

# **Prospective Synergy of Biogas Upgrading Technologies with Carbon Capture and Sequestration (CCS) Techniques**

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## **ABSTRACT**

The alternatives options pertaining to the use of separated impurities such as carbon dioxide (CO<sub>2</sub>) have attracted high interest over the recent years. CO<sub>2</sub> utilization is offering many additional economic benefits to biogas upgrading technologies. By means of these technologies, the separated CO<sub>2</sub> can be used for various purposes as an eco-friendly and financially sustainable product in enhanced oil recovery (EOR), algae production, mineralization and carbon sequestration. On the one hand, the anthropogenic climate change and the mitigation thereof are the main reasons of the increasing interest for CCS technologies. On the other hand, the financial aspect of upgrading technologies is still an important criterion always considered before the commencement of projects due to the production of impurities. To optimize the economic viability of biogas, this study proposes a synergy between biogas upgrading and CCS technologies to generate tremendous benefits in the future. This study aims at promoting the deployment of biogas and CO<sub>2</sub> utilization to avail governmental incentives and subsidies in order to eradicate present hurdles in the energy sector.

## **Keywords**

Biogas, carbon, capture, synergy, upgrading.

## **I. BIOGAS UPGRADING TECHNOLOGIES**

Biogas is essentially a mixture of  $\text{CH}_4$  and  $\text{CO}_2$  [1]. It is formed from the Anaerobic digestion (AD) or anaerobic co-digestion of organic compounds [2]. During AD, a bio-degradation takes place in the presence of microorganisms (bacteria) that develop an affiliation with hydrogen ( $\text{H}_2$ ), carbon dioxide ( $\text{CO}_2$ ) and acetic acid ( $\text{CH}_3\text{COOH}$ ) by decomposing materials while simultaneously producing methane ( $\text{CH}_4$ ) [1]. AD is defined as the process that involves various metabolic interactions among distinct groups of bacteria [3]. It is carried out in digesters that are maintained at temperatures ranging from 30 to 65 Degree Celsius [3]. Biogas under AD is formed from organic compound such as manure, food waste (FW), crop residues and sewage considered as biomass [1, 4] which can be liquid or solid and used as waste feedstock for biogas [5].

When raw biogas is produced, it is afterwards purified through the capture of  $\text{CO}_2$  and removal of impurities such as  $\text{H}_2\text{S}$ . Upon  $\text{CO}_2$  capture and contaminants removal,  $\text{CH}_4$  subsequently enriches. The raw biogas becomes purified and upgraded into biomethane by means of upgrading technologies. This section outlines pros and cons of most preferred upgrading technologies.

Technologies of biogas upgrading can be divided into four main groups: Adsorption, absorption, permeation and cryogenic techniques [6]. The adsorption group includes techniques such as pressure swing adsorption (PSA). Other technologies such as absorption occurs in two ways: Physical and chemical absorption. Both types can be operated using inorganic or organic solvents meanwhile the permeation technique usually takes place in high pressure or low pressure depending on the desiring final product. Cryogenic technique also makes uses of high pressure and low-pressure separation. In certain cases, the cryogenic method incorporates chemical absorption to produce a high purity liquid and biomethane to minimize the loss in methane [4, 6].

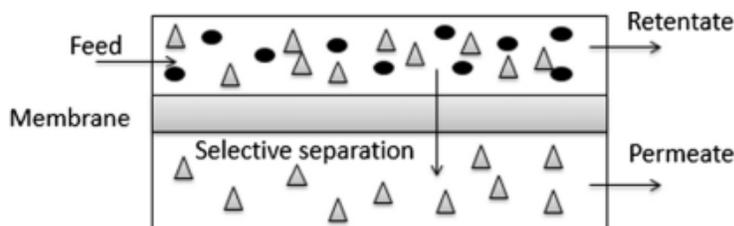
The PSA method is an absorptive biogas upgrading technology firstly introduced in the 1960s which consists in retaining gas compounds (adsorbates) on the surface of solids (adsorbents) according to the molecular size [4, 6]. Usually activated carbon, zeolites, or carbon molecular sieves are used as adsorbents. The PSA approach enables the separation of methane from  $\text{N}_2$ ,  $\text{O}_2$  and  $\text{CO}_2$  due to the fact the methane molecule is larger than any other gas molecules [6]. Often, the irreversibility of the adsorption  $\text{H}_2\text{S}$  caused by the toxicity of the compound is the reason of the prior removal of  $\text{H}_2\text{S}$  before PSA [4]. It is recommended that  $\text{H}_2\text{O}$  be removed prior to PSA as water dew points in the product gas of -100 to -80 °C can be reached. It has emerged that 96-98 % of methane is obtained after upgrading while 2 - 6 % of methane can be lost in the process. Other studies conducted by Lindberg [7] and Patterson et al., [8] have shown that a loss of 10 to 12 % of methane was also possible in some instances.

Water scrubbing is an absorptive technology often used in the upgrading of biogas in which water is used as the only inorganic solvent water [4,6]. Contrarily to adsorption, water scrubbing is an absorptive method which consists in dissolving gas in liquid [6]. Due to the difference existing between physisorption and chemisorption,

water scrubbing is classified as a physisorption based on the reversibility caused by physical bonding forces commonly known as the Van der Waals Force [6].

Physical absorption is also called physisorption like water scrubbing. However, in this physisorption process, an organic reagent such as polyglycol dimethyl ethers is used as an absorption agent in processes such as the commonly called Selexol process [6]. This organic reagent has a higher absorption rate for CO<sub>2</sub> compared to water. The particularity of this method lies in the capability to parallelly absorb CO<sub>2</sub>, H<sub>2</sub>S and H<sub>2</sub>O in the absorption column. Due to the formation of unwanted reaction products in the process, NH<sub>3</sub> is separately absorbed [6].

Conventionally known as gas permeation, membrane separation consists in purifying raw gas by means of permeabilities of different gas compounds through polymer membranes [6]. In other words, it is a separation technology at molecular scale. It has the merits of being cost effective, easy to process, and energy efficient [4]. It consists of permeating CO<sub>2</sub> through the membrane while CH<sub>4</sub> is retained at the inlet side. Beil and Wiebkebeyrich, [6] defines the membranes systems in three important flows: the feed at the inlet side (where the raw biogas is injected), the permeate (through which CO<sub>2</sub> penetrates) and the retentate (where methane is retained).



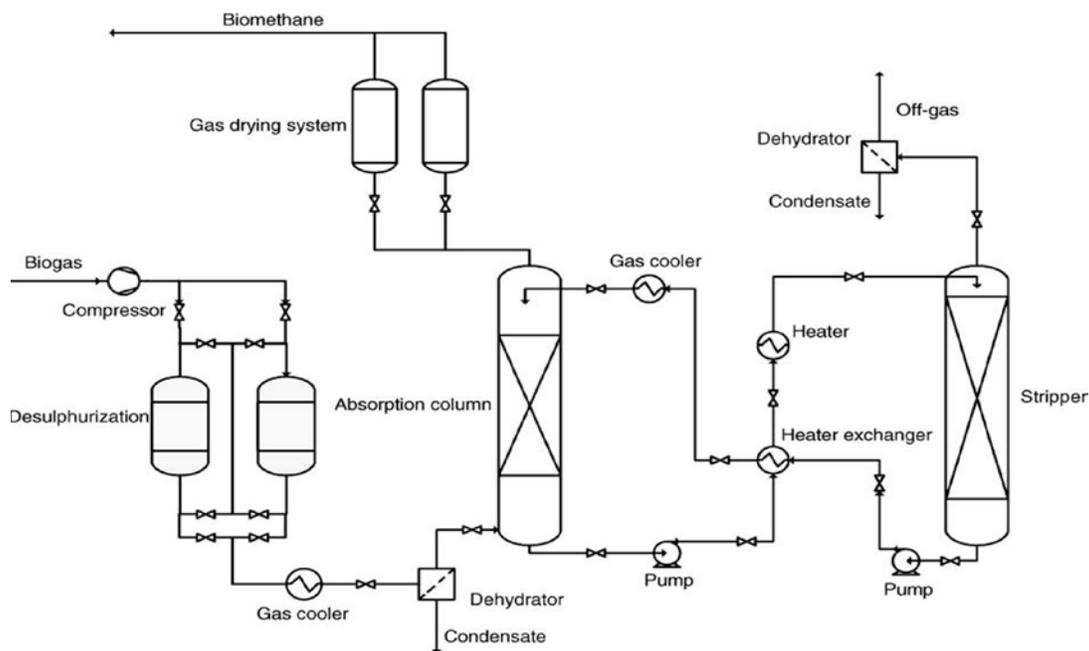
**Figure 1: Process flow diagram of membrane separation [10].**

The Cryogenic upgrading is a technology under development which aims at separating CH<sub>4</sub> and CO<sub>2</sub> based on the effect of different boiling points of CH<sub>4</sub> (-161°C) and the sublimation points of CO<sub>2</sub> (-78,48°C) [4,6]. Although this method is still under development at a pilot scale, there are presently few commercial plants in operation. The advantage of cryogenic processes is the production of CH<sub>4</sub> of 98% purity with a possible loss of more or less 1%. However, it shows a great disadvantage of energy consumption as it can require up to 200 bar to compress raw biogas to obtain a large amount of energy [4, 6].

Chemical absorption is a chemisorptive technology that utilizes organic solvents to bond impurities such as CO<sub>2</sub> and H<sub>2</sub>S to an organic scrubbing agent. It is also known as an amine scrubbing method due to different alkanolamine and ethanolamine solutions mainly used for the separation of CO<sub>2</sub> in chemical absorption process [6]. Since the 1970s, monoethanolamine (MEA), diethanolamine (DEA) and methyldiethanolamine (MDEA) are proven to be the most relevant and preferred for biogas upgrading and especially in the separation of CO<sub>2</sub> and H<sub>2</sub>S in pungent gases [8, 9].

The chemical absorption is well-preferred because of its capability to recover up to 99% of methane from the raw biogas due to the minimal loss of methane in the system compared to other upgrading technologies. In figure 2, it is shown that the absorption process takes place without a flash column as the raw biogas is fed and desulphurized by means of the amino acid salt solution used in the system and afterwards interact with the amine solution in the absorption column in cross-flow [6].

Under the system depicted in figure 2, CO<sub>2</sub> and H<sub>2</sub>S can be co-absorbed in chemical absorption processes. However, as in previous upgrading technologies, a desulphurization step is often installed prior to the absorption phase occurring in the absorption column to reduce uncontrollable factors such as the energy demand caused by the regeneration process [6].



**Figure 2: Process flow diagram of chemical absorption [6].**

The chemical absorption has the merits of producing methane of high purity compared to others aforementioned technologies. The high upgrading and cost-effectiveness of the technology are among the Nowadays, there are many optimization designs proposed to improve the efficiency and availability of these technologies. For instance, water scrubbing has been optimized in terms of fresh water demand. PSA has been improved to reduce the number of costly pressure valves and use cost-effective absorbents. Chemical absorption has been optimized to improve the regeneration process of appropriate solutions. Presently, cryogenic upgrading is being improved to produce liquefied biogas (LBG) that meets the natural gas standards [10].

## **1.1. COMPARATIVE ANALYSIS OF BIOGAS UPGRADING TECHNOLOGIES**

The financial aspect in the selection of biogas is one of the most important criterion which should be considered before the commencement of any biogas upgrading project. However, the quality of desired outcomes is also paramount and must not be overlooked at any cost. For this reason, table 2 presents a comparison of the most dominant biogas upgrading technologies in relation to cost parameters such as investment, running and maintenance which are membrane separation, PSA, water scrubbing, chemical absorption and cryogenic upgrading separation [10]. Few years ago, membrane separation was reported a very expensive and inefficient technology. It has attracted high interest in the green market due to the availability of highly selective and low cost polymeric membranes. The challenges of high loss of methane, purity and uncertainty of the technology lifespan are still barriers to its wide implementation [10]. The rate of biogas upgrading plants have grown exponentially in Europe. Table 1, provides a summary of the drastic growth which occurred in selected European countries.

**Table 1: Biogas upgrading plants and respective capacities in selected European countries [10]**

<b>Country</b>	<b>Biogas plants</b>	<b>Biogas upgrading plants</b>	<b>Upgrading capacity (Nm<sup>3</sup>/h)</b>
<b>Germany</b>	94	120	204 082
<b>Austria</b>	9066	10	5160
<b>Italy</b>	1264	1	540
<b>Sweden</b>	187	53	38 858
<b>Netherlands</b>	211	16	16 720
<b>UK</b>	-	-	18 957
<b>Switzerland</b>	-	-	6310

Results in table 1 reveals the large development of biogas technology over the years and the gap between the production of raw biogas and the upgrading process. In Germany, it appears that the reduction of CO<sub>2</sub> is highly important based on the higher number of biogas upgrading plants compared to biogas plants presently established. In countries such as Austria, Italy and Netherlands there are seemingly large discrepancies between the number of biogas plants and biogas upgrading plants presents. This evaluation shows how crucial the economy of biogas needs to be reviewed to obtain cost effective strategies of implementation.

Table 2 provides an overview of prerequisites of different biogas upgrading technologies. Although there are five technologies presently attracting interests, only four of them are in constant operation. Cryogenic upgrading separation proposes many positive outcomes which are still under thorough development [10].

**Table 2: Biogas upgrading plants and respective capacities in selected European countries**

<b>Factors</b>	<b>Water Scrubbing</b>	<b>Chemical Absorption</b>	<b>PSA</b>	<b>Membrane Separation</b>	<b>Cryogenic Upgrading</b>	<b>Sources</b>
<b>Total investment cost (€)</b>	265 000	869 000	680 000	749 000	908 500	[11]
<b>Total running cost (€)</b>	10 000	179 500	187 250	126 750	397 500	[11]
<b>Gas Processing cost (€/Nm<sup>3</sup>)</b>	0.13	0.28	0.25	0.22	0.44	[11]
<b>Gas Processing cost (€/ Nm<sup>3</sup>)</b>	0.16	0.35	0.31	0.27	0.55	[11]
<b>Product Flow rate (Nm<sup>3</sup>)</b>	144	137	139	130	161	[11]
<b>Efficiency</b>	High	High	Low	Low	High	[6, 10]
<b>Economic viability</b>	Low	Moderate	Moderate	High	Low	[6, 10]
<b>Regeneration</b>	Possible	Recommended	Possible	-	-	[11]
<b>MJ/ton of CO<sub>2</sub> removed</b>	770	433	915	1264	1275	[10]
<b>Technical Availability per year (%)</b>	96	91	94	98	-	[10]
<b>Maintenance Cost (€/year)</b>	15000	59 000	56 000	25 000	-	[10]

Nowadays, there are many optimization designs proposed to improve the efficiency and availability of these technologies. For instance, water scrubbing has been optimized in terms of fresh water demand. PSA has been improved to reduce the number of costly pressure valves and use cost-effective absorbents. Chemical absorption has been optimized to improve the regeneration process of appropriate solutions. Presently, cryogenic upgrading is being improved to produce liquefied biogas (LBG) that meets the natural gas standards [10].

**Table 3: Key selection factors of preferred biogas upgrading technologies**

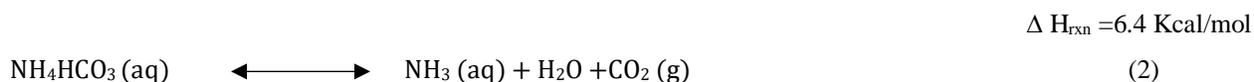
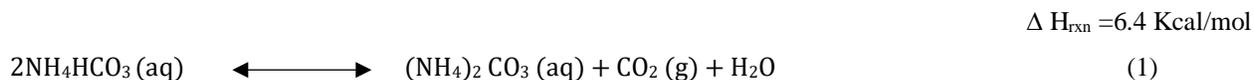
	PSA	Water Scrubbing	Physical Absorption (Organic Solvents)	Chemical Absorption (Organic Solvents)	Membrane (High Pressure /Dry)	Cryogenic Upgrading	Sources
<b>Electricity Demand (Kwh/ m<sub>n</sub><sup>3</sup> BG)</b>	0.16-0.35	0.20-0.30	0.23 - 0.33	0.06 - 0.17	0.18-0.35	0.18-0.25	[6,12,13]
<b>Heat demand (Kwh/ m<sub>n</sub><sup>3</sup> BG)</b>	0	0	0.10 - 0.15	0.4 - 0.8	0	0	[6, 12]
<b>Temperature process Heat (in the column) (°C)</b>	-	-	40-80	106-160	-	-	[6, 14]
<b>Operation Pressure (Bar)</b>	1-10	4-10	4 - 8	0.05 - 4	7-20	10 - 25	[6,14,15, 16]
<b>Methane Loss (%)</b>	1.5 -10	0.5 -2	1- 4	~ 0.1	1- 15	0.1-2.0	[6,16, 17,18]
<b>Methane Recovery Rate (%)</b>	90-98.5	98-99.5	96-99	~ 99.9	85-99	98-99.9	[6,12,14, 19]
<b>Precision Desulphurization required</b>	Yes	No	No	Yes	Recommended	Yes	[13,43]
<b>Water demand</b>	No	Yes	No	Yes	No	No	[6]
<b>Chemical substances demand</b>	No	No	Yes	Yes	Yes	No	[6]
<b>Removal O<sub>2</sub> / N<sub>2</sub></b>	Possible	No	No	No	Possible	Possible	[4,6,8,9]

## 1.2. REGENERATION PROCESSES OF ORGANIC SOLVENTS

The regeneration process is a method that consists in separating the impurities absorbed by organic solvents during biogas upgrading in order to re-use them for future biogas upgrading processes. In other words, regeneration processes consist in restoring organic solvents and absorption by-products to their initial stages by means chemical interactions. There are several regeneration processes selected based on organic solvents used in absorption [11].

### 1.2.1. Ammonia (NH<sub>3</sub>) regeneration processes

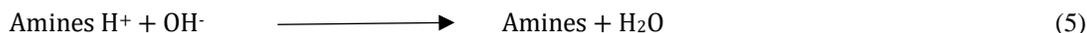
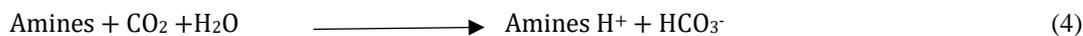
NH<sub>3</sub> is often used to reduce acid pollutants and unwanted components in gases such as CO<sub>2</sub>. Studies reveals that 80% removal efficiency is guaranteed during absorption using NH<sub>3</sub> [20]. During the regeneration process, NH<sub>3</sub> is thermally decomposed by giving off the absorbed CO<sub>2</sub> so that the solvents as well as other absorption by-products can be used in the absorber again. In the following equations, there is an illustration of the thermal decomposition which takes place.



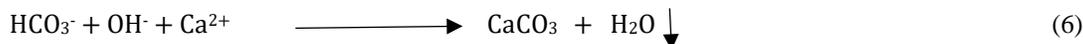
### 1.2.2. Amines regeneration processes

Amines are highly preferred chemical absorbents for CO<sub>2</sub> capture due to their high efficiency proven by low methane loss and high methane recovery. Unlike many other solvents, amines are reported to be easily regenerated for re-utilization and absorption processes. García-Abuín et. al, [21] illustrates in the following equations, chemical interactions of amines during absorption and regeneration processes.

- **Absorption**



- **Regeneration**



As shown in the above equations, Calcium Hydroxide Ca(OH)<sub>2</sub> is often used for tertiary amines such as Pyrrolidine (PYR), methyl diethanolamine (MDEA), triethanolamine (TEA) and tri-isopropanolamine (TIPA).

As CO<sub>2</sub> passes through aqueous solution of the amines, bicarbonate and carbamate are produced as main products. Most regeneration processes aim at obtaining bicarbonate and not carbamate. For this reason, carbamate avoidance is always recommended for an effective regeneration process [21]. Among all amines, triethanolamine and tri-isopropanolamine are reported to have a high potential regeneration capacity of almost 100 % when effectuated with Ca(OH)<sub>2</sub>.

### **1.2.3. Alkali regeneration processes**

The alkalis are known as good absorbents due to their capacities to improve absorption in physiosorption processes. However, their high energy demand inconveniences their regeneration [22]. The alkali regeneration can take in two major ways: electro-dialysis and causticization [23, 24]. Electro-dialysis consists in regenerating aqueous solution of Sodium Hydroxide (NaOH) by means of an electro-membrane separation process in which an electro-membrane separation process in which an electric field is placed to cause a migration of ions from one solution to the other through membranes permeation [23]. Due to its negative environmental impact, bipolar membrane electro-dialysis was proposed because of its lower voltage drop [23]. Causticization consists in regenerating NaOH from Sodium Carbonate using lime and titanium oxide in conventional or direct ways, respectively [24, 11].

### **1.2.4. Regeneration processes of H<sub>2</sub>S removing absorbents**

The chemicals often used in H<sub>2</sub>S removal are the following NaOH, FeCl<sub>2</sub>, FeCl<sub>3</sub>, FeSO<sub>4</sub>, Fe<sub>2</sub>O<sub>3</sub>, Fe (OH)<sub>3</sub> and ZnO. By means of the following reactions, these components react with H<sub>2</sub>S [11].



Despite H<sub>2</sub>S removal as shown in equations 7 to 9, only few components can be regenerated. In studies conducted by Maile [11], it has emerged that Na<sub>2</sub>S (Sodium Sulfide) is non-regenerative and FeS, very insoluble for an effective regeneration. As such, only Fe<sub>2</sub>S<sub>3</sub> can be regenerated using Oxygen as illustrated in equation 10.

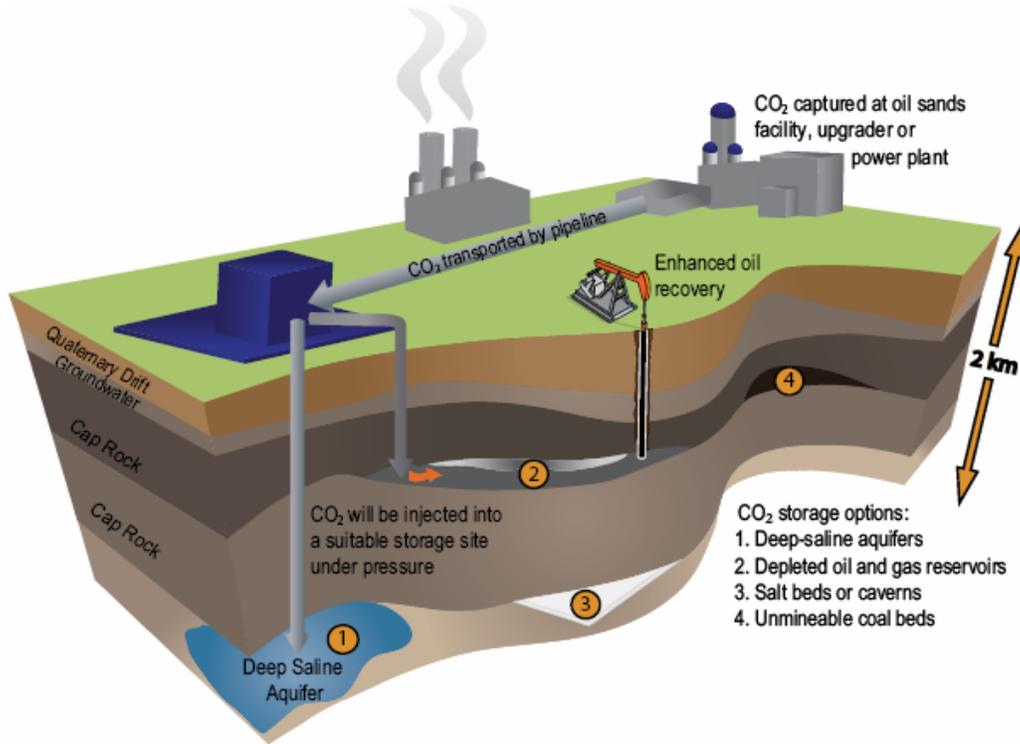


## **II. Carbon capture and sequestration (CCS) technologies**

The anthropogenic climate change and the mitigation thereof are the main reasons of the increasing interest for CCS technologies as they promote the capture, storage and utilization of CO<sub>2</sub> [10]. CCS is the ensemble of processes that shelves CO<sub>2</sub> from emission to the atmosphere. It consists on capturing CO<sub>2</sub> at an emission source such as biogas plant or power plant, compressing and transporting it to a storage site by using pipelines. As illustrated in figure 3, the captured, compressed and stored CO<sub>2</sub> is injected into rock formations where it could possibly remain for millennium [10, 25, 26].

As shown in figure 3, there are four storage options of CO<sub>2</sub> after their transportation through pipeline. CO<sub>2</sub> can be stored in deep saline aquifers, depleted oil and gas reservoirs, salt beds or caverns and finally in unmineable coal beds.

Unmineable coal beds can be used to recover methane by enhanced coal bed methane (ECBM) recovery although it can be inconvenient due to coal use for CO<sub>2</sub> storage. Saline aquifers are potential materials in storage project onshore and offshore. Salts bed, also called as basalts represents an enormous volume for CO<sub>2</sub> storage which can be fixed or cleaned with carbonate minerals treatment [27, 28].



**Figure 3: Carbon capture and sequestration (CCS) [28].**

Current studies have shown that large volumes of CO<sub>2</sub> can be transported through pipelines by considering the purity of CO<sub>2</sub>. It is reported that the suitable method for CO<sub>2</sub> transportation capable to produce the high-purity CO<sub>2</sub> is cryogenic separation technology [4, 10, 26]. Table 4 presents a recommendation of CO<sub>2</sub> transportation standards to be considered.

**Table 4: Pipeline standards recommendations [4].**

Component	Concentration		
	DYNAMIS	CO <sub>2</sub> Europe	NETL
CO <sub>2</sub>	> 95.5% (mol)	> 95.5% (mol)	95.5% (mol)
H <sub>2</sub> O	500 ppm	No free water	300 ppm <sub>wt</sub>
H <sub>2</sub> S	200 ppm	235 ppm	0.01% (mol)
CO	2000ppm	4750ppm	35ppm
CH <sub>4</sub>	Aquifer < 4% (mol)	Along with Ar, H <sub>2</sub> , N <sub>2</sub> , O <sub>2</sub> < 5 % (mol)	4 % (mol)

<b>N<sub>2</sub></b>	< 4 % (mol.,all non-condensable gases)	Along with Ar, H <sub>2</sub> , N <sub>2</sub> , O <sub>2</sub> < 5 % (mol)	4 % (mol)
<b>Ar</b>	< 4 % (mol.,all non-condensable gases)	Along with Ar, H <sub>2</sub> , N <sub>2</sub> , O <sub>2</sub> < 5 % (mol)	4 % (mol)
<b>H<sub>2</sub></b>	< 4 % (mol.,all non-condensable gases)	Along with Ar, H <sub>2</sub> , N <sub>2</sub> , O <sub>2</sub> < 5 % (mol)	4 % (mol)
<b>O<sub>2</sub></b>		Along with Ar, H <sub>2</sub> , N <sub>2</sub> , O <sub>2</sub> < 5 % (mol)	4 % (mol)
<b>NO<sub>2</sub></b>		75 ppm	100 ppm
<b>SO<sub>2</sub></b>		75 ppm	100 ppm
<b>HCN</b>		70 ppm	Trace
<b>COS</b>		235 ppm	Trace
<b>NH<sub>3</sub></b>		550 ppm	50 ppm
<b>Glycol</b>			46 ppb
<b>C<sub>2</sub> H<sub>6</sub></b>			1 % mol

Given different recommendation from expert such as DYNAMIS, Europipe and National Energy Technology Laboratory, CO<sub>2</sub> must have a purity of not less that 95% with trace element composition specified therein. Similarly to biogas upgraded into biomethane, CO<sub>2</sub> can also be purified and upgraded into a more valuable component by means of upgrading technologies such as chemical and physical adsorption, PSA, and membrane technologies. These technologies enables to obtain a purity over 85 %.

Although chemiabsorptive regeneration processes were proposed as potential ways of reducing cost by reconsidering chemical solvents used in CO<sub>2</sub> capture, the cost of regeneration processes remained high and stopped its wide adoption. Although, membranes technologies seemed more economically effective than others, the loss of methane remained high and low methane recovery remained low. As such, there was an urge to reduce further operational cost by increasing the profitability of technologies in place. In this perspective, alternatives options pertaining to the use of separated impurities often discarded such as CO<sub>2</sub> has attracted high interest over the last recent years.

The synergy between biogas upgrading and carbon capture, storage and utilization (CCSU) technologies is paramount. Merging these two approaches will certainly require a very high start-up investment capital but will generate a tremendous benefit in the future. By means of considerable economic incentives and subsidies, hurdles pertaining to renewable energy deployment will be resolved.

### **III. CONCLUSION**

The economic viability of biogas upgrading technologies is still an important question today. Although regeneration processes were proposed as potential ways of reducing cost by reconsidering chemical solvents used in CO<sub>2</sub> capture chemical absorption, the cost of regeneration processes remained high and stopped its wide adoption. Although,

membranes technologies seemed more economically effective than others, the loss of methane remained high and low methane recovery remained low. As such, there was an urge to reduce further operational cost by increasing the profitability of technologies in place. In this perspective, alternatives options pertaining to the use of separated impurities often discarded such as CO<sub>2</sub> has attracted high interest over the last recent years. CO<sub>2</sub> utilization is offering many additional economic benefits to biogas upgrading technologies. Upon appropriate separation, the separated CO<sub>2</sub> can be used for various purposes as an eco-friendly and financially sustainable product.

#### **IV. ACKNOWLEDGEMENT**

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