on high surface area carbon-based adsorbents at high temperature. The results showed an adsorption capacity for carbon dioxide of 0.28 and 0.22 m mol/g at 300±C and 1 Bar pressure.

Biofixation of carbon dioxide by photosynthetic microorganism (usually microalgae) is an efficient *biological method* applied for the capture and storage of CO_2 that has been extensively investigated as an alternative of chemical and physical processes being an economically feasible and environmentally sustainable technology [18]. According to Milne *et al*. [19] the photosynthetic microorganisms are able to "capture sunlight and use that energy to store carbon in forms useful to humans such as fuels, food additives, and medicines". The CO_2 removal rates were examined by Jacob-Lopes *et al*. [20] in a bubble column photobioreactor using a culture of the cyanobacteria *Aphanothece microscopica Nägeli.* The authors underline the importance of operational parameters (temperature, light intensity, CO_2 concentration) of the photobioreactor on the kinetics of carbon dioxide removal. They observed that the ratios between constant for the rate of carbon dioxide removal as a function of the constant for the rate of carbon dioxide lost to the atmosphere were higher under the conditions of 25ºC, 15% of carbon dioxide concentration and independent of the light intensity. Sydney *et al*. [21] evaluated the amount of the carbon dioxide assimilation of four microalgae: *Dunaliella tertiolecta*, *C. vulgaris*, *Spirulina platensis* and *Botryococcus braunii*. The authors obtained a carbon dioxide fixation rate (mg L^{-1} day^{-1}) equal to 251.64 for *C. vulgaris*, 496.98 for *B. braunii*, 318.61 for *S. platensis* and 272.4 for *D. tertiolecta*. Lee *et al*. [22] studied the CO_2 fixation from concentrated sources such as power plant exhaust using two species of algae *Chlorella sp*. and *Chlorococcum littorale*. The growths of microalgae are influenced by temperature, CO_2 concentration and light intensity. They obtained a maximum grow of *Chlorella sp*. of 0.21 dry cell at 6 Klux light intensities. In the same time the growth of *Chlorella sp*. was influenced by the concentration of CO_2, therefore at concentration of 10% and 20% CO_2 the growth of the algae was not inhibited but was declined to about half at 30% CO_2 concentration. At temperature of 26ºC and 30ºC the two species showed a maximum growth.

As a conclusion of this section the following aspects must be notice and taken into account in order to choose the optimum method of removing carbon dioxide from contaminated gas streams:

− the main disadvantages of absorption process is that the pollutant is transferred in to another phase and the pollution problems still remain because the pollutant is not destroyed [23];

− chemical absorption involves high energy consumption resulting from the chemical reaction and during heating of absorber and is recommended for low gas flow rates and low concentrations of impurities in gas [24];

− biological processes are more effective and more economical methods compared with conventional physical and chemical methods; an important disadvantage of this method is that it can be applied usually for low concentrations of pollutants, higher levels affecting the activity of microorganisms.

4. Application of Airlift Reactors in Gaseous Streams Treatment

Airlift reactors are simple gas-liquid or gas-liquid-solid contactors characterized by pneumatic agitation, specific flow pattern and particular hydrodynamic characteristics that make them more efficient and attractive compared with other devices such as stirred tanks and bubble columns. As a result many applications of airlift reactors may be found in chemical and biotechnological industry for several type of processes (catalytic conversion of hydrocarbons, coal liquefaction, selective oxidation of alcohols, absorption processes, extraction, Fischer-Tropsch synthesis, production of antibiotics, proteins, vitamins, aerobic fermentation, aerobic cultivation, wastewater and flue gas treatment) [25], [26].

Initially airlift reactors were developed for wastewater treatments but these devices showed a significant potential for application in gas separation, purification and waste gas treatment, in particular using biological methods. Biological waste gas treatment in bioreactors is a new treatment alternative for conventional techniques (adsorption, absorption) [23]. The airlift system having a well-distributed and low shear stress creates optimal conditions for cultivation of organisms sensitive to intense shear forces. Therefore, according to Lo and Hwang [27] the airlift system "provided a more advantageous environment for the biological processes".

A baffled airlift reactor was applied by Ensley and Kurisko [28] for removal of *trichloroethylene* (TCE) from vapor phase using two species of microorganism: *Pseudomonas cepacia* or *Pseudomonas mendocina*. The authors varied TCE concentrations, air flow rates between 100 and 350 mL/min and cosubstrate feed rates. According to their results the concentrations of TCE up to 4,000 μg/liter of inlet air were degraded by at least 90% removal efficiency (RE). Increasing airflow rate leads on the decrease of TCE removal efficiency to about 60%. Zuber [29] compared the performances of an airlift reactor and trickling filter applied for the removal of *dichloromethane* (DCM) from air, operating with a biocatalyst fixed on sand. The airlift reactor showed a higher performance (the maximum DCM removed was 2300 g/m^3 h) compared with trickling filter (the maximum DCM removed was 450 DCM g/m^3·h). A novel airlift bioscrubber configuration was developed by Edwards and Nirmalakhandan [30] for the removal of air phase *benzene, toluene* and *xylene* (BTEX) compounds. According to the mathematical model developed by authors for this process, the removal rates higher than 99% can be achieved for benzene and toluene in the air stream with concentrations <1000 ppmv. An airlift inner-loop bioreactor immobilized with *Achromobacter sp* was applied by Quan *et al*. [31] for biodegradation of 2,4-*dichlorophenol* and *phenol*. The removal efficiency of 2,4-dichlorophenol decreased from 100 to 87.9% with increasing of phenol loading rates while the RE % of phenol were about 99.6%. An airlift bioreactor with biological membrane was applied by Jianping *et al*. [32] for the removal of *ethyl acetate* in air streams. Under the optimum operation conditions: temperature of 20–30ºC, pH 6.5–7.5 and waste gas influx of 0.05 m^3/h, the average removal efficiency of ethyl acetate was higher than 98%. A laboratory and pilot scale new air-lift-tube absorber with recycle and reaction tubes was applied by Bekassy-Molnar *et al*. [33] for the absorption of *sulfur dioxide* from SO_2/air mixture into sodium citrate buffer solution. The removal efficiency of SO_2 was significant (85% for both absorbers). Wang *et al*. [34] developed a three-dimensional transient computational fluid dynamics (CFD) in order to simulate the biotreatment of *toluene* waste gas in a laboratory scale airlift reactor. The authors observed that superficial gas velocity influenced significantly the treatment process. Therefore, the RE varied from 88 to 94% at gas phase inlet concentration of 500 mg/m^3 and superficial gas velocity variation from 0.01 to 0.02 m/s. At superficial gas velocity value of 0.0075 m/s and variation of gas phase inlet concentration from 500 to 2000 mg/m^3, RE was around 93%. These results are available for simulated transient RE. In case of experimental data, the RE obtained was 99%, therefore the model fit well with experimental data.

CO_2 removal in airlift reactors. In the last decades airlift reactors claimed to be essential for the design and development of a new generation of photobioreactors for cultivation and growth of photosynthesis organism [35]. For instance, Xu *et al.* [35] designed and developed a simple and low-cost airlift photobioreactor for microalgal mass culture. In a further work, Xu *et al*. [36] compared the effect of airlift and bubble column photobioreactors on growth and photosynthesis in vegetative gametophytes of *U. pinnatifida*. As it was expected higher growth rate of microorganism were achieved in airlift reactor compared with bubble column.

Many investigations have been made in order to demonstrate the applicability of airlift photobioreactors in CO_2 removal from gas streams. For instance, Yamada *et al*. [37] cultivated *Cyanobacterium anacystis nidulans* R2 in an airlift bioreactor in order to study cell-growth characteristics of this strain and its capability for CO_2 removal. At concentration of 5% CO_2 the highest growth rate of strain (0.046 h^{-1}) was achieved. The authors notice that the cell mass concentration was less affected when the CO_2 concentration varied from 5% at 30%. Mazzuca Sobczuk *et al*. [38] obtained a maximum CO_2 uptake efficiency of 63% by microalga *Phaeodactylum tricornutum* in a tubular airlift photobioreactor. Chiu *et al*. [39] applied an airlift photobioreactor with a porous centric tube in order to assess biomass production and CO_2 removal using a marine microalga *Chlorella sp*. Under the conditions of 5.15 g/L biomass concentration, 0.125 vvm aeration, aeration gas containing 10% CO_2, the maximum CO_2 removal efficiency was 63%. Hincapie *et al*. [40] designed and developed a novel 30-liter laboratory scale airlift photobioreactor for growing algae that can be further applied for CO_2 remediation in diverse industries. Cultivation of wild mixed microalgae in an airlift photobioreactor was developed by Du *et al*. [41]. The authors obtaining a better growth rate of microalgae feeding with 10% CO_2 compared to of that feeding with air. Fan *et al.* [42] compared the performances of a membrane-photobioreactor applied to remove CO_2 using *Chlorella vulgaris* with other three reactors: draft tube airlift photobioreactor, bubble column and a membrane contactor. The authors obtained a CO_2 fixation in the membrane-photobioreactor of 1.95, 2.15, and 6.40 times higher than that in the other three reactors. However it is important to notice that the membrane photobioreactor was designed based on the airlift reactor construction, replacing the cross- shaped air sparger with a dead-end hollow fiber membrane.

5. Conclusions

Carbon dioxide is one of the most important greenhouse gases responsible for the intensification of global warming potential. Several methods (physical and chemical absorption, adsorption, membrane separation processes, cryogenic separation processes, biological methods) and devices (venturi scrubbers, spray tower, stirred tanks, packed columns, bubble columns, airlift reactors) for removing of CO_2 from flue gas streams are investigated and developed. Chemical absorption using aqueous alkanolamine is currently the most promising industrial process for removal of CO_2 from different gas streams in spite of the disadvantages of the absorption process: the pollutant is transferred in to another phase and the pollution problems still remain because the pollutant is not destroyed. In addition, chemical absorption process involves high energy consumption resulted from the chemical reaction and during heating of absorber and in the same time it is recommended for low gas flow rates and low concentrations of impurities in gas. Biological processes are more effective and more economical methods compared with conventional physical and chemical methods. An important disadvantage of this method is that it can be applied, usually, for low concentrations of pollutants, higher levels affecting the microorganism's activity.

Airlift reactors have extensively been studied and applied in chemical and biotechnological process as well as in environmental remediation. ALRs showed a significant potential for application in gas separation, purification and waste gas treatment, in particular using biological methods. Numerous applications have been reported for treatment of several flue gas contaminants (toluene, trichloroethylene, BTEX, ethyl acetate, SO_2, 2,4-dichlorophenol and phenol). In addition, ALRs claimed to be essential for the design and development of a new generation of photobioreactors for cultivation and growth of photosynthesis organism for CO_2 removal. Studies are still necessary to be done in order to demonstrate the applicability of these devices for carbon dioxide removal using physical and chemical methods. But it is believed that ALRs are suitable devices for treatment of contaminated CO_2 gas effluents using different physical-chemical and biological methods due to the high efficiency removals of a large class of pollutants from contaminated media.

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ÎNDEPĂRTAREA CO_2 DIN FLUXURI GAZOASE CONTAMINATE ÎN REACTOARE GAZ-LIFT

(Rezumat)

Emisiile de dioxid de carbon (CO_2) precum şi a altor gaze cu efecte de seră au devenit o problemă majoră de mediu datorită contribuției intensive a acestora asupra fenomenului de încălzire globală. În ultimele decenii îndepărtarea CO_2 din amestecurile de gaze a devenit o problemă industrială importantă datorită necesității purificării diverselor surse de gaze precum şi din motive de recuperare având în vedere numeroasele aplicații ale CO_2 în scopuri industriale şi alimentare. Pentru îndepărtarea CO_2 din fluxurile gazoase contaminate s-au aplicat diverse tehnologii. Aceste tehnologii au la bază diferite procese fizice, chimice şi biologice. În acelaşi timp pentru îndepărtarea CO_2 au fost raportate diverse tipuri de contactoare precum: vase cu amestecare, coloane cu umplutură, coloane cu barbotare, reactoare membrană. Lucrarea de față prezintă cele mai recente aplicații ale reactoarelor gaz-lift la tratarea fluxurilor gazoase contaminate, în particular potențialul aplicării la îndepărtarea CO_2 din fluxurile de gaz, având în vedere cele mai importante avantaje ale acestor aparate: proiectare şi construcție simplă, lipsa pieselor în mişcare, viteze mari de transfer de masă şi căldură, amestecarea intensă a fazelor, forțe de forfecare reduse care creează condiții optime de cultivare a unor organisme sensibile la forțe de forfecare intense, consum energetic redus, costuri operaționale scăzute. Pe baza informațiilor din literatura de specialiate lucrarea prezintă o analiză a celor mai relevante caracteristici şi proprietăți ale CO_2 precum şi a celor mai aplicate tehnologii de îndepărtare a CO_2.