



State of the Art in Separation Processes for Alternative Working Fluids in Clean and Efficient Power Generation

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Abstract: Gas turbines must now comply with much stricter emission control regulations. In fact, to combat the greenhouse effect, regulatory authorities have drastically reduced allowable emission levels. For example, in less than 12 years, the United States' Clean Air Act issued the New Source Performance Standards (NSPS), which tightened the NOx emission margin of natural gas combustion (from 75 ppm to 10 ppm). On the other hand, despite those efforts, the high demand for energy produced by fossil-fueled gas turbines in power plants has resulted in dramatic increases in anthropogenic CO₂ and NO_x emitted by gas combustors. Most systems responsible for these undesirable emissions are directly linked to power generation, with gas turbines playing a pivotal role. Yet, gas turbines are still widely used in power plants and will continue to meet the growing demand. Therefore, sequestration and separation techniques such as Carbon Capture and Storage (CCS) and Air Separation Units (ASU) are essential to reduce CO_2 and NO_x emissions while allowing large amounts of power to be generated from these systems. This paper provides an in-depth examination of the current state of the art in alternative working fluids utilized in the power generation industry (i.e., gas turbines, combustion). In addition, this paper highlights the recent contribution of integrating separation techniques, such as air separation, steam methane reforming, and watergas shifting, to the power generation industry to facilitate a continuous and adequate supply of alternative working fluids.

Keywords: air separation; steam methane reforming; water gas shift; alternative fluids; gas turbine

1. Introduction

This paper examines the state of art in integrating CCS and separation units to a variety of gas turbine cycle configurations, highlighting their role in producing a continuous and sustainable supply of complex blends of working fluids, thus reducing carbon and NO_x emissions and increasing the power generation cycle efficiency.

Although the progress of separation processes has been reported previously, this paper aims to correlate this progress with the potential of producing a continuous and sustainable supply of alternative working fluids to gas turbine and combustion chambers,



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Copyright: © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). hence delivering unique state-of-the-art evaluation. The method of evaluating the involved literature in this paper is based on providing a pros–cons evaluation of each separation process and efficiency comparison tables for each aspect in reference to its potential to produce a continuous and sustainable supply of alternative working fluids.

The literature [1–5] is enriched with research on the use of alternative working fluids, which have significant potential for improving gas turbine efficiency and power outputs. These increases are necessary to ensure the integration of other high-energy-consuming processes into the global carbon footprint reduction effort. Helium in the nuclear industry, ammonia–water in organic Rankine cycles, and humidified injection techniques are all examples of alternative working fluids [5–10].

Alternative working fluids are based on replacing air with an oxygen-based mixture to increase the cycle efficiency and reduce the greenhouse effect. By replacing air with oxygen-based blends, the N₂ content is eliminated from the combustion process; thus, this technique eliminates NO_x emissions while a cost-effective and straightforward condensation process captures carbon emissions [11–16]. The captured carbon is typically circulated back to serve as a diluting agent to avoid turbine overheating associated with oxyfuel combustion [17–20]. As far as efficiency is concerned, the cycle efficiency can be increased by increasing the overall heat capacities (i.e., specific heat at constant pressure (C_p)) of the working fluid in the expansion stage (i.e., turbine). This is achieved by injecting components with high heat capacities (i.e., inert gases) into the working fluid, thus increasing the work output as described in Equation (1). Another approach to improving the work output is to increase the mass flow rate (m) in the expansion stage by injecting steam into the working fluid mass flow rate, isobaric mass heat capacity, turbine total inlet temperature, and turbine total outlet temperature, respectively.

$$W_t = \dot{m} C_p (T_{03} - T_{04}) \tag{1}$$

As NO_x emissions are eliminated from the combustion process, alternative working fluid-based gas turbines utilize diffusive combustors due to their high combustion stabilities compared to premixed combustors.

Because of their non-reactive nature combined with their thermodynamic properties, inert gases present a unique opportunity among the potential gases that can be used to increase power and efficiency. Because of its stable radioactive properties, helium is known to be the best choice in high-temperature gas-cooled nuclear reactors (HTGR) [21,22]. Efficiency losses, on the other hand, are the cost of making necessary changes to the compressor and turbine blade geometry. This results in forming an end-wall boundary layer and secondary flow, impacting the final efficiency [23]. In the literature, attempts have been made to compromise between mechanical requirements and high efficiency by introducing inert gases into the working fluid, such as neon (Ne) and helium (He). For example, by optimizing the compositions of both components, an optimal expansion ratio can be achieved.

Nonetheless, such a composition has no discernible impact on cycle efficiency. As a result, additional gases are required to improve overall efficiency, lowering maintenance and operating costs. Furthermore, to accommodate carbon sequestration at the end of the line, the gas blend should include CO_2 .

Oxyfuel combustion is one method for using carbon dioxide in carbon sequestration systems that is currently being developed. Oxyfuel combustion is a modern technique that substitutes high oxygen concentrations for air (i.e., with a 21 percent v. oxygen content) as the primary oxidant [24]. Compared to air–fuel combustion, oxyfuel combustion produces flue gases with approximately 75% lower mass and volume (primarily, steam, and CO₂). Compared to air–fuel combustion, CO₂ is captured and circulated back to the oxyfuel combustion-based gas turbine with lower heat losses, resulting in lower efficiency losses [25].

Using CO₂ without injecting inert gases in oxyfuel combustion systems, on the other hand, lowers the turbine inlet temperature, necessitating higher pressure ratios to maintain the same temperature. Essentially, this entails the use of larger compressors, which impact the gas turbine's compactness. Furthermore, existing gas turbine arrangements would necessitate significant changes, delaying the adoption of CO₂ injection [25,26]. As a solution, injecting argon (Ar) into the working fluid increases the overall heat capacity by introducing another inert gas into the working fluid. Injecting Ar is a more economically sustainable option than He, Ne, or other inert gases because of its abundance in the atmosphere. By injecting argon, the turbine inlet temperature can be maintained at a constant level while maintaining high specific heat ratios for improved cycle efficiency. As a result, significant changes to existing gas turbine configurations can be avoided. Furthermore, argon's high heat capacity boosts power outputs, offsetting the efficiency reduction penalty associated with CCS installations.

However, if only Ar-O₂-CO₂ blends were used as working fluids, the costs would remain significant. Therefore, advanced humidified systems could be an alternative to increase output power and efficiencies while lowering the overall costs. Complex cycles are defined as those that combine heat recovery (HR) with humidified injection techniques [27]. By recovering about 60% of heat losses, these cycles improve efficiency and reduce emissions [28]. Heat exchange and recuperation techniques, such as gas-to-gas recuperation, are examples of heat recovery methods for gas turbines with low-pressure ratios [29]. Furthermore, higher thermal efficiencies and output power are demonstrated when heat-exchanged gas turbines are compared to similar simple gas turbines. On the other hand, steam injection is a more preferable approach for higher pressure ratios [29].

As far as the evaporative cycles are concerned, they have a higher power output but lower efficiency than a similar steam injection cycle. For evaporative cycles, water can evaporate below its boiling point; thus, it does not require extensive heating (i.e., superheaters). The compressed air counter-contacts the heated water surface by placing the evaporator after the compressor. This arrangement causes molecules to collide, allowing water molecules to escape and overcome the vapor pressure [30].

Humidification can also be used for other applications, such as cooling and overcoming compressor sizing limitations, a process known as wet compression [31,32].

CARSOXY [33] is a new set of gas turbine working fluid blends created by combining all of these components, namely carbon dioxide, argon, steam, and oxygen. Previous research [33] has demonstrated that using humidification and evaporation methods can improve the performance of a gas turbine. Furthermore, when a CARSOXY mixture is used as the working fluid, heat recovery methods are used, thus increasing cycle efficiency. High techno-economic benefits can be achieved if a suitable cycle configuration is used under the specific operating condition. Furthermore, further carbon capture and storage techniques, which are considered e imperative requirements for future fossil-fueled gas turbines [34,35], have the potential to improve efficiency while maintaining a relatively low level of CO_2 emissions by using a CO_2 –Ar–steam mixture. However, increasing the amount of CO_2 in a gas turbine's working fluid can cause carbon hydrates and blockage in cooling channels in the turbine blades [36], necessitating extra caution in determining the right amount of CO_2 in the working fluid.

As discussed, evaluating the proposed novel approaches of utilizing alternative working fluids in gas turbines is based on the implementation simplicity, cycle efficiency, power outputs, economic sustainability, and acceptable combustibility criteria. Therefore, within those criteria, this paper provides an in-depth examination of the current state of the art in alternative working fluids utilized in the power generation industry (i.e., gas turbines, combustion). In addition, applying alternative working fluids in power plants implies using separation processes (i.e., air separation, carbon capturing, steam methane reforming, water gas shifting, pressure swing adsorption, distillation, etc.). Therefore, promoting the concept of alternative working fluids to the industry is determined by the feasibility level of implementing those separation processes. Thus, this paper highlights the current contributions of integrating separation techniques, such as air separation, steam–methane reforming, and water–gas shifting to the power generation industry to facilitate a continuous and adequate supply of alternative working fluids.

As far as the integration of separation processes to gas turbines is concerned, there are two major schemes to increase the cycle efficiency or/and reduce the greenhouse effects. The first scheme uses alternative fuels, such as decarbonized fuel blends (i.e., H_2 and NH_3/H_2), amongst many other examples in the literature [37,38]. The second scheme is to use alternative working fluids, which requires the implementation of separation processes to ensure a continuous supply of the components of the alternative working fluid. Therefore, this paper spots the integrated separation techniques to supply the components of the alternative working fluids (oxidized gas turbine cycles, H_2O for humidified gas turbine cycles, CO_2 -diluted gas turbine cycles, and inert gas-enhanced gas turbine cycles).

In addition, air may be taken from a gas turbine facility for various reasons, such as for use as a feed to an air separation unit, bleed cooling air for the turbine, or other pressured air requirements inside the facility. The ejected air (Figure 1) includes useful heat recovered by boiling liquid fluids at discrete temperature levels or by sensible heat transfer to other fluids. Solvent regeneration, a process characterized by a gas-liquid absorption stage followed by heat transfer to the liquid to desorb a gaseous product or pollutant, is one type of use for recovered heat. The following unit activities, which may be found in hydrocarbon gasification or hydrocarbon processing facilities, are examples of processes that potentially benefit from this heat integration, including the regeneration of a cryogenic air separation unit's liquid-based air pretreatment system. Extraction air heat recovery might benefit a liquid-based absorption phase for removing impurities from the air supply stream to an ASU. The heated air would be cooled by an absorption column against the liquid bottoms in one embodiment. The cooled air would enter the column and come into contact with the liquid absorbent, absorbing the pollutants in the air stream. The pollutants would be desorbed from the absorbent liquid, then returned to the absorbent column through the air to begin the absorbent heating process. For better efficiency removal, the absorbent system may include one or more fluids in several absorption phases or utilize specialized absorbents to remove specific pollutants in the air stream. Heating from other sources and a pressure decrease in conjunction with heating might be used to desorb contaminants from absorbents. Impurities are desorbed by heating. A gas cleaning solvent, such as that used in the syngas cleanup process, is regenerated. Absorption devices to remove acid gases from the partial oxidation process (POX) plants are widely recognized. Extraction air heat might be utilized in place of or in addition to other heat sources for solvent regeneration. A solvent-based system might be used to recover a salable by-product such as carbon dioxide from a facility's off-gas stream. Again, heat recovered from the extracted air would be used to liberate the product stream from the solvent, either whole or partially.



Figure 1. ASU of Chemical PSA [39].

As illustrated in Figure 2, some of these notions can be combined. Heat can be recovered from extracted air by contacting it indirectly with a process fluid or transferring heat from the air to a working fluid such as steam or inert gas. High-level heat from the extracted air source is transferred to a nitrogen stream that returns to the gas turbine in this case. Contact with the rich bottoms of an absorption column used to pretreat air supply to the ASU further cools the extracted air. Other absorption systems in the plant's POX or POX product workup parts might potentially perform this heat transfer phase. The high-level heat recovery phase may be omitted, and all of the extracted air heat may be utilized for absorber renewal, depending on the solvent and substance to be absorbed. Suppose the pressure was considerably different from the supplement air source. In that case, the extracted air might be mixed with supplementary compressed air entering the pretreatment process, or it may be handled independently. Carbon dioxide is collected as a by-product from the air pretreatment region in this embodiment. The carbon dioxide might be treated as a by-product for sale or utilized within the plant. Returning the carbon dioxide to the gas turbine as a diluent is one example.



Figure 2. Application of extracted air for heat recovery [39].

2. Carbon Capture and Storage (CCS)

The most prevalent carbon dioxide capturing techniques are discussed in this section. Pre-combustion, post-combustion, and oxyfuel combustion are the three main types. Based on multidisciplinary aspects, the literature has evaluated these techniques (technology maturity state, economic sustainability, advantages, and disadvantages).

2.1. The Main Categories of the CCS

CO₂ is captured from flue gases in post-combustion CCS with no significant changes required. Post-combustion CCS can be easily integrated into existing power plants [40]; however, the electricity costs are increased by approximately 70% [41].

The Technology Centre Mongstad in Norway [42] is one of the most prominent examples of deploying post-combustion CCS commercially. According to the reference [42], the process begins by separating liquid and solid particulates from combustion flue gases in a separation unit, as shown in Figure 3. Flue gases rise from the absorber's bottom against a counter-current stream of lean solution. This allows CO_2 to be absorbed and the treated flue gases to exit the absorber at the top. Meanwhile, the CO_2 -rich solution travels from the absorber's bottom to the stripper's top. Against a counter-current water vapor stream, the CO_2 -rich solution flows downward. The water vapor stream absorbs the majority of the remaining CO_2 . Finally, CO_2 is separated from water vapor using a condensation process.



Figure 3. A simplified diagram of a reactive solvent-based post-combustion capture process [42].

Pre-combustion CCS, unlike post-combustion CCS, is difficult to integrate into existing power plants. It necessitates extensive pretreatment, particularly for coal-fired power plants [43], which reduces the system's compactness. On the other hand, the efficiency losses are significantly lower than those seen in post-combustion [44]. The pre-combustion CCS process begins with a coal gasification process, in which coal is converted into hydrogen, carbon monoxide, and carbon dioxide, as shown in Figure 4. This is followed by a water–gas shift reaction, which converts carbon monoxide to carbon dioxide and increases hydrogen production. Carbon dioxide can be captured at this stage using absorption, adsorption, membrane separation, hydrate-based separation, and cryogenic distillation techniques. Natural gas fuel is processed in the same way. The gasification process, on the other hand, is replaced by a reforming process. The CO₂ removal efficiency of natural gas fuel pre-combustion CCS can reach up to 80% [35].



Figure 4. CCS of pre-combustion [44].

Regarding oxyfuel combustion in CCS, the air is not directly supplied to the combustion chamber, as seen in Figure 5. It is first separated into argon, oxygen, nitrogen, and other gases in an air separation unit. The oxygen is extracted and fed into a combustion chamber. Carbon dioxide is highly concentrated in oxyfuel combustion flue gases [41], thus allowing easier carbon capturing. However, the use of the ASU for an oxyfuel lignite-fired power plant, for example, has been reported to reduce plant efficiency by up to 10% [44].



Figure 5. CCS of oxyfuel combustion [44].

Table 1 summarizes the three CCS technologies' maturity states, economic status, and benefits and drawbacks [44].

2.2. Methods for Separating Carbon Dioxide

 CO_2 is separated by absorption using liquid sorbents, and stripping techniques can be used to extract the latter. Monoethanolamine (MEA) ensures a CO_2 absorption efficiency of 90% [45]. Adsorption [45] is the technique used when the sorbent is in the solid phase (such as hydrotalcite, molecular sieves, and so on). Another well-known technique is membrane separation, which allows CO_2 to pass through a porous composite polymer selectively. Because different gases have different molecule sizes, pores are carefully sized to match the size of the CO_2 molecule [46]. Table 2 [46] compares and contrasts the benefits and drawbacks of the most common CO_2 separation techniques.

Capturing Technology	Technology Status	Cost	CO ₂ Concentration	Thermal Efficiency	Pros	Cons
Post- combustion	Fully Developed	The lowest	The lowest	The highest	Easily integrated into existing power plants.	The electricity costs are increased by approximately 70%
Pre- combustion	Fully Developed	Medium	Medium	The lowest	The efficiency losses are significantly lower than those seen in post- combustion. Enables hydrogen production. The CO ₂ removal efficiency of natural gas fuel pre-combustion CCS can reach up to 80%	Difficult to integrate into existing power plants. Reduces the system's compactness.
Oxyfuel combustion	Developed air separation	The highest	The highest	Medium	Eliminates NO _x emission of the power plant.	Reduces plant efficiency by up to 10%

 Table 1. The current state of CCS technologies.

Table 2. CO₂ pros and cons.

CO ₂ Separation Method	Pros	Cons
Absorption	 The most developed technology The highest CO₂ recovery efficiency (approximately 90%) 	 Low efficiency for low CO₂ concentration, i.e., Post-combustion High heat requirements
Adsorption	 Recyclable absorbent Relatively high CO₂ recovery efficiency (approximately 85%) 	• High heat requirements
Membrane separation	• Relatively high CO ₂ recovery efficiency (approximately 80%)	Operational issues
Hydrate-bases separation	 Negligible energy penalty 	• Not fully developed technology
Cryogenic distillation	Fully developed technology	• High-energy penalty

2.3. CO₂ Storage

Structural and stratigraphic, residual, solubility, and mineral trapping are the four primary trapping mechanisms for CO_2 storage [47]. Once CO_2 is injected underground, it rises to the top of geological structures, owing to the buoyancy effect. Still, it remains below the impermeable caprock with structural and stratigraphic trapping, which is the most prominent trapping process. The injected CO_2 displaces formation fluid as it passes through the formation rock in residual trapping. Due to the capillary force, the displaced fluid disconnects and traps the remaining CO_2 inside the pores of rocks [48]. The saturation of trapped CO_2 in the residual trapping mechanism is at least 10% and can reach more than 30% of the pore volume in some reservoir rocks [49]. CO_2 dissolves in formation fluids and becomes immobile, resulting in a reduction in the amount of free CO_2 [36]. The density of the formation fluid will be slightly increased by about 1% due to the dissolved CO_2 . Such a small density differential [36] is adequate to induce convection flow, which is also beneficial to CO_2 trapping. CO_2 solubility in groundwater varies between 2% and 6%, decreasing with increasing temperature and salinity [50]. CO₂ is held in the mineral trapping mechanism by geochemical processes in the reservoir, which generally precipitate as carbonate, effectively trapping CO_2 in immobile secondary phases [50].

In a period ranging from 1 to 10,000 years, various trapping processes play distinct roles in CO_2 storage. Structural trapping is vital in the early stages of CO_2 storage, but its impact fades over time. Within decades, residual trapping and solubility trapping significantly influence and lock up a large quantity of CO_2 for thousands of years. Mineral trapping begins to have a substantial influence around a hundred years and plays a critical function in a geological timeframe.

For various reasons, storing CO₂ in hydrocarbon reservoirs is one of the most preferred CO_2 storage options. For example, oil and gas reservoirs contain a large quantity of existing equipment on the surface and underground that may be utilized for CO₂ storage with only minimal modifications. Furthermore, the caprock's seal quality and integrity are assured, having been thoroughly defined during the exploration and production process. Moreover, because of the long-term extraction of oil and gas, the magnitude of pressure perturbations and associated stress shifts in depleted oil and gas reservoirs is significantly smaller than in aquifers. Depleted gas reservoirs are more suitable for CCS than depleted oil reservoirs because of gas's higher ultimate recovery and compressibility and a bigger storage capacity per pore volume. When comparing the types of reservoirs utilized in this type of storage, condensate gas reservoirs outperform wet and dry gas reservoirs due to the small amount of residual gas, the phase behavior of the combination of condensate gas, and CO₂, and its high injectivity. Additionally, the amount of CO_2 sequestered per pore volume in depleted condensate reservoirs is relatively high: around 13 times that of a comparable aquifer. Yet, the phase transition may occur in depleted condensate reservoirs but not in dry and wet gas reservoirs; therefore, caution is advised.

3. Air Separation Unit (ASU)

The concept of oxyfuel combustion is based on utilizing pure oxygen as the primary working fluid while eliminating nitrogen to avoid NO_x emissions. As previously mentioned, pure oxygen is typically accompanied by inert gases, such as argon, for its high heat capacity, to increase the cycle efficiency. Therefore, to separate oxygen and argon from the air while excluding nitrogen from the combustion process (i.e., to avoid NO_x emission), air separation techniques are integrated into oxyfuel combustion-based power plants [51].

3.1. Types of Cryogenic Distillation

Cryogenic distillation can be performed using a single distillation column or multiple distillation columns. The multi-column process [52] is, however, the traditional method. A low-pressure column, a high-pressure column, and a side rectifier or strippers are the main components of this process. The cost of multi-column distillation remains a challenge.

10 of 23

However, several cost-effective improvements have been proposed in the literature [52] and are detailed in the following sections.

3.1.1. Thermally Linked Distillation Column, with a Side Rectifier

According to the reference [53], A ternary mixture made up of A, B, and C components (i.e., N₂, Ar, and O₂ in air, respectively) are separated, as shown in Figure 6. The most volatile component is A (i.e., N₂ in the air), followed by B and C (i.e., Ar and O₂ in air, respectively), the intermediate and least volatile components, respectively. The mixture is supplied to the main distillation column, which extracts component A (i.e., N₂ in air) from the top. From the bottom, component C (i.e., O₂ in air) is extracted. The ABC (i.e., air) mixture's main feed is located approximately in the middle of the distillation column. The side feed is connected to the bottom of the rectifier and located just beneath the main feed. B and C (i.e., Ar and O₂ in air, respectively) are the components of the side feed mixture. B (i.e., Ar in the air) is produced from the top of the side rectifier. At the same location as the side feed, component C (i.e., O₂ in the air) from the side rectifier is recycled back into the main column. When the relative volatility of the two components (B and C (i.e., Ar and O₂ in the air) from the side feed is located is low, this method is used.



Figure 6. Thermally linked distillation column, reproduced from the reference [53].

3.1.2. Thermally Linked Distillation Column, with a Side Stripper

A side stripper is a better choice if the low relative volatility is between (A and B) rather than (C and B). The side feed is linked to the top of the stripper and is located above the main feed, as shown in Figure 6. The side feed mixture comprises A and B, with B produced at the stripper's bottom, whereas A is returned to the main column.

3.1.3. Side Rectifier vs. Side Stripper

The concept of using ternary mixture distillation on air is depicted in Table 3. Because argon and oxygen (B and C) have lower relative volatility than argon and nitrogen (B and A) [54], a side rectifier is a more proper choice for air separation.

3.2. Low-Pressure (LP) Distillation Column vs. Elevated-Pressure (EP) Distillation Column

Low-pressure distillation columns are used if the by-products of nitrogen production (oxygen and argon) are not stored and discharged near atmospheric pressure. The storage of oxygen and argon, on the other hand, necessitates an additional increase in pressure. As a result, evaluated-pressure distillation columns are utilized. To put it another way, if the by-products of producing nitrogen are compressed and used as main products, an elevated-pressure distillation column is required [52]. The elevated-pressure distillation column provides better matching between the ASU and the gas turbine unit; for example, the working fluid of CARSOXY gas turbines is required to operate at relatively high pressure; thus, elevated-pressure (EP) distillation columns are required [50].

Component	Volatility	Boiling Point	in Air	Relative Volatility
А	High	Low (-195.8 °C)	Nitrogen	The relative volatility of argon and oxygen (B and C) is lower than that of argon and nitrogen (B and A).
В	Intermediate	Intermediate (-185.8 °C)	Argon	
С	Low	High (-183 °C)	Oxygen	

Table 3. Air components relative volatility [54].

3.3. Single Distillation Column vs. Double Distillation Columns

In comparison to conventional double distillation columns, self-heat recuperation of a single distillation column saves 36% of energy consumption, according to the reference [46]. Liquefaction and air compression are the significant losses in a double-column air separation process. Double distillation columns, on the other hand, are still the most popular. Table 4 provides a critical assessment of the air separation methods.

Table 4. Pros and cons of the air separation methods (distillation).

Air Separation Method.	Pros	Cons
Cryogenic distillation	Can be performed using a single distillation column or multiple distillation columns, depending on the specifications and requirements of the integrated power plant	The cost of multi-column distillation remains a challenge
Thermally linked distillation column, with a side rectifier	Used for alternative working fluids with high contents of oxygen and argon.	Excessive production of nitrogen for nitrogen-independent working fluid.
Thermally linked distillation column, with a side stripper	_	Because argon and oxygen (B and C) have a lower relative volatility than argon and nitrogen (B and A) [54], a side rectifier is a more proper choice for air separation.
Low-pressure (LP) distillation	Less energy consumption compared to the EP distillation	Not suitable for gas turbines operating at high compression ratios
The elevated-pressure distillation column	Provides better matching between the ASU and the gas turbine unit compared to Low pressure (LP) distillation	High energy consumption
Single distillation column	In comparison to conventional double distillation columns, self-heat recuperation of a single distillation column saves 36% of energy consumption	Less popular, compared to the double distillation columns
Double distillation columns	More popular compared to the single distillation column	Liquefaction and air compression cause significant losses

3.3.1. Conventional Argon Production

Air is fed to the ASU through a heat exchanger at about 5.5–6.5 bar, as shown in Figure 7, to be cooled by its products. The first stage is a high-pressure (HP) stage, and the second stage is a low-pressure (LP) stage in a two-stage distillation column that separates oxygen and nitrogen. At the HP stage, the air feed is separated into N_2 and oxygen-enriched liquid (liquid oxygen (LOX)) at the HP stage. The latter is passed through the LP stage, where it is separated into N_2 and O_2 . There is a temperature difference between the two stages due to the pressure difference. This allows heat exchange between the two stages (between LP and HP), in which the N_2 vapor-boiling stream at the top of the HP stage is then condensed by the colder liquid O_2 at the bottom of the LP stage. From the top of the HP column, N_2 is produced. The highest argon concentration is found in the lower section of the LP column due to its intermediate boiling temperature. A vapor stream is drawn to feed the rectifier from this location. The vapor argon-boiling stream at the top of the rectifier is condensed by the colder LOX liquid, similar to the main distillation column. LOX that has been vaporized is returned to the LP column.



Figure 7. Thermally linked distillation column, reproduced from the reference [53].

3.3.2. EP Argon Production

As shown in Figure 7, oxygen in the liquid phase is drawn from the bottom of the LP to provide total condensing duty, while argon is drawn from the top of the rectifier column to improve argon recovery due to the elevated-pressure process's tight volatility. The vaporized oxygen serves a second purpose after completing the total condensing duty. As the EP process reaches higher temperatures, it is used as a coolant agent in the system. Because of the heat pump effect between the argon rectifier and the oxygen at the bottom of the LP column, the pressure at the top of the rectifier remains lower than the pressure at the bottom of the LP column, requiring no compressor.

3.4. Membrane Air Separation

One of the most important ways to generate oxygen from air separation is using new ceramic membranes with mixed electronic ionic conducting (MIEC) properties. Figure 8 depicts a schematic of an oxyfuel combustion power plant using an MIEC membrane. As shown in Figure 8, an air compressor may pressurize air to a pressure of 10 bar. Both the membrane and the air must be subjected to temperatures exceeding 800 °C, as well as an oxygen partial pressure ratio across the membrane, to accomplish air separation. A combination of air compression, flue gas recycling, and boiler heat exchange can provide the heat necessary to reach this high temperature. Surprisingly, the front end of an oxyfuel combustion plant can accommodate the ceramic membrane module. Because of the continuous oxygen partial pressure drop across the membrane, which is one of the technological aspects of such integration, using recovered CO₂ from the combustion process might result in a strong oxygen driving force. Oxygen penetrates selectively through a membrane into the recycled CO₂ stream from the compressed air side, enriching it to around 20% (v/v) [55].



After that, the oxygen-enriched CO_2 stream may be burned directly in the boiler, resulting in superheated steam that can be used to power steam turbines and generate electricity.

Figure 8. Diagram of an oxyfuel power plant using an MIEC membrane module [55].

As shown in Figure 9, there are two distinct approaches for integrating an MIEC membrane into the oxyfuel combustion process. The notion of enabling recirculated flue gas to pass over the membrane surface is used in Figure 9a, allowing oxygen to be absorbed directly. This so-called four-end approach [56–58] is intended for Integrated Gasification Combined Cycle (IGCC) processes; however, CO₂-resistant membrane materials such as Ba0.5Sr0.5CO0.8Fe0.2O3I (BSCF) do not exist. As a result, BSCF membranes can only be used when the membrane material is not in direct contact with the flue gas. On the other hand, the three-end idea [59,60] depicted in Figure 9b appears to be an appealing alternative, as it avoids direct contact between the membrane and the flue gas. A vacuum pump is used to remove oxygen from the membrane module in this scenario. Although the first concept achieves a higher thermal efficiency in the oxyfuel combustion process, the three-end concept is more likely to be feasible because no membrane materials exist that can withstand direct contact with the flue gas; therefore, membrane material development is critical [61–63]. Kneer et al. [56] effectively provided the requirements of coal combustion in a CO_2/O_2 environment, including associated burner design, as well as the cleaning of hot flue gas from oxycoal combustion, in their article on membrane-based air separation modules and their design for oxycoal circumstances.



Figure 9. (**A**) four-end approach and (**B**) the three-end concept for oxygen separation in oxyfuel processes [62].

It is worth noting that there are several problems with using this innovative approach for obtaining oxygen from air separation. To offer high oxygen penetration flux, the membrane separation system must first be run at temperatures exceeding 900 °C. As a result, because the heated air includes around 80% (v/v) nitrogen as waste gas, there will be significant energy penalties in this operation. Although process efficiency losses can never be entirely recovered, energy losses can be minimized by adequately implementing

heat recovery. Second, while the strong oxygen driving force of high CO_2 partial pressure is appealing to membranes, the aggressive circumstances would lead these membrane materials to break down, limiting their efficacy in generating oxygen for the oxyfuel combustion process. The high need for oxygen for the oxyfuel combustion process is likely to lead to the deployment of oxygen membrane separation technology in the near future; nevertheless, continuing research and development is critical to overcome the significant challenges. In summary, the oxyfuel power plant is an appealing choice due to a considerable cost decrease in oxygen generation when carbon capture becomes a requirement in the future. As a result, ceramic membranes are thought to be an effective high-temperature air separation technology that might result in a significant cost decrease in the generation of oxygen for this application.

It should be mentioned that an IGCC coal-fired power station combines two technologies, coal gasification and combined cycle, to produce energy in the most efficient manner possible [63,64]. Traditionally, the oxygen supply for coal gasification has been obtained by cryogenic distillation; however, it has been observed that this technique has many inefficiencies [65]. The use of solid oxygen carriers as an alternate way of producing oxygen has also been investigated, including CaSO₄ [66,67] and other oxygen carriers such as manganese [68], nickel [69], and iron [70,71] oxides. However, there are drawbacks to using this technique, including sluggish response speeds, solid handling, and a huge solid oxygen carrier inventory [70,71]. Because these membrane modules have no moving parts and are thus simple to run and maintain, introducing a revolutionary technology based on ceramic membranes for oxygen generation from air separation is thought to enhance the efficiency of IGCC power plants.

Table 5 provides a critical assessment of the membrane air separation methods.

Membrane Air Separation Method	Pros	Cons	
MIEC membrane	 Can be well-integrated with CCS units within the power plant. Because these membrane modules have no moving parts, they are simple to run and maintain. 	 High temperature and pressure are required (approximately 800 °C and 10 bar). The aggressive circum-stances would lead these membrane materials to break down, limiting their efficacy in generating ox-ygen for the oxyfuel com-bustion process. 	
Four-end approach	• Suitable for Integrated Gasification Combined Cycle (IGCC) processes.	• Can only be used when the membrane material is not in direct contact with the flue gas.	
Three-end approach	• Avoids direct contact between the membrane and the flue gas.	• A vacuum pump is required to remove oxygen from the membrane.	

Table 5. Pros and cons of the membrane air separation methods.

4. Steam Methane Reforming (SMR)

SMR is an essential step in the carbon capture and storage (CCS) process, as discussed in Section 5.1. Furthermore, it is one of the fully developed technologies for producing hydrogen. The results and conditions in real life are identical to those in theory [72]. When a light hydrocarbon fuel (such as methane) reacts with steam, it produces hydrogen as the primary product and carbon monoxide and carbon dioxide as by-products. Since WGS processes accompany SMR in most applications, some literature includes WGS as a step in SMR [72]. Two endothermic reactions occur during the reforming process. One mole of methane to one mole of steam requires 206 kJ in the first reaction. This yields 3 moles of hydrogen and 1 mole of carbon monoxide as a by-product. WGS makes use of the hydrogen produced by this reaction by converting carbon monoxide to hydrogen and carbon dioxide as a by-product [73]. The second reaction, which produces hydrogen and carbon dioxide directly instead of one mole of methane reacting with two moles of steam, does not require WGS because it directly produces hydrogen and carbon dioxide. It uses 165 kJ and emits 4 moles of hydrogen and 1 mole of CO_2 . Finally, the CCS process is used to capture all of the carbon dioxide produced during the entire process. Figure 10 depicts the whole procedure.



Figure 10. SMR process, reproduced from the reference [74].

The SMR process is characterized by harsh reaction conditions [74]. Because reactions 1 and 2 are both endothermic, the temperature rises to around 1000 K. It has even been described as a "brutal" process [73]. Both reactions take place in a catalytic environment. Table 6 [74] explains that the catalyst must be carefully selected to withstand these extreme conditions.

Table 6. SMR catalyst [75].

Category	Temperature Range (°C)
nickel-based	550-850
carbon-based	300–900
zinc-based	300–500
aluminum-based	750–800
cerium-based	300-800
zirconium-based	250–500

Because of well-developed methane infrastructures and methane's favorable hydrogento-carbon ratio, methane steam reforming is commonly regarded as a readily available technique for producing hydrogen on a large scale [76]. Fuel cell technology needs small, low-cost reformers [77,78]. Compact reformers should be operated at low temperatures (<1000 K) and pressures (3 bar). As a result, the present large-scale reformer technology, which operates at high temperatures (>1100 K) and pressures, is incompatible with smaller-scale reformers for fuel cell applications [79]. Methane steam reforming is a substantially endothermic process. The exothermic water–gas shift process is also included, which is more favorable at low temperatures (500–850 K). The benefit of executing the steam reforming process at low operating temperatures is that the applied temperature promotes the water–gas shift reaction by suppressing the CO quantity [80]. Creating an active catalyst capable of achieving conversion up to equilibrium levels is the primary disadvantage of methane steam reforming processes at low temperatures [81–83]. Several groups investigated catalyst performance in methane steam reforming at temperatures above 1000 K [84–86].

5. Water–Gas Shifting (WGS)

Water–gas shifting is a chemical reaction that converts carbon monoxide and water into carbon dioxide and hydrogen. It is one of the most essential processes in carbon capture and storage. About 40 kJ is generated when one mole of carbon monoxide is reacted in the WGS reaction. Two types of catalysts (iron-based and copper-based) are commonly used to speed up the reaction [87].

5.1. WGS Applications

Water–gas shifting is an area of interest whenever carbon monoxide is an unwanted by-product gas. It is, in fact, a very reliable method of purifying hydrogen produced by steam hydrocarbon reforming processes. This process is typically integrated with the SMR process, Reactions (2) and (3) [26], as discussed in Section 7.

Steam reforming of methane: $H_2O + CH_4 \leftrightarrow CO + 3H_2$ (2)

Water-gas shift reaction
$$H_2O + CO \leftrightarrow CO_2 + H_2$$
 (3)

Ammonia production is another industrial application that relies heavily on water– gas shifting to prevent carbon monoxide from reacting with the catalyst [88]. Ammonia production is likely to be a part of the gas turbine cycle, primarily if evaporation injection techniques are used [88].

5.2. WGS Catalyst

The two most important factors to consider when selecting catalysts for the WGS process are sulfur tolerance and operation temperature. Iron-based catalysts have a low sulfur tolerance and operate at a high temperature (approximately 680 K). Carbon-based catalysts are used at lower operating temperatures (around 480 K). On the other hand, it has a low tolerance level for sulfur [89]. The main properties of the iron-based and copper-based catalysts are shown in Table 7 [89].

Property	Composition	Stabilizer	Promoter
Iron-based catalyst	74.2% Fe ₂ O ₃ , 10% Cr_2O_3 , 0.2% MgO, and the remaining is to balance volatiles	Cr ₂ O ₃	_
Copper-based catalyst	24% ZnO, 24% Cr ₂ O ₃ , 3% Mn, Al, and Mg-oxides. The remaining is to balance volatiles	Cr ₂ O ₃	ZnO

 Table 7. WGS catalysts.

5.3. WGS Reactors

The conventional WGS process consists of two reactors. It uses an iron-based catalyst in a high-temperature reactor (HTWGS). The high-temperature reactor's products are cooled before being purified from CO in a low-temperature reactor (LTWGS) with a copperbased catalyst. This option is ideal if the reactor's inlet boundary condition is high, and the desired product (H₂ and CO₂) must be produced at a low temperature with very low CO residuals. Figure 11 [51] shows a WGS reactor with a shell-tube design. A hydrogen-selective membrane tube is inserted into a shell. The catalytic WGS reaction is carried out around the tube (inside the shell). The membrane performs the separation function by allowing only H_2 to pass through (inside the tube), while the remaining WGS products are extracted as a CO₂-rich mixture outside the tube.



Figure 11. Shell-tube WGS reactor, reproduced from the reference [80].

6. Helium as a Working Fluid

Many nations, including Russia, Europe, South Africa, Japan, and the United States, are studying direct and indirect Closed Brayton Cycle (CBC)-based high-temperature gascooled reactors (HTGRs) that employ helium as a coolant for energy conversion. Because of its outstanding transport characteristics, helium is regarded as the best coolant. According to the data, helium has been utilized as the working fluid and coolant in 9 of the 15 closed Brayton cycle nuclear reactor systems built across the world since the 1960s [90]. However, because helium is less compressible than air, more stages are necessary to compress it to the desired pressure ratio in an axial compressor. As a result, every helium compressor ever built has a large number of stages. The Oberhausen II type 50 MW reactor's high- and lowpressure compressors each contain 25 stages [90]. As a result, the helium turbo equipment is rather substantial in terms of both size and weight. This situation is unfavorable because it raises various losses, such as the formation of a multi-stage narrow fluid flow channel. As a result, there is an increase in aerodynamic loss due to leakage, separation, and boundary layer loss. As a result, the stator and rotor stages are mismatched. Aside from that, compressors with narrow rotors cause dynamic difficulties [91,92]. As a result, there is a pressing need to address this problem since smaller shafts with two spools turbo machinery enhance plant efficiency over longer shafts because they are easier to maintain, perform better dynamically, and have high rigidity [93]. An extensive study is being carried out to identify the most viable working fluid for utilization in HTGR systems with increased thermal efficiency, electrical productivity, decreased size, and lowest rotational loss to solve the problem mentioned above.

On the other hand, researchers devised a novel design approach for a heavily loaded helium compressor to minimize the number of stages in the helium compressor. This design approach is based on changing the form of the airfoil or blade when torsional velocity, axial velocity, and negative pre-spin increase. As a result, stage loading increases [15]. Numerical simulations of a heavily loaded helium compressor were carried out using the commercial program Ansys, and it was discovered that this design method can minimize the number of stages. Still, no experiment has been carried out using it. Noble gases and their mixes are not yet used as a working fluid in CBCs. Certain factors influence the choice of working fluid for the HTGR system's power conversion unit. Noble gases and their mixes are the subjects of many studies for use as a working fluid in CBC HTGR plants and axial flow turbomachinery [94,95]. Many investigations have been carried out to evaluate the heat transmission process of the HeXe mixture in triangular channels [95], cylindrical channels [96], beginning pipe sections [97], and heated channels with varied cross-sectional forms [98]. The HeXe gas mixture's thermodynamic characteristics and transport constants

were also calculated [98]. HeXe was also used in the Prometheus project. It was based on a Brayton energy conversion loop with a single reactor heat source [99].

7. Conclusions

Local and regional regulations have been established to limit NO_x and CO_2 emissions as part of this global commitment. Most systems responsible for these undesirable emissions are directly linked to power generation, with gas turbines playing a pivotal role. As a result, stationary gas turbines have been widely used in power plants and will continue to be used to meet the growing electricity demand. As a result, novel concepts are needed to reduce emissions while allowing large amounts of power to be generated from these systems. One promising technology for reducing harmful emissions while recirculating CO_2 in the combustion process is alternative working fluids. The idea behind using alternative combustion working fluids is to replace air with oxygen, carbon dioxide, steam, or inert gases, either individually or collectively.

Using carbon dioxide as a working fluid in an oxyfuel gas turbine keeps the temperature of the turbine within acceptable limits and reduces heat losses, all while removing NO_x emissions. Chemical kinetics, flammability regions, and flame compactness, on the other hand, are negatively affected. Increased oxygen levels could lessen the severity of those negative consequences. However, air separation units will be required to provide more oxygen, resulting in additional costs. As a result, it can be concluded that using CO₂- O₂ as a working fluid (without any additional components to improve overall thermodynamic properties) poses significant technical challenges and is unlikely to be implemented on a large scale.

When compared to conventional air-driven simple cycle gas turbines, the main benefit of injecting steam into gas turbines is an increase in cycle efficiency and specific power outputs. However, based on the research on O_2 -H₂O-CH₄, it can be concluded that O_2 -H₂O-CH₄ power plants do not always achieve higher efficiencies and power outputs than air-driven cycles (i.e., aero-derivative and industrial combined cycles achieve higher efficiencies and power outputs than steam-injected or (O_2 -H₂O-CH₄) gas turbines). As a result, humidification techniques should be used with caution to achieve higher efficiency than air-driven cycles. O_2 -H₂O-CH₄ combustion/power plants showed more advanced properties, as shown in Table 7. However, those properties (laminar flame speed, CO emissions, and exergy efficiency) are only compared to the literature's O_2 -CO₂-CH₄ combustion/power plants. They must be benchmarked against air-CH₄ combustion/power plants in future studies.

Due to their higher specific heat ratios and potential for increasing cycle efficiency compared to air, argon, xenon, and helium have been studied as working fluids in the context of using inert gases in combustion and power generation. In comparison to airdriven cycles, using helium as a working fluid in closed cycles has the potential to improve cycle efficiency. However, these techniques have not yet matured to the point where they can be used on a large industrial scale (i.e., helium leakage is not easily controlled due to its low molecular weight).

Due to the high specific heat ratio of xenon (1.677) and argon (1.667) compared to nitrogen (1.401), using oxygen–argon and oxygen–xenon mixtures is expected to increase cycle efficiency while eliminating NO_x . However, because argon has a higher abundance and mixing rate than xenon, it is more likely to be used in combustion in the future.

The advantages of using carbon dioxide to keep turbine temperatures within acceptable ranges, argon to increase specific heat ratio, steam to increase mass flow rate, and oxygen to eliminate NO_x emissions are incorporated into the CARSOXY working fluid concept.

As discussed, evaluating the proposed novel approaches of utilizing alternative working fluids in gas turbines is based on the implementation simplicity, cycle efficiency, power outputs, economic sustainability, and acceptable combustibility criteria. Therefore, within those criteria, this paper provides an in-depth examination of the current state of the art in alternative working fluids utilized in the power generation industry (i.e., gas turbines, combustion).

In addition, applying alternative working fluids in power plants implies using separation processes (i.e., air separation, carbon capturing, steam methane reforming, water gas shifting, pressure swing adsorption, distillation, etc.). Therefore, promoting the concept of alternative working fluids to the industry is determined by the feasibility level of implementing those separation processes. Consequently, this paper has highlighted the current contributions of integrating separation techniques, such as air separation, steam methane reforming, and water–gas shifting to the power generation industry to facilitate a continuous and adequate supply of alternative working fluids.

Finally, the decision of choosing the composition of the optimal working fluids can be made through numerical analyses that correlate the individual effect of each component to the overall composition with respect to the gas turbine cycle efficiency. As an example of this practice, the reference [48] has related the components of the alternative working fluid with respect to the cycle efficiency through a three-dimensional efficiency surface that correlates three intervals of variable molar fractions of carbon dioxide, argon, and steam. The highest cycle efficiency is visualized as the highest peak on the efficiency surface within the tested intervals of molar fractions. As a result, the optimal blend was determined by selecting the corresponding molar fractions (to the highest peak).

As highlighted previously, the idea behind using alternative combustion working fluids is to replace air with oxygen, carbon dioxide, steam, or inert gases. Therefore, ASU is utilized to produce the required oxygen and argon content. Steam and carbon dioxide are supplied to the alternative working fluid mixture by integrating humidification and CCS techniques. However, to produce the required molar fractions to achieve the optimal working fluid blends, the operating conditions of the integrated facilities (i.e., ASU, humidification, and CCS facilities) have to be calibrated through performing extensive sensitivity analyses. An example of this practice can be found in the reference [48].

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20 of 23

Abbreviations

CCS	Carbon Capture and Storage
ASU	Air separation units
HTGR	High-temperature gas-cooled nuclear reactors
HR	Heat recovery
CARSOXY	Carbon dioxide, argon, steam, and oxygen
POX	Partial oxidation process
SMR	Steam methane reformer
WGS	Water–gas shift
PSA	Pressure swing adsorption
MEA	Monoethanolamine
LP	Low pressure
EP	Elevated pressure
HP	high pressure
LOX	Liquid oxygen
MEIC	Mixed electronic ionic conducting
IGCC	Integrated gasification combined cycle
CBC	Closed Brayton cycle
NSPS	New source performance standards
BSCF	Ba0.5Sr0.5CO0.8Fe0.2O3I
HTWGS	High-temperature water-gas shift
LTWGS	Low-temperature water–gas shift

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