Review

Methods and Techniques for CO$_2$ Capture: Review of Potential Solutions and Applications in Modern Energy Technologies

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Abstract: The paper presents and discusses modern methods and technologies of CO$_2$ capture (pre-combustion capture, post-combustion capture, and oxy-combustion capture) along with the principles of these methods and examples of existing and operating installations. The primary differences of the selected methods and technologies, with the possibility to apply them in new low-emission energy technologies, were presented. The following CO$_2$ capture methods: pre-combustion, post-combustion based on chemical absorption, physical separation, membrane separation, chemical looping combustion, calcium looping process, and oxy-combustion are discussed in the paper. Large-scale carbon capture utilization and storage (CCUS) facilities operating and under development are summarized. In 2021, 27 commercial CCUS facilities are currently under operation with a capture capacity of up to 40 Mt of CO$_2$ per year. If all projects are launched, the global CO$_2$ capture potential can be more than ca. 130–150 Mt/year of captured CO$_2$. The most popular and developed indicators for comparing and assessing CO$_2$ emission, capture, avoiding, and cost connected with avoiding CO$_2$ emissions are also presented and described in the paper.

Keywords: carbon capture and storage installation; CO$_2$ capture methods; CO$_2$ emission level assessment indicators

1. Introduction

With the increase in electricity consumption around the world, electricity demands are increasing every day. During electricity generation using energy technologies based on fossil fuels, the emission of harmful pollutants into the environment (gaseous, liquid, and solid) occurs as the emission of NOx, SOx, dust, CO$_2$, and wastewater (e.g., from flue-gas treatment installations) [1–3]. Last year, a great deal of effort in modern low-emission energy technologies was directed at activities leading to decreased gaseous pollutant emissions [4,5]. The emission of carbon dioxide (CO$_2$), which is treated as one of the main reasons for global warming when fossil fuel is burned, cannot be avoided. Fossil-fueled power-production technology plays a significant role in contributing to the emission of greenhouse gases into the atmosphere. By reducing the emission of CO$_2$ into the atmosphere, and by switching to an alternative power generation with zero-emission, it is possible to prevent future catastrophic effects. The carbon capture utilization and storage (CCUS) methods and technologies are among the many ways to reduce CO$_2$ emissions. CCUS technologies aim to capture CO$_2$ from large industrial sources and store it in underground structures, or use it through conversion into useful products [6]. All this is happening while emissions from the industrial and energy sectors are reduced, which makes this process one of the most current scientific research endeavors, while also presenting socio-economic challenges. The current fees related to CO$_2$ emissions in the European Union amount to over 50 EUR/tCO$_2$ [7], and there is an expected upward trend for coming years, forced by political declarations and treaties.

There are four different ways to reduce CO$_2$ emission levels [8]:

1. Reducing the use of fossil fuels by:
   ○ improving the efficiency of energy conversion processes;
   ○ reducing the demand for energy;
   ○ using renewable (non-fossil fuel) energy sources, such as hydropower, wind, biomass, solar cells, and nuclear power;
   ○ increasing the use of green hydrogen, which is produced by splitting water using electricity from renewable energy.
2. Replace technologies using fossil fuels with a low carbon to hydrogen C/H_2 ratio by replacing coal and oil with gaseous fuels.
3. Capturing CO_2 from fuel combustion in power plants and other industrial processes and storing it in appropriate geological structures, in exhausted or exploited gas or crude oil deposits (intensification of crude oil extraction, enhanced oil recovery (EOR)), or at the bottom of oceans.
4. Limiting deforestation processes and thus storing more CO_2 in biomass.

Carbon capture utilization and storage (CCUS) is a family of methods to reduce the emission of CO_2 from fossil-fueled power plants. The CCS can be coupled with Power Plants (PPs) and Combined Heat and Power Plants (CHPPs) to reduce the emission of CO2 in the flue gas. First-generation carbon capture technology had a lower efficiency in carbon capture, and was challenging to integrate with the complex structures of a power plant. With improved research and development, the second- and third-generation power plants using carbon capture technology showed improved efficiency and a low cost compared to first-generation CCS technology [9]. The three different methods (pre-, post-, and oxyfuel combustion) for CO_2 capture and separation are under development. The oxyfuel combustion method is considered a promising solution from an energy-efficiency power-generation point of view. Authors have noted, that the energy penalty for the oxy-combustion method can be around 4%, in comparison to 8–12% for post-combustion methods [10].

The implementation of CCS in Europe is focused on two major factors, the development of power generation technology with carbon capture at a low cost and selling CO_2 at a high price to reimburse the cost of CO_2 transportation. In 2008, the European Parliament approved the "draft CCS directive", which aims to guide CO_2 geological storage. Due to public opposition from European Union countries for underground CO_2 storage, many countries allow only offshore storage projects [11]. CCUS technologies are considered as crucial technologies for the European Commission, and are explicitly included in, e.g., the European Green Deal. Nowadays, more and more important projects at the industrial scale are funded by the Innovation Fund (https://ec.europa.eu/clima/eu-action/funding-climate-action/innovation-fund/large-scale-projects_en, accessed on 20 January 2022). The International Energy Agency forecasts that CCS will contribute up to 21% of the reduction in CO_2 emissions into the atmosphere. Many countries, including Asian countries, still depend on coal-fired power production because of the low cost and reliability, and these cannot be wholly replaced with renewable energy systems. Some European countries, such as Poland, will depend on fossil-fuel power production for at least 30 more years. According to Polish Energy Policy, by the year 2050, electricity demand in Poland will be produced by renewable energy and future nuclear power projects. The use of coal for the next few years makes CCS technology inevitable [12]. Carbon capture and storage is a method for capturing the concentrated CO_2 in flue gas from fossil-fueled power plants and store them in one place. There are three methods of CO_2 capture: pre-combustion carbon capture, post-combustion carbon capture, and the oxy-combustion carbon capture method.

- Pre-combustion carbon capture occurs before the combustion process (through fuel gasification with oxygen, e.g., integrated IGCC coal gasification technology).
- Post-combustion carbon capture occurs after the combustion process (capturing CO_2 from flue gas, e.g., using chemical absorption, physical adsorption, membrane separation, or the use of a chemical loop).

- Oxy-combustion carbon capture occurs after the combustion process in an oxygen atmosphere by separating CO2 generated during the oxy-combustion process, e.g., using an oxygen gas turbine. Oxygen atmosphere can be obtained by removing nitrogen from the air before the combustion process.

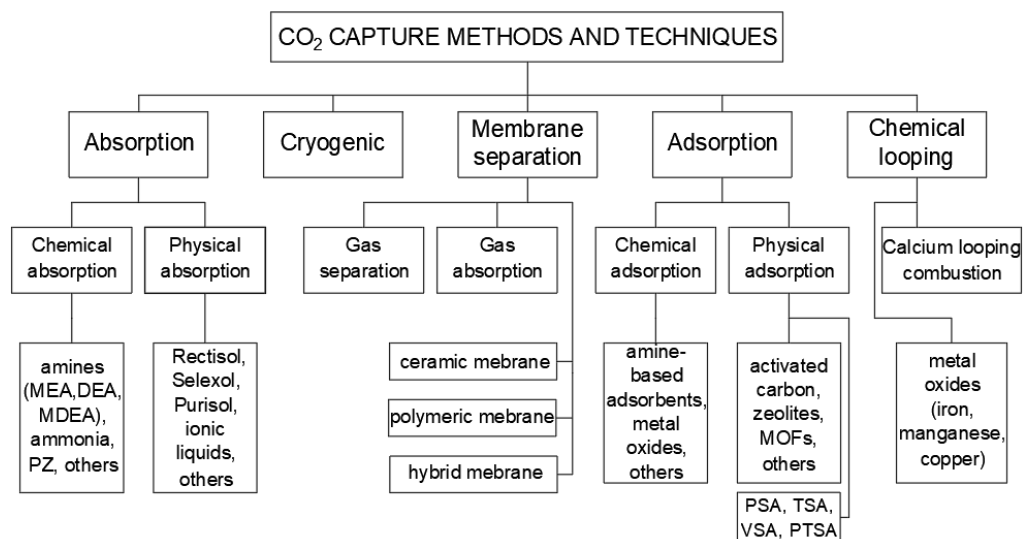A diagram explaining the methods and techniques for CO_2 capture is shown in Figure 1.

Figure 1. Methods and techniques of CO_2 capture.

In pre-combustion carbon capture, fuel is oxidized using a gasification process, which produces syngas with a composition of hydrogen (H_2) and carbon monoxide (CO). The produced CO is converted into CO_2, which is captured before combustion. Shijaz et al. showed a comparison between power generation using coal gasification without carbon capture, pre-combustion carbon capture, or chemical looping combat (CLC). The results showed that the overall efficiency of a power plant with pre-combustion carbon capture and CLC is reduced compared to a plant without CO_2 capture. CO_2 captured from fuel reduces the fuel volume sent to the turbine, reducing power generation. However, due to environmental concerns, including a CO_2 capture unit is unavoidable [13]. Mukherjee et al. compared each type of carbon capture method using an IGCC power plant without CO_2 capture. CLC was combined with a coal-fired IGCC, and analyses were performed based on electrical efficiency and carbon capture efficiency. The CLC and oxyfuel combustion methods showed a value of around a 100% carbon capture rate compared to pre-combustion CO_2 capture, which achieved 94.8% CO_2 capture. Another method of coal direct chemical looping combustion, where coal is fed directly to a boiler without gasification, increased the electrical efficiency and achieved 100% CO_2 capture. The results from the comparison of IGCC-CLC, pre-combustion, and oxyfuel combustion showed that the methods' energy penalties were 4.5%, 7.1%, and 9.1%, respectively [14]. This article describes the various carbon capture and storage methods and technology used in large-scale units. The CO_2 emission from a coal-fired ultra-supercritical power plant is calculated.

The SO_2 and NO_x content in the flue gas has a higher chance of affecting the purity of CO_2 during CO_2 capture. The high purity of the CO_2 stream is very important for recycling methods. A pilot, dual-reflux VPSA unit, installed in the Łagisza Power plant in Poland, was installed for post-combustion CO_2 capture. Before the flue gas from the boiler is sent to the DR-VPSA unit, the flue gas is passed through an absorber, an adsorber, and a glycolic gas dehydration system to remove SO_2 and NO_x. Activated carbon works effectively for the removal of SO_2 compared to the removal of NO_x, which leads to a high-purity CO_2 [15].

CO$_2$ emissions from the power sector are mainly caused by modern technologies, such as coal-fired, gas-fired, oil-fired, and combined cycle gas turbine (CCGT) power plants. Table 1 presents the CO$_2$ emissions and lower heating value depending on fossil fuel. From Table 1, it can be seen that CO$_2$ emissions depend on the content of the fuel. The higher the fuel content, the higher the CO$_2$ emissions will be. The increasing share of H$_2$ gives better properties to fossil fuels, taking into account the LHV and CO$_2$ emission levels. Natural gas consists mainly of CH$_4$ and is characterized by almost two times lower emissions than hard and lignite coal, and an almost two times larger LHV. This fact comes from gas fuel composition, where four atoms of hydrogen are inside the methane molecule of every carbon atom. Other modern power generating technologies, such as nuclear, renewable energy sources (RES), and hydrogen-based technologies, are less likely to produce emissions. A newly built CCGT power plant has CO$_2$ emissions of 350 kg$_{CO2}$/MWh without carbon capture, indicating the same emissions as a gas-fired power plant [16]. In the case of an ultra-supercritical power plant fired with coal, CO$_2$ emissions up to 700 kgCO$_2$/MWh can be produced. According to the type of coal-fired critical power plant used, CO$_2$ emissions range from 690 to 830 kgCO$_2$/MWh [17]. The use of fossil fuel in modern energy technology will continue until it is replaced with alternative technologies, such as renewable energies and power production without emission. Until these technologies are replaced with those without CO$_2$ emissions, CO$_2$ capture is unavoidable to reduce greenhouse gases and protect the environment.

Table 1. Emission of carbon dioxide (CO$_2$) during combustion of different hydrocarbon fuels [1–3,8].

Fuel	LHV	Emission
	MJ/kg	kgCO$_2$/GJ
Hard coal	>23.9	94.60
Lignite	<17.4	101.20
Crude oil	43.0	74.07
Petrol	43.4	66.00
Paraffin oil	41.5	71.50
Heating oil	42.8	77.37
Diesel	42.6	74.07
Natural gas	47.1	56.10
Hydrogen	120	0.00

2. Pre-Combustion CO$_2$ Capture

In this method, the fuel (coal, gas, biomass) is not completely combusted in the reactor, but is converted into a mixture of CO and H$_2$ in the reforming or gasification process. Subsequently, using the water–gas shift, CO$_2$ and H$_2$ are produced. Figure 2 shows a block diagram of the pre-combustion CO$_2$ capture method in a power plant.
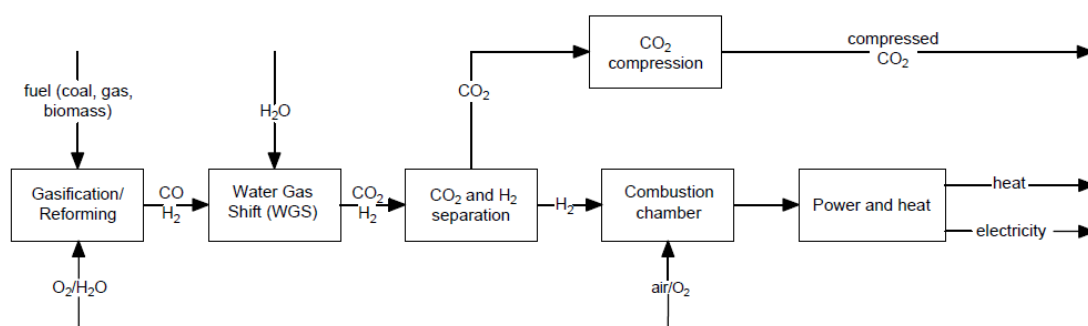

Figure 2. Block diagram of electricity generation heat production with the use of the pre-combustion CO$_2$ capture method.

Pre-combustion capture is used, e.g., in an integrated gasification combined cycle (IGCC). Carbon dioxide is removed after the gasification process. An example of a typical process for power and heat generation in a gas turbine with pre-combustion capture is shown in Figure 3.
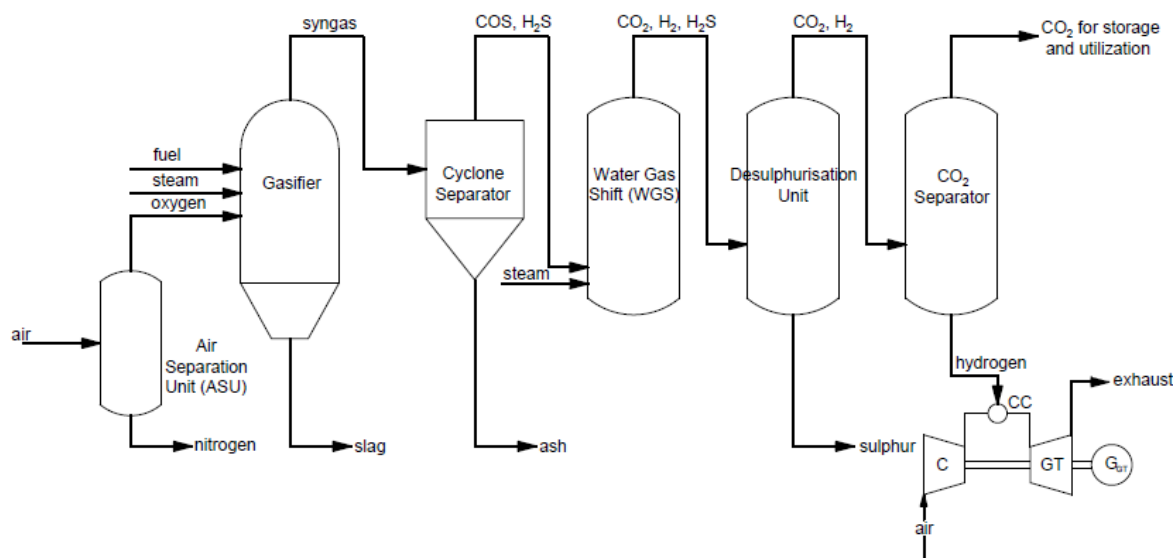

Figure 3. Scheme of integrated gasification combined cycle (IGCC) for electricity generation using a gas turbine using the pre-combustion CO_2 capture method.

In this process, steam and oxygen are provided to the gasifier to produce syngas enriched in hydrogen and carbon monoxide. Then, syngas is sent to a cyclone separator, where it is filtered to remove ash. After, the conversion of syngas and steam to CO_2 and H_2 occurs in the water–gas shift reactor. The received gas needs to be purified of sulfur in the desulfurization unit. Subsequently, CO_2 is captured in the CO_2 separator and is sent for storage or utilization. Received hydrogen is provided to the gas turbine as fuel [18]. Pre-combustion methods are very effective in CO_2 separation on the grounds of the high concentration of CO_2 in fuel before combustion. On the other hand, these processes are expensive due to the need for a gasification unit.

Pre-combustion carbon capture uses physical and chemical methods to capture CO_2 from processed syngas. Chemical absorbents, such as carbonates and physical solvents, such as polypropylene glycol and methanol, are commercially used in industries to capture CO_2. The cost expenditure and energy consumption of carbon capture depend on the utilities and capture process. An effective solvent or absorbent pre-combustion carbon capture technology can achieve more than 90% CO_2 capture, but, at the same time, reduces plant efficiency [19]. The calcium looping process is another method of pre-combustion CO_2 capture, where CO_2 capture is achieved effectively at a low cost. This method involves the sorption of CaO with CO_2 and the desorption of $CaCO_3$ to release CO_2 at an optimal temperature. This cycling process repeats multiple times, and waste heat from the gasifier is used to reduce the heat consumption of the CO_2 capture process. The CaL pre-combustion carbon capture method is highly effective. Low-cost and CO_2 capture are achieved by decreasing energy consumption [20]. The pre-combustion carbon capture demo plant in Port Arthur, United States, has successfully captured 1 million tons of CO_2 since it started operations, without problems. This plant proved that, using the dual pressure swing adsorption (PSA) technology method, purification of hydrogen >99.9% and a high efficiency CO_2 capture can be achieved. When the streaming gas has a low pressure, hydrogen purification is performed and the tail gas is sent to undergo vacuum pressure swing adsorption (VPSA) to separate purified CO_2. If the streaming gas has a high pressure,

CO_2 capture is achieved without VPSA first, and hydrogen purification is achieved from the exiting gas [21].

3. Post-Combustion CO_2 Capture

Post-combustion CO_2 capture methods are based on removing carbon dioxide from flue gas. The capture unit is placed after the purification systems, such as desulphurization, denitrogenation, and dedusting installations. Figure 4 shows a general block diagram of the post-combustion capture technique.
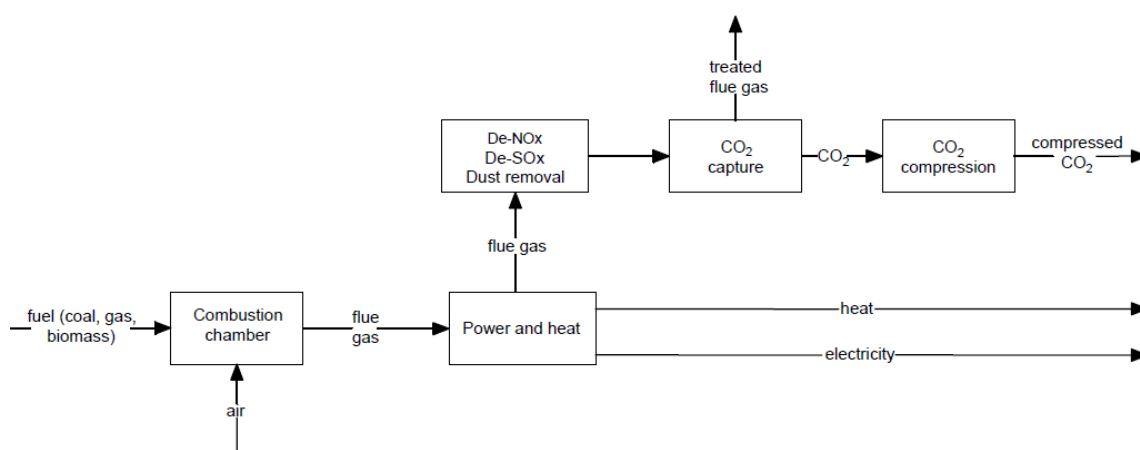
POST-COMBUSTION

Figure 4. Block diagram of electricity generation and heat production through the use of the post-combustion CO_2 capture method.

In existing conventional power units, post-combustion technologies are the most frequently considered [2,8]; nevertheless, there is one main barrier to using these methods. Since the partial pressure of CO_2 in the flue gas is low (flue gas is under atmospheric pressure and the concentration of carbon dioxide is within 13–15%), the driving force for CO_2 is also low [22]. Post-combustion technologies can be divided up according to the type of process used for capturing carbon dioxide, as follows:

(a)   Absorption solvent-based methods

Chemical absorption is the most recognizable method of CO_2 capture. It relies on a reaction between carbon dioxide and a chemical solvent. Solvents that are usually used are alkanolamines, such as monoethanolamine (MEA), diethanolamine (DEA), or methyl diethanolamine (MDEA) in aqueous solution [23]. A schematic diagram of chemical absorption is shown in Figure 5. The process takes place in two stages. In the first stage, the flue gas reacts with the solvent in the absorber to capture CO_2. Subsequently, the rich loading solution is carried to the stripper to regenerate CO_2 at elevated temperatures. The solution without CO_2 (lean-loading solution) is sent back to the absorber column. A high purity carbon dioxide stream from the desorber is transferred for compression and storage or utilization. The chemical absorption process has been used for a long time in the chemical industry. The typically used 30% MEA and MDEA solutions achieve a high process efficiency and a high degree of carbon dioxide purity [23]. The chemical absorption method is a very energy-consuming process due to the need to supply a large amount of heat to the desorber. It is assumed that approx. 30% (37%) of the heat supplied to the steam in the boiler should be directed to the CCS installation in the case of a steam unit fired with hard coal (lignite), depending on the absorber used (for ammonia, the amount of heat needed for regeneration is 22% for hard coal and 27% for lignite). Chemical absorption technologies are used in power plants fired with solid fuel, and they are the only ones

that are commercially available. It is assessed that the amine method can capture approx. 85–95% carbon dioxide included in flue gas with a purity above 99.95% [2].
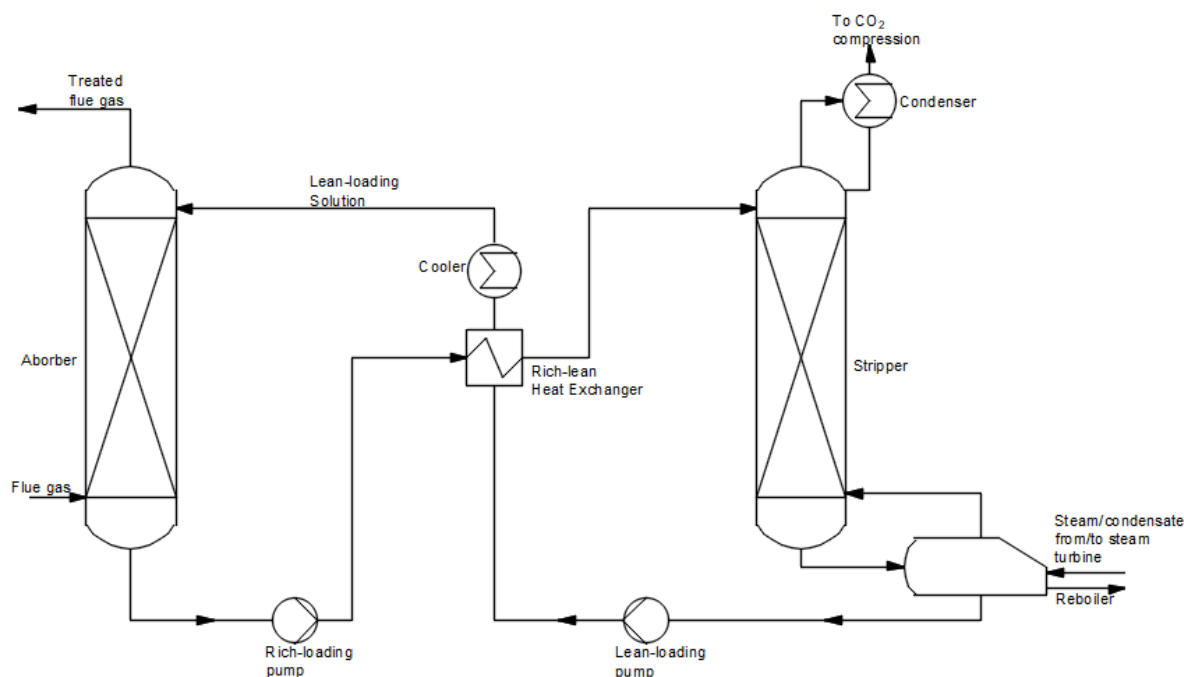

Figure 5. Scheme of the post-combustion CO_2 capture method using a chemical absorption process (based on [22]).

Nowadays, apart from conventional solvents (amine-based-MEA, DEA, ammonia, piperazine), there are other solvents developed for the CO_2 capture process. Solvent blends offer the ability to improve absorption properties by combining types properly. Primary and secondary amines have high absorption rates, and tertiary amines are characterized by a high capacity [8]. For example, blending MEA with a little PZ can improve the absorption rate (PZ is 50 times faster than MEA) [24]. Another possibility is to use a solution of 2-amino-2-methyl1-propanol (AMP) promoted with PZ. Artanto et al. showed that a mixture of 25 wt% AMP and 5 wt% PZ can be a good substitution for MEA [25]. Ionic liquids (ILs) are novel alternatives for amines. These low melting salts are comprised of a large organic cation and an arbitrary anion, which can be combined freely, obtaining a great variety of compound properties. ILs can physically or chemically absorb CO_2, depending on pressure [8]. A review [26] and articles [24,27] have presented deep insight into this technology. In the case of reducing energy consumption, there are new generation solvents—water-free solvents and biphasic solvents—that have been proposed. The presence of water in a solvent enhances the energy demand for the regeneration process. Novel water-free solvents, such as non-aqueous organic amine blends (methanol, ethylene glycol), aminosilicones, or amines with a superbase have been observed [24]. In [28], researchers showed that solvent mixtures based on ethylene glycol, used in the chemisorption process, can achieve CO_2 capture efficiencies of up to 95%. Deep eutectic solvents (DESs), such as choline chloride and ethylene glycol at a 1:2 mole ratio are getting more attention. They are fluids consisting of organic halide salts and metal salts or a hydrogen bond donor. DESs have similar properties to ILs, but they are cheaper and environmentally friendlier. According to [29], using DESs can decrease the vapor pressure of a solvent, achieve a lower effect of corrosion, and needs less energy in the regeneration process.

The physical absorption method is based on using a chemically inert solvent, which absorbs CO_2 physically. Absorption occurs in water or organic absorbers (methanol, N-methyl-2-pyrrolidone, dimethyl ether). This method achieves the best results for low

temperatures and high pressures of the separated gas. Therefore, it is used to capture CO_2 from the coal gasification process. In this method, there are distinguished processes, with the use of solvent such as Selexol^{TM}, Rectisol^{TM}, Ifpexol^{TM}, Fluor^{TM}, Purisol^{TM}, Sulfinol^{TM}, and Morphysorb^{TM} [23,24,30].

(b)   Adsorption–physical separation

Adsorption is a process that uses a solid surface to remove carbon dioxide from a mixture. Physical separation relies on adsorption, absorption, and cryogenic separation methods. It can be physical (Van der Waals forces for adhesion CO_2—physisorption) or chemical (covalent bonding between compounds—chemisorption) [31]. Physical adsorption uses various porous materials (such as activated carbon, alumina, metallic oxides, or zeolites [6]) to absorb carbon dioxide. Activated carbons contain amorphous carbon, and it is low-cost material with the advantage of having a large surface area and the possibility of modifying its pore structure. However, the weak binding energy with carbon dioxide causes this material to need to be highly microporous to be useful for carbon capture [8]. Zeolites (crystalline aluminosilicates) have good adsorption properties for CO_2 capture, but they are hydrophilic. The presence of water weakens these properties by reducing the strength of interactions between coupled compounds [26]. A new approach is to use metal–organic frameworks (MOFs) in adsorption processes. MOFs consist of metal ions or ion clusters linked by organic ligands and bridges that create strong coordination bonds. On account of this, MOFs are characterized as having benefits such as ease of design and synthesis, a high porosity, and tailored pore properties [32]. One of the other adsorption materials is silica. Silicas are non-carbonaceous substances with a large surface area and pore size, and they are highly mechanically stable. Mesoporous silica materials use amine-based substances for CO_2 capture [8,31]. The methods of adsorption are as follows: pressure swing adsorption (PSA), temperature swing adsorption (TSA), vacuum swing adsorption (VSA), and pressure–temperature swing adsorption (PTSA).

(c)   Membrane separation

Figure 6 depicts the membrane separation process. In the first place, flue gas is directed to an absorber to cool to the operating temperature of the membrane. Subsequently, flue gas is transported to the membrane. This method uses a spiral wound, flat sheet, and hollow fiber modules [30].
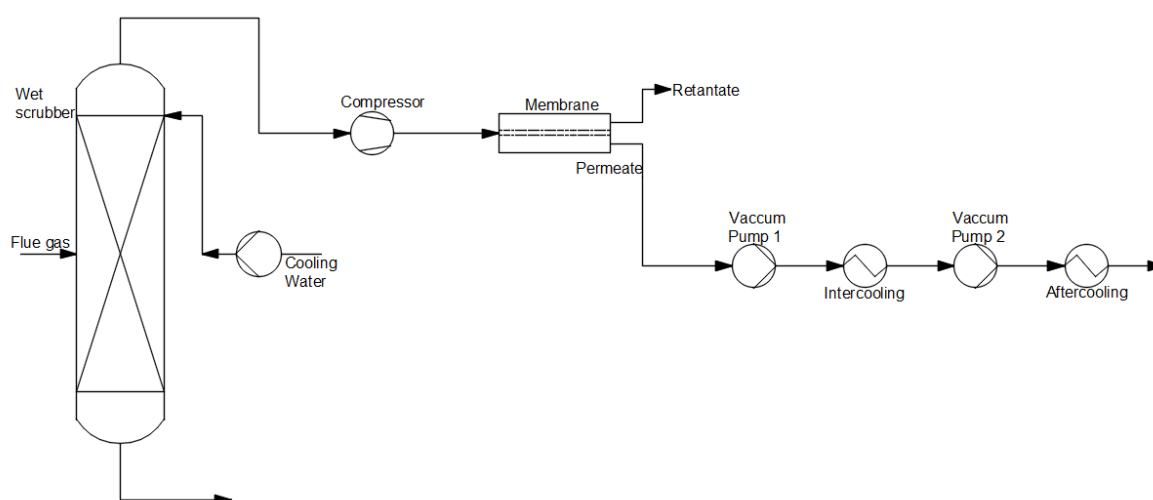

Figure 6. Scheme of the post-combustion CO_2 capture method using a membrane separation process (based on [22]).

There are two types of membrane capture technology: gas separation membranes and gas absorption membranes. With a gas separation membrane, gas with CO_2 is introduced

at the high-pressure side of the membrane. Carbon dioxide is recovered at the low-pressure side. A solid microporous membrane is used to enable gas flow and absorption in the gas absorption system. This system has a high removal rate of CO_2, on the grounds of minimization of flooding, foaming, channeling, and entrainment. The principles of both membrane systems are shown in Figure 7 [22,30].
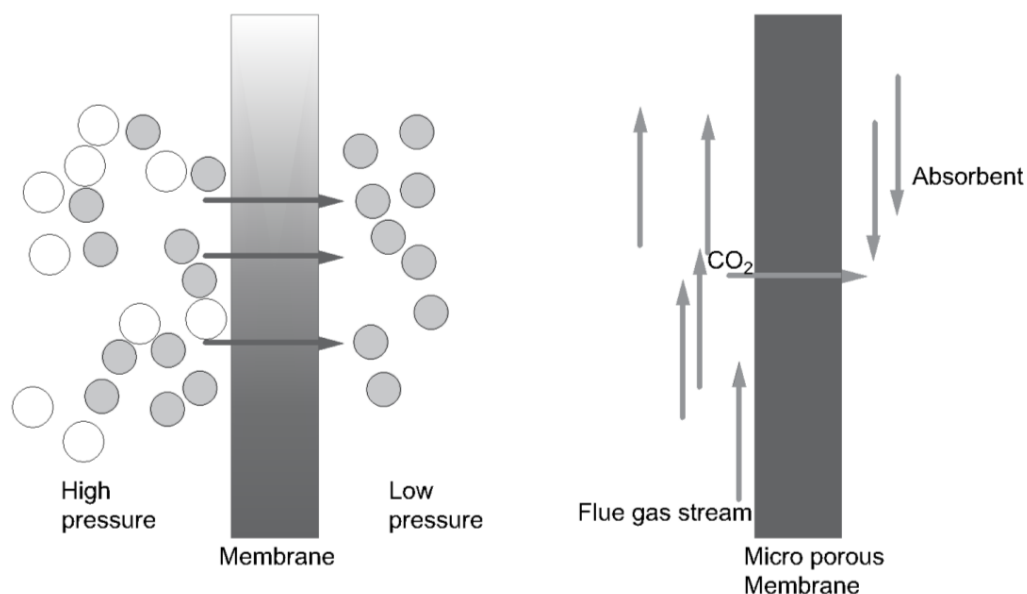

Figure 7. Principle of (**left**) a gas separation membrane, (**right**) a gas absorption membrane (based on [30]).

Membranes should characterize relevant properties for gas separation—proper permeability and selectivity. There are three types of membrane materials: polymeric membranes (organic), ceramic membranes (inorganic), and hybrid membranes [22]. A polymeric membrane has a lower cost of production than the others with a relatively high gas flux and it is mechanically stable [33]. Nevertheless, it has generally low selectivity CO_2/N_2—less than 100, and it is supposed to be 200 [32]. Ceramic membranes, especially zeolites and their derivatives, obtain high selectivities. However, the production of ceramic membranes is more difficult [22]. Hybrid membranes (modified on the surface of inorganic membranes) provide advantages of both membranes, polymeric and ceramic. They have the flexibility and low cost of production of a polymer and the high selectivity of an inorganic material [8,22]. For post-combustion capture, commercially available polymeric membranes, such as PRISM, Polaris, PolyActive, PermSelect, and Medal, are introduced in [34]. A new approach is to use metal–organic frameworks (MOFs) in the experimental stage. These offer many properties that are useful for membranes, such as large surface areas, adjustable pore sizes, and controllable pore-surface properties [32].

(d)　Chemical looping combustion (CLC) and calcium looping process (CLP)

Chemical looping technology uses two reactors, an air reactor and a fuel reactor. These reactors typically circulate fluidized beds that are coupled for carrier transport. In the air reactor, oxidation of an "oxygen carrier", usually metal particles, such as iron, manganese, or copper, occurs with the oxygen from air. As a result of the reaction, metal oxides are formed. These compounds are carried to a second reactor, where they react with the fuel. Metal oxides are reduced during combustion, producing energy and flue gas as a stream of CO_2 and H_2O. The flue gas can be condensed to receive pure CO_2 [35,36].

The calcium looping process is a type of chemical looping. The process (Figure 8) is based on a reversible reaction between calcium oxide and carbon dioxide. The reaction of bounding CaO and CO_2 is called carbonation, and takes place in the first reactor. Subsequently, the formed calcium carbonate in the carbonator is transported to the second

reactor, called a calciner, where the reversible reaction occurs and high purity CO_2 stream is produced (>95%). In the calciner, the heat needed for the reaction is generated by burning fuel in the oxygen atmosphere, and sometimes the CLC capture method is considered a kind of oxy-combustion method. The reactors (circulating fluidized bed (CFB)) are coupled to transport solid and cyclones separate solid and gaseous mass streams. Calcium looping technology has a few advantages. It uses a cheap sorbent (lime) and the flue gas is partially desulfurized. Moreover, the process uses fluidized beds, and this mature high-temperature technology can generate power [6,37].
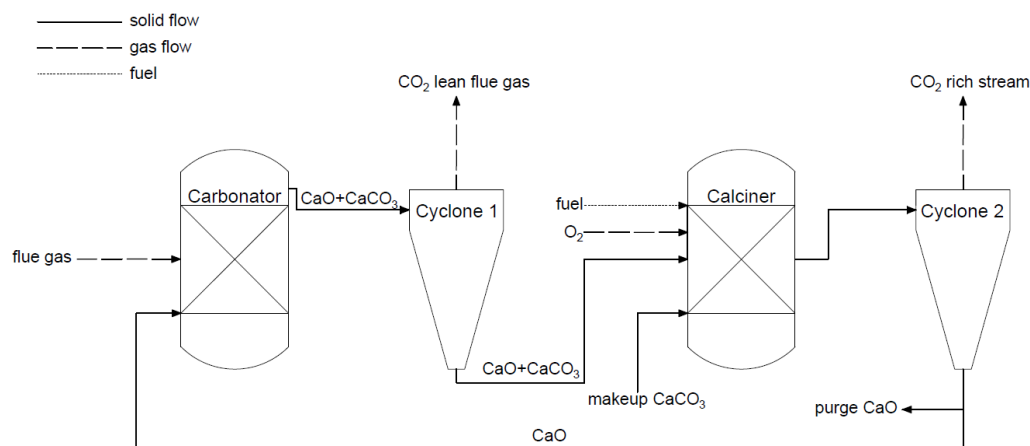

Figure 8. Scheme of the calcium looping process (adapted from [37]).

(e)   Cryogenic method

The cryogenic method of carbon capture technology uses liquefied natural gas (LNG) to provide cold energy to capture CO_2. Cryogenic CO_2 capture is used in oxyfuel combustion technology as well as in post-combustion carbon capture technology to separate CO_2 from flue gas. With cryogenic CO_2 capture, it is possible to produce high purity CO_2 of up to 99.17%. This method includes a few processes, such as compression, expansion, separation, and cooling. The cryogenic method is less preferred because of its high operational cost [38]. The cryogenic method used in post-combustion carbon capture is carried out using various methods [38,39].

(f)   Application of absorption-based post-combustion capture method

The absorption-based post-combustion capture is the most widely used method due to its efficiency and lower energy consumption. Monoethanolamine (MEA), methyl diethanol amine (MDEA) and piperazine (PZ) are the most extensively used amine solvents in large-scale industries [40]. There are many classified technologies used in adsorption and absorption, as well as in membrane separation. Chao et al. analyzed the challenges and compared the commercial use of PCC technology using solvents for absorbents, bed configurations for adsorption, and membrane processes. Among the adsorbent processes, temperature swing adsorption is very effective for both adsorptions, using solid and solvents, compared to pressure swing adsorption (PSA) and vacuum swing adsorption (VSA). TSA is more efficient than PSA and VSA, but it consumes a large amount of energy during regeneration. In the case of chemical absorption, MEA is the best and most used solvent for CO_2 capture. MEA shows a good absorption and desorption rate when mixed with other solvents as well. However, the solvent absorption process requires a high energy consumption for the regeneration of solvents, and solvent losses may occur due to evaporation and chemical degradation, leading to reduced absorption capacity [41]. Lungkadee et al. showed simulation analyses of retrofitting a post-carbon capture unit with a 300 MW power plant. The amine-based PCC technology used MEA amine for the carbon capture process, and was estimated to cost less than 55 \$/ton of CO_2 capture. The absorber

and desorber used in this process were designed to have 90% CO_2 capture capacity and 30 wt% of MEA. About 63.075 kg/s of CO_2 was captured from flue gas at a flow rate of 458 kg/s with 15% CO_2 content [42]. Another simulation analysis of natural gas combined cycle (NGCC) power plants with PZ solvent showed better performance when compared to that of MEA solvent. The use of 40 wt% PZ solvent showed significant improvements in capture efficiency, energy consumption, and capture cost compared to that of using 30 wt% MEA solvent. The lowest CO_2 capture cost was obtained at 34.65 \$/ton of CO_2 using 40 wt% PZ solvent [43]. Hadri et al. showed a comparison of 30 different amine solutions (30 wt%) used for post-combustion carbon capture. The amine solutions were analyzed using a solvent screening setup, where amine was passed at 1 bar pressure with a gas containing 15% CO_2, and CO_2 loading was measured. Compared to that of other amines, hexamethylenediamine had the highest CO_2 loading of 1.35 and triethanolamine had the lowest CO_2 loading of 0.39 [44].

(g)　　Converting CO_2 into value-added chemicals

CO_2 can be utilized to satisfy the needs of various industries as fuels and chemicals or beverages and food [45]. Technologies that allow to convert CO_2 into value-added chemicals are still being developed because of their economic and environmental benefits. In contrast to physical processes, the valence state of CO_2 changes [46]. This process can be used to produce chemical feedstock (polymers, plastics, carbonates [47]), as well as energy carriers (methane, ethane, methanol, syngas). Among the chemical conversions, it is possible to distinguish thermochemical, electrochemical (photoelectrochemical [48]), and biological processes, where enzymes are used [49]. Because of the high stability of CO_2, there is a thermodynamic barrier in the CO_2 conversion process [50]. A crucial component in most processes connected with converting CO_2 into value-added chemicals is hydrogen. It should be produced using renewable energy sources to maintain an environmentally friendly effect.

4. Oxy-Combustion CO_2 Capture

The exhaust gas from combustion in an oxygen-enriched atmosphere (oxy-combustion) consists mainly of carbon dioxide and water vapor (nitrogen content is minimized). From the condensation of water vapor, CO_2 separation is possible. The condensation temperature is higher than ambient conditions, except for very low partial pressures during the condensation process. The oxygen for combustion is produced using the air separation process, which gives an oxygen purity of about 95%. The general scheme for the process using the oxy-combustion method is presented in Figure 9.

Application of oxy-combustion technology mainly concerns solid fuel-fired boilers, including pulverized coal boilers (PCs) or circulating fluidized bed boilers (CFBs), but more and more consideration is being given to the possibility of using oxy-combustion in energy systems with gas turbines. In oxy-combustion technologies, low-temperature and high-temperature boilers can be distinguished. The combustion process in low-temperature boilers usually takes place in oxygen mixed with recirculated exhaust gases. The flame temperatures are similar to those of air-powered units. In the case of high-temperature boilers, the temperature can exceed 2400 °C.

The strengths of oxy-combustion are nitrogen oxide (NO_x) reduction, boiler dimension reductions, a simplified CO_2 capture method compared to other technologies, the possibility of applying in existing technologies, and less mass flow rate of exhaust gases (about 75% less compared to combustion in air). The weaknesses of oxy-combustion are the high material requirements because of the high temperatures, an efficiency decrease (oxygen production process is energy-consuming), and a high capital cost.
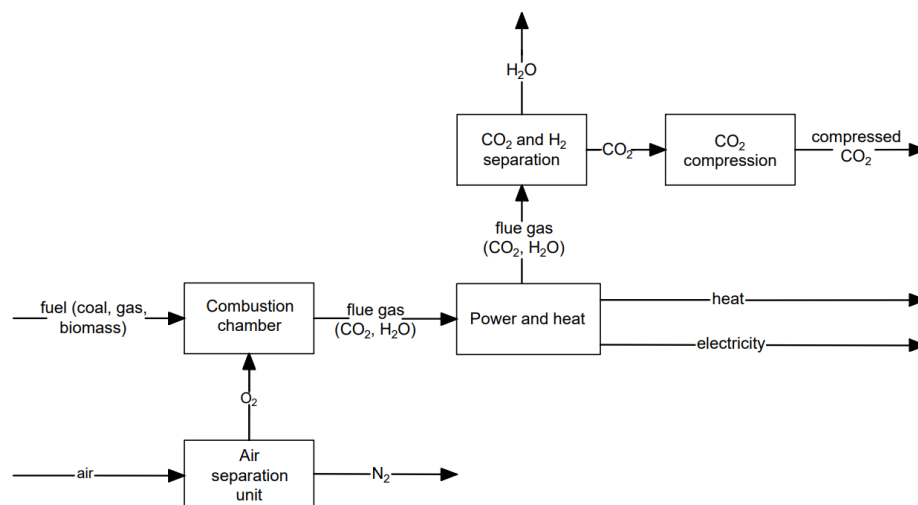
OXY-COMBUSTION

Figure 9. Block diagram of electricity generation heat production with the oxy-combustion CO_2 capture method.

Oxy-combustion methods are mainly used at the laboratory scale, and for pilot installations, among which are the Callide Power Station [51] and Compostilla Thermal Power [52]. A 30 MW_e experimental unit in the Callide Power Station started operation in 2012. The nominal flow of 98% pure oxygen, which was supplied to the boiler, was 19,200 m_n^3/h. Various types of fuel have been the object of investigation (Callide coal, Minerva coal, etc.). Daily production of CO_2 based on cryogenic capture technology was 75 t. The unit was closed after a successful demonstration in 2016. A pilot power plant unit with CO_2 capture, based on oxy-combustion technology, was developed as part of the OXYCFB 300 project at Compostilla Thermal Power. The CO_2 capture installation was equipped with a circulating fluidized bed boiler with thermal power of 30 MW_{th}, and a pulverized coal-fired boiler with thermal power of 20 MW_{th}. The flue gas stream was 800 m^3/h. The daily capacity of the separation process, which was carried out using the cryogenic method, was 3–5 tons of CO_2.

The development of oxy-combustion technology is not only connected with solid-fired fuel units. It also concerns systems equipped with gas turbines, where combustion in an oxygen-enriched atmosphere takes place. Within the framework of the project Negative CO_2 Emission Gas Power Plant [53,54], the concept of a negative CO_2 emission gas power plant based on oxy-combustion combined with CO_2 capture from flue gas was developed. Application of CO_2 neutral fuel, such as sewage sludge in combination with oxy-combustion and CO_2 capture, will allow negative emission levels [55]. The CO_2 capture process will be possible, among other things, through the use of a prototype spray-ejector condenser (SEC). The main task of the SEC will be to condense the water vapor from the exhaust gases.

Among other CO_2 capture technologies, oxy-combustion carbon capture does not require many process modifications. Oxygen is used for the combustion process instead of air to eliminate the nitrogen content in flue gas, which leaves flue gas with carbon dioxide and water vapor as the major contents. This flue gas does not require high energy consumption for CO_2 separation. The boiler's temperature is controlled by again sending part of the flue gas (about 70%) to the boiler. The air separation unit (ASU), which separates oxygen from air, is the most energy-consuming part of the oxyfuel combustion process. An industrial-scale cryogenic ASU consumes up to 200–225 kWh/t on an industrial scale [56]. To improve efficiency and CO_2 capture, oxyfuel combustion carbon capture is combined with moderate and intense low-oxygen dilution (MILD). MILD oxy-combustion carbon capture (MOFC) holds many benefits, such as improving the efficiency of the plant, improv-

ing the purity of the CO_2, and reducing energy consumption [12]. The oxy-fuel combustion CCS technology used in the Allam cycle shows a higher efficiency of 55–59%, which is higher when compared to that of a combined cycle power plant with a carbon capture unit. In the Allam cycle, the heat generated from the ASU is sent to the regenerator to heat up the CO_2 to 400 °C, which is then reused in the combustion chamber, improving the cycle efficiency [57].

5. Indicators for CO_2 Emission Level Assessment

In order assess the CO_2 emission level and CO_2 capture, many variants of indicators, depending on what they want to present, can be used and have been presented [8,58–63]. The most popular indicators that allow evaluating CO_2 technologies are as follows:

- Specific emission of carbon dioxide, eCO_2 (kg$_{CO2}$/kWh):

$$eCO_2 = \frac{\dot{m}_{CO_2}}{N_{net}} \cdot 3600 \tag{1}$$

where $\dot{m}_{CO_2}$—mass flow rate of the emitted CO_2, kg/s; N_{net}—net power of electricity generation, kW.

- Relative emissivity of carbon dioxide, $e_r CO_2$ (kg$_{CO2}$/kWh):

$$e_r CO_2 = \eta_{net} \cdot eCO_2 \tag{2}$$

$$e_r CO_2 = \frac{N_{net}}{\dot{Q}_{CC}} \frac{\dot{m}_{CO2}}{N_{net}} \cdot 3600 \tag{3}$$

$$e_r CO_2 = \frac{\dot{m}_{CO2}}{\dot{Q}} \cdot 3600 \tag{4}$$

where $e_r CO_2$ in (Equations (2)–(4)) is defined as the amount of emitted CO_2 divided by heat input from the fuel (kg$_{CO2}$/kWh); or as the net efficiency of electricity production of the cycle, η_{net} ($\eta_{net}=N_{net}/\dot{Q}$), multiplied by the specific CO_2 emission, eCO_2 ($eCO_2 = \dot{m}_{CO2}/N_{net}$). $\dot{Q}$ is the chemical energy rate, kW.

- CO_2 capture ratio *CCR* (unitless):

$$CCR = \frac{\dot{m}_{CO_2,capt}}{\dot{m}_{CO_2,gen}} \tag{5}$$

where *CCR* (unitless) is defined as the mass flow rate of the captured CO_2, $\dot{m}_{CO_2,capt}$ (t/h) divided by the generated mass flow rate of CO_2, $\dot{m}_{CO_2,gen}$ (t/h).

- CO_2 emission index, χ (kg$_{CO2}$/kJ):

$$\chi = \frac{m_{CO_2,gen}}{Q} \tag{6}$$

Equation (6) defines new factor, χ (kg$_{CO2}$/kJ), which is the amount of CO_2 mass generated (kg) to the heat input in the fuel (kJ). $m_{CO_2,gen}$ is the mass of generated CO_2, kg; Q is the heat input by the fuel, kJ.

- *CO_2 captured* (kg$_{CO2}$/kWh):

$$CO_2\ captured = \frac{\chi}{\eta_{p,CCS}} \eta_{cap} \tag{7}$$

$$CO_2\ captured = \frac{m_{CO_2,gen}}{Q} \frac{\eta_{cap}}{\eta_{p,CCS}} \tag{8}$$

The term CO_2 *captured* (kg_{CO2}/kWh), defined by Equations (7) and (8), refers to the amount of CO_2 *captured* (kg_{CO2}/kJ) per unit of main product of the plant (e.g., power in power plant, kWh); $\eta_{p,CCS}$ is the efficiency of the plant with capture; η_{cap} is the efficiency of the CO_2 capture.

- CO_2 *emitted* (kg_{CO2}/kWh):

$$CO_2 \text{ emitted} = \frac{\chi}{\eta_{p,CCS}} \left(1 - \eta_{cap}\right) \tag{9}$$

The term CO_2 *emitted* (kg_{CO2}/kWh) is specified as the amount of CO_2 *emitted* per main product of the plant (e.g., power in the power plant, kWh).

- CO_2 *avoided* (kg_{CO2}/kWh):

$$CO_2 \text{ avoided} = \frac{eCO_{2ref} - eCO_{2p,CCS}}{eCO_{2ref}} \tag{10}$$

The indicator CO_2 *avoided* (-) evaluates the direct CO_2 emission reduction from the plant, taking into account the emissions related to the capture processes, e.g., steam generation, and the emissions of the flue gas. eCO_{2ref} is the specific emission from the reference plant (kg $_{CO2}$/kWh), and $eCO_{2p,CCS}$ is the specific emission from the plant with capture (kg_{CO2}/kWh).

The term CO_2 *avoided* can also be characterized by the following form:

$$CO_2 \text{ avoided} = \frac{\chi}{\eta_{ref}} - \frac{\chi}{\eta_{p,CCS}} \left(1 - \eta_{cap}\right) \tag{11}$$

This parameter is specified as the net reduction of CO_2 emission per unit of net power output (kg_{CO2}/kWh) comparing with a reference power plant without CO_2 capture and compared to that of a similar power plant with CO_2 capture. η_{ref} is the efficiency of the reference plant without capture, $\eta_{p,CCS}$ is the efficiency of the reference plant with CO_2 capture installation, and η_{cap} is the efficiency of the CO_2 capture process.

- Specific primary energy consumption for CO_2 *avoided* SPECCA (kWh/kgCO_2):

$$SPECCA = \frac{HR_{p,CCS} - HR_{ref}}{eCO_{2ref} - eCO_{2p,CCS}} = \frac{\frac{1}{\eta_{p,CCS}} - \frac{1}{\eta_{ref}}}{eCO_{2ref} - eCO_{2p,CCS}} \tag{12}$$

The indicator from Equation (12) defines the amount of energy used to avoid 1 kg of emitted CO_2 (kWh/kg$_{CO2}$). $HR_{p,CCS}$ and HR_{ref} (kJ/kWh) are the heat rate of the plant with and without CO_2 capture, respectively.

- Specific primary energy consumption cost for CO_2 *avoided* (€/kgCO_2):

$$SPECCA_{cost} = \frac{HR_{p,CCS} - HR_{ref}}{eCO_{2ref} - eCO_{2p,CCS}} EC = \frac{\frac{1}{\eta_{p,CCS}} - \frac{1}{\eta_{ref}}}{eCO_{2ref} - eCO_{2p,CCS}} EC \tag{13}$$

Equation (13) is defined as the product of the *SPECCA* index multiplied by the primary energy cost, (€/kgCO_2), where HR_{ref} and $HR_{p,CCS}$ are the heat rates of the plant without and with CCS installation, respectively (kJ/kWh); eCO_{2ref} and $eCO_{2p,CCS}$ are the CO_2 emission rates in the plant without and with CCS installation, respectively (kg_{CO2}/kWh); EC is the primary energy cost (€/kWh).

- Levelized costs of electricity (USD/MWh):

$$LCOE = \frac{\sum (Capital_t + O\&M_t + Fuel_t + Carbon_t + D_t) \cdot (1+r)^{-t}}{\sum MWh(1+r)^{-t}} \tag{14}$$

The levelized costs of electricity (*LCOE*), according to IEA, indicates the economic costs of generic technology. It allows comparing technology over operating lifetimes at plant-level unit costs, at different baseloads. Equation (11) calculates the average lifetime levelized costs based on the costs of construction, operation and maintenance, fuel, carbon emissions, and decommissioning and dismantling, where $Capital_t$ is the total capital construction costs in year (t), $O\&M_t$ is the operation and maintenance costs in year t, $Fuel_t$ is the fuel costs in year t, $Carbon_t$ is thecarbon costs in year t, D_t is the decommissioning and waste management costs in year t, MWh is the amount of the electricity produced annually (MWh), $(1+r)^{-t}$ is the real discount rate corresponding to the cost of capital, and the subscript t means the year, when is a sale of production or takes place the cost disbursement.

6. Applications of CO_2 Capture Technologies on a Large-Industrial Scale

Carbon capture, utilization, and storage (CCUS) will have a key role in efforts that will lead the world to a net-zero CO_2 emission path. CCUS technologies will have to play an important role, alongside electrification, hydrogen technologies, and sustainable energy based on biofuels. It is the only group of technologies that directly contributes to a reduction in CO_2 in crucial sectors and CO_2 removal that cannot be avoided. Stronger climate and investment incentives are driving forces for CCUS technology. Since 2017, the rapid growth of newly announced integrated CCUS units has been observed (Figure 10). These are mainly located in the United States and Europe and in Australia, China, Korea, the Middle East, and New Zealand [6]. If all projects are launched, the global CO_2 capture potential will more than triple to about 130–150 Mt_{CO2}/year of captured CO_2 (currently it is about 40 Mt_{CO2}/year), as shown in Figure 10. In 2021, 97 CCUS facilities were in early stages and announced, 66 were in advanced development and 5 were under construction [64].

Figure 10. Commercial CCS facilities operating and under development [6,64–67].

Currently, there are 27 CCUS facilities in the world with a capture capacity of up to 40 Mt CO_2 per year [6,64]. The total capacity of all CCUS installations developed since 1972, and the capacity of new installations built every year, are presented in Figure 11.
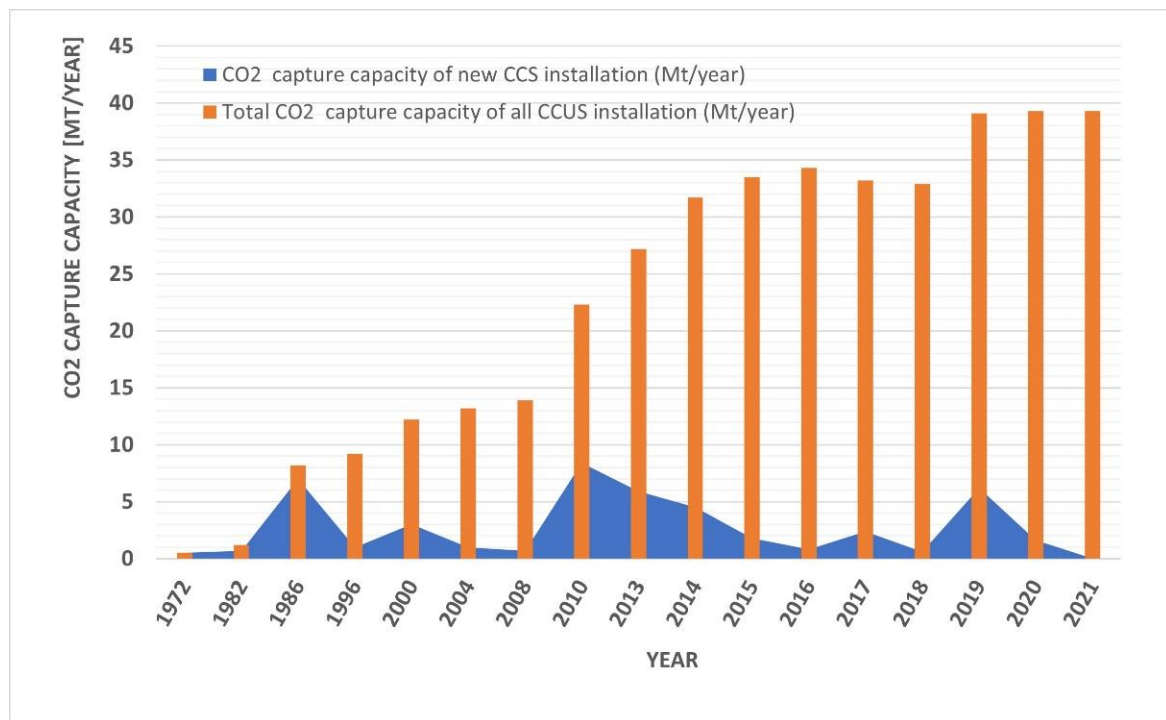
Figure 11. Total capacity and new large-scale CCUS installations capacities in 1972–2021 [6,64,66–70].

Some of these have been operating since the 1970s and 1980s when natural gas processing plants in Texas began capturing CO_2 and delivering it to local oil producers. The first large-scale CO_2 capture and injection project, with dedicated CO_2 storage and monitoring systems, was put into operation at the undersea Sleipner gas field in Norway [66,67] in 1965. Commercial large-scale operations in 2021 in terms of CO_2 capture facilities are presented in Table 2. Commercial large-scale is defined as a scale covering the capture of at least 0.8 Mt/year CO_2 for coal-fired power stations and 0.4 Mt/year CO_2 for other industrial facilities (including natural gas-fired power generation). Data presented in Figure 11 include facilities that are out of service: that in Salah (Algeria) closed in 2011, Los Cabin Gas Plant (USA) closed in 2018, Kemper County IGCC Project (Canada) closed in 2017, and Petra Nova (USA) closed in 2020.

Table 2. Commercial large-scale CCUS facilities in operation between 1972 and 2021 ([1] facility out of operation in 2021) [6,66–70].

Country	Project	Operation Date	Source of CO_2	CO_2 Capture Capacity (Mt/Year)	Primary Storage Type
United States (USA)	Terrell natural gas plants (Val Verde Gas Plants)	1972	Natural gas processing	0.4–0.5	United States (USA)
USA	Enid Fertilizer	1982	Fertilizer production	0.7	EOR
USA	Shute Creek gas processing facility	1986	Natural gas processing	7.0	EOR
Norway	Sleipner CO_2 storage project	1996	Natural gas processing	1.0	Deep saline formation
USA/Canada	Great Plains Synfuels (Weyburn/Midale)	2000	Synthetic natural gas	3.0	EOR
Algeria [1]	In Salah CO_2 Injection	2004	Natural gas processing	1.0	Deep saline formation
Norway	Snohvit CO_2 storage project	2008	Natural gas processing	0.7	Deep saline formation
USA	Century plant	2010	Natural gas processing	8.4	EOR
USA	Air Products steam methane reformer	2013	Hydrogen production	1.0	EOR
USA [1]	Lost Cabin Gas Plant	2013	Natural gas processing	0.9	EOR
USA	Coffeyville Gasification	2013	Fertilizer production	1.0	EOR
Brazil	Petrobras Santos Basin pre-salt oilfield CCS	2013	Natural gas processing	3.0	EOR
Canada	Boundary Dam CCS	2014	Power generation (coal)	1.0	EOR
Canada [1]	Kemper County IGCC Project	2014	Natural gas processing	3.5	EOR
Saudi Arabia	Uthmaniyah CO_2 EOR demonstration	2015	Natural gas processing	0.8	EOR
Canada	Quest	2015	Hydrogen production	1.0	Deep saline formation
United Arab Emirates	Abu Dhabi CCS	2016	Iron and steel production	0.8	EOR
USA [1]	Petra Nova	2017	Power generation (coal)	1.4	EOR
USA	Illinois industrial	2017	Ethanol production	1.0	Deep saline formation
China	Jilin oilfield CO_2 EOR	2018	Natural gas processing	0.6	EOR
Australia	Gorgon Carbon Dioxide Injection	2019	Natural gas processing	3.4–4.0	Deep saline formation
Qatar	Qatar LNG CCS	2019	Natural gas processing	2.2	Dedicated geological storage
Canada	Alberta Carbon Trunk Line (ACTL) with North West Redwater Partnerships	2020	Hydrogen production	1.3–1.6	EOR

7. Conclusions

Growing energy demands are still observed, and a large part of energy is produced with the use of fossil fuels. The gaseous pollutant emissions from fossil fuel power plants include carbon dioxide, which is the major cause for the emission causing global warming and climate change. The Paris Agreement aims for sustainable energy with zero-emission by capturing CO_2 released into the atmosphere from anthropogenic activities. The International Energy Agency report in 2020 recommends that the global energy transition can be carried using renewable energy, bioenergy, green hydrogen, and CCUS to reduce emissions in large-scale industries. This paper presents developed methods and technologies for carbon dioxide capture. Crucial issues connected with the progress of contemporary global technologies based on pre-combustion, post-combustion, and oxy-combustion methods have been discussed.

Pre-combustion capture is connected with removing carbon compounds before introducing fuel into the combustion chamber. This method is mainly used in integrated gasification in combined cycle (IGCC) processes, and can achieve a high efficiency of CO_2 removal—more than 90% of CO_2 capture.

Post-combustion methods are the only solutions for existing coal-fired units. CO_2 capture from flue gases is based on chemical absorption, physical separation, membrane separation, cryogenic methods, and combustion in a chemical loop. MEA solvents are the most mature technology; however, research showed less energy consumption for ammonia and PZ-AMP solvent, which must be investigated more.

In oxy-combustion technologies, fuel is burnt in an oxygen-enriched atmosphere. Therefore, exhaust gases consist mainly of CO_2 and steam, which are then condensed and carbon dioxide is separated. Currently, oxy-combustion methods are used at the laboratory scale and in pilot installations. This technology has many advantages and opportunities for development (reduction of NO_x, less amount of exhaust gases, does not require many process modifications, possibility to use CO_2 neutral fuel, and Allam cycle).

Different ways of reducing CO_2, when the fossil fuels are used as the energy input, are presented in this paper:

- In the case of fossil-fueled power plants, there is a need to use carbon capture utilization and storage methods to reduce CO_2 emissions, and, at the end, to minimize the impact of greenhouse gases on the environment.
- Currently, there are 27 CCUS commercial facilities with which the global CO_2 capture potential is about 40 $MtCO_2$/year, but this can increase by three times after launching announced CCUS units.
- Based on the prepared review, it can be concluded that most of the operating, large-scale, commercial CCUS facilities are connected with natural gas processing and use CO_2 to enhance oil recovery.

Reduction of CO_2 emissions in energy technologies, especially in high-power fossil-fueled technologies, requires constant development in order to achieve relevant capacities. The indicators for CO_2 emission level assessment, as specific emissions of CO_2, CCR, *CO_2 avoided*, or *SPECCA*, are helpful in the evaluation process of different developed technologies for CO_2 capture and were described in detail in this paper.

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