OVERVIEW OF PROVEN AND EMERGING CO_2 PURIFICATION PROCESSES FOR CO_2 CAPTURE, UTILIZATION, AND STORAGE

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Ray McKaskle, P.E.
Trimeric Corporation
PO Box 826 / 100 S. Main St.
Buda, Texas USA 78601
ray.mckaskle@trimeric.com

Zeke Pickering
Trimeric Corporation
PO Box 826 / 100 S. Main St.
Buda, Texas USA 78601
zeke.pickering@trimeric.com

Kevin Fisher, P.E.
Trimeric Corporation
PO Box 826 / 100 S. Main St.
Buda, Texas USA 78601
kevin.fisher@trimeric.com

Brian Visioli
Trimeric Corporation
PO Box 826 / 100 S. Main St.
Buda, Texas USA 78601
brian.visioli@trimeric.com

ABSTRACT

More than 3.5 billion standard cubic feet of carbon dioxide (CO_2) are transported through pipelines in the United States every day [1]. In addition, smaller quantities of CO_2 are captured and used locally in applications such as food, beverage, dry ice, chemical feedstocks, and even for developing technologies such as sustainable aviation fuel and concrete. Depending on the application, specifications for product CO_2 may include limitations on water, oxygen, nitrogen oxides, sulfur compounds, and other contaminants. Established and evolving specifications often present challenges for project developers and engineers. These specifications have a significant impact on project feasibility and economics, the selection of capture technology, and the equipment, operation, and cost of CO_2 purification.

Current CO_2 purification technologies range from i) proven, common, and affordable, to ii) proven and widely available, but significantly more expensive, to iii) less available, unproven, and potentially expensive and/or complex purification options for post-combustion impurities like nitrogen oxides (NO_x).

This paper will discuss well-established purification approaches including glycol absorption, solid desiccant adsorption, refrigeration, liquefaction, distillation, and emerging catalytic processes for oxygen removal. The paper will also review options for removing sulfur dioxide, carbonyl sulfide, organic sulfur compounds, and hydrogen sulfide, and provide an overview of potentially applicable NO_x removal methods for CO_2 purification – likely to be a key challenge in meeting stringent purity specifications for CO_2 from post-combustion sources. CO_2 purification techniques for other contaminants including mercury, radon, and oxygenated and non-oxygenated volatile organic compounds will also be discussed.

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Ray McKaskle, Trimeric Corporation, Buda, TX
Zeke Pickering, Trimeric Corporation, Buda, TX
Kevin Fisher, Trimeric Corporation, Buda, TX
Brian Visioli, Trimeric Corporation, Buda, TX

Introduction

CO_2 Capture, Utilization, and Storage (CCUS) techniques and designs have been developing continuously for the last several decades in response to increasing demand for food and beverage grade CO_2, dry ice, use as a refrigerant or industrial feedstock reagent, crude oil (tertiary or enhanced oil recovery), reduction of greenhouse gas emissions, and even conversion of CO_2 into fuels, cement, or other useful products. Carbon dioxide (CO_2) is captured from a variety of sources, and its use in a variety of applications requires different levels of purification. Every combination of a CO_2 source (producer) and CO_2 sink (consumer) could require unique specifications and purification approaches.

CO_2 product specifications may include limits on components such as water (H_2O), oxygen (O_2), sulfur oxides (SO_x, including SO_2), nitrogen oxides (NO_x, typically the sum of NO and NO_2), methane (CH_4), nitrogen (N_2), hydrocarbons, amines, mercury, and particulate matter (PM, solids). For example, guidelines have been set forward by the U.S. Department of Energy (DOE) National Energy Technology Laboratory (NETL) [2]. These guidelines are not rigid specifications but instead represented industry consensus on expectations for product purity when published. Specifications have been published by the Northern Lights Project for liquid CO_2 cargo (transportation) [3] and the Compressed Gas Association for use in food and beverage applications [4]. Table 1 shows a comparison of these three specifications.

Table 1. Comparison of three specifications of CO_2 purity

Parameter	**DOE NETL (Pipeline Spec)**	**Northern Lights (liquid CO_2 Spec)**	**CGA G-6.2 (Beverage Grade)**
CO_2 Purity	≥ 95–98 vol%	≥ 99.81 mol%	≥ 99.9 vol%
Water (H_2O)	≤ 500 ppmv	≤ 30 ppm-mol	≤ 20 ppmv
Oxygen (O_2)	≤ 10 ppmv	≤ 10 ppm-mol	≤ 30 ppmv
Sulfur Compounds	≤ 10–20 ppmv	SO_x ≤ 10 ppm-mol H_2S ≤ 1 ppm-mol	SO_2 ≤ 1 ppmv Total (non SO_2) S ≤ 0.1 ppmv
Carbon Monoxide (CO)	≤ 100 ppmv	≤ 100 ppm-mol	≤ 10 ppmv
Nitrogen (N_2)	≤ 4 vol%	≤ 50 ppm-mol	Trace only
Methane (CH_4)	≤ 4 vol%	≤ 100 ppm-mol	≤ 20 ppmv
Organic Hydrocarbons	Specifications on methanol, ethanol, MEG, TEG, BTEX, C_3+, and VOCs	Specifications on methanol, ethanol, MEG, TEG, BTEX, ethane, ethylene, C_3+, and VOCs	Specifications on methanol, acetaldehyde, benzene, VOCs, and non-volatile residue
Particulates	Dry, free of solids	≤ 1 µm	No visible solids

In this paper, Trimeric provides an overview of some common treatments used to purify CO_2, with a focus on CCUS applications. Each treatment is aimed at removal of a specific contaminant or set of contaminants. This paper focuses primarily on commercially available and demonstrated technologies, though a few select emerging/developing technologies are included since the more widespread use of increasingly stringent CO_2 purity specifications has created a demand for them.

Water Removal (Dehydration)

Water is a common contaminant in CO_2 CCUS source streams either as a product of combustion or due to passing through a packed-bed water scrubber or through an absorber with a water-based solvent (e.g., MEA) in upstream processes. Depending on the source, pressure, and temperature of the captured CO_2, its water content may be as high as 5 vol% (2,375 lb H_2O per MMSCF). This water usually needs to be removed to mitigate risks of freezing, $(CO_2\text{-}H_2O)_s$ hydrate formation, and corrosion in downstream equipment. Water removal specifications may be provided as water content (e.g., < 7 lb/MMSCF or 148 ppmv) or as dew point (e.g. -75°F at 1 atm), which can be readily converted to other units.

The most commonly applied methods for dehydration of CO_2 include cooling and condensation, absorption using liquid solvents, and adsorption using solid desiccants. Selection between these methods depends on the final water specification, downstream unit operations, and the temperature, pressure, and water content of the feed gas.

Cooling and Condensation

Cooling and condensation are often selected when the final water specification is not strict. This method takes advantage of the relationship between the saturated water content of CO_2 and pressure [5]. This behavior is different from sweet natural gas which exhibits a decreasing saturated water content with increasing pressure. With CO_2, water content decreases with increasing pressure to a minimum water content around 700 psig and then increases again with increasing pressure as the density of the CO_2 increases through liquid and/or supercritical phases. When CO_2 compression and water removal are both required, one of the compressor interstage pressures is usually selected to be near the minimum on the water content curve that corresponds with the intercooler outlet temperature. For example, at an intercooler outlet temperature of 122°F, the minimum water content occurs around 950 psig and is approximately 170 lb/MMSCF (3,600 ppmv). At an intercooler outlet temperature of 65°F, the minimum water content occurs around 750 psig and is approximately 40 lb/MMSCF (422 ppmv). Process engineers can leverage understanding of the relationship between water content and pressure of CO_2 to design an optimal cooling and condensation process configuration.

Absorption (Liquid Desiccants)

Various liquid desiccants have been used to remove water from CO_2, including glycerol, mono-, di-, tri-, and tetra- ethylene glycol, and propylene glycol. The most common liquid desiccant used for CO_2 dehydration is triethylene glycol (TEG), which is also used frequently in natural gas dehydration. The achievable water content using TEG absorption is similar for CO_2 and natural gas (±10%) [6]. Although the dehydration of these two gases with TEG can be treated similarly, care must be taken to account for the differences in achievable water specifications as well as materials of construction for dehydration unit components. TEG-based dehydration units for treating CO_2 can be designed to achieve a water content as low as 10 lb/MMSCF (211 ppmv) with a 380°F reboiler temperature. Achieving a lower specification such as 7 lb/MMSCF (147 ppmv) requires higher reboiler operating temperatures (e.g. 395°F) and/or the addition of a stripping column to remove additional water from the glycol after the reboiler. However, exposing glycol to excessively high temperatures, localized heat flux, or both can accelerate thermal degradation of the glycol.

Process descriptions are readily available regarding the use of TEG dehydration for natural gas [7], and these unit operations are similar for TEG dehydration of CO_2. Due to corrosion concerns, a TEG unit for dehydration of CO_2 typically requires more 304/304L or 316/316L stainless steel components in the more corrosive sections of the process. Many design guidelines for TEG dehydration of natural gas, such as TEG circulating rates, number of stages in the contactor, or stripping gas flowrate, may be extended to CO_2 dehydration [8].

If the CO_2 requiring dehydration is already at a pressure above the critical pressure of CO_2 (1,071 psia), then glycerol is often used as the liquid desiccant in place of TEG. The solubility of TEG in CO_2 increases to unacceptable levels at pressures above about 1,000 psig, which would lead to excessive glycol loss into the high-pressure CO_2. There are many design differences that must be considered when designing glycerol dehydration units compared to TEG based units [9]. For example, absorber operating temperatures are typically higher because glycerol has higher viscosity than TEG. Regenerator conditions also differ in most glycerol applications. For example, the regenerator operates at a lower temperature to minimize solvent degradation, as glycerol degradation occurs at lower temperatures than TEG. Additionally, supercritical CO_2 holds more water than CO_2 below its critical pressure, requiring a higher solvent circulation rate. Accurate property data or validated predictions are essential for designs involving supercritical CO_2.

Adsorption (Solid Desiccants)

Adsorption-based dehydration systems are generally capable of drying CO_2 to lower water content levels than absorption-based technologies. Molecular sieves are often used when the minimum water content in CO_2 is required, for example, upstream of CO_2 liquefaction. Based on Trimeric project experience, capital and operating costs are approximately 50% higher for adsorption dehydration units than for TEG absorption dehydration units. However, molecular sieves can consistently achieve CO_2 water content as low as 1 ppmv (-150°F dew point) [5].

When there is tolerance for slightly higher water content, activated alumina or silica gel may be used as the dehydration media instead of molecular sieves. Activated alumina and silica gel are less expensive than molecular sieve media and can achieve 10 ppmv water content in the product CO_2 (-90 to -60°F dew point) [5], which is satisfactory for many CCUS projects.

Adsorption dehydration is typically based on a two-bed system with one media contacting bed in dehydration mode and the second bed in regeneration mode. In dehydration mode the wet inlet gas flows down the bed at process pressure (typically > 300 psig) and exits as dry gas at the bottom. In regeneration mode, heated, dry regeneration gas flows up the bed and wet regeneration gas exits the top of the bed. Regeneration frequently takes place at low pressure for CO_2 dehydration (typically < 50 psig / 0.7 bar(g)) which allows for more rapid and more complete desorption of the water.

Regeneration gas is typically supplied as a slipstream of the dry gas exiting the filter downstream of the dryer bed, heated in the regeneration gas heater to at least 400°F. The regeneration temperature required depends on the adsorption media used [10]. Distillation column overhead vent gas streams are an alternate source of regeneration gas at CO_2 plants using distillation, but these plants typically also have a backup of dry product gas taken immediately downstream of the dehydration unit. The water saturated regeneration gas leaving the regeneration bed can be recycled (requiring compression / recompression) or it can be vented to the atmosphere. Switching the beds between regeneration mode and dehydration mode is done with automated valves that are usually controlled by timers in the programmable logic controller (PLC) based on the dehydration and regeneration times selected during the design phase [5]. The drying media typically requires replacement every three to five years in a CO_2 drying application.

Oxygen Removal

Oxygen (O_2) is a contaminant in some CO_2 streams. CO_2 pipelines and CO_2 liquid applications often have a stringent limit for O_2. It is difficult to treat a CO_2 stream to remove O_2, so the preferred approach is to try and limit the introduction of oxygen at the source, but that is not possible in all cases. In some enhanced oil recovery (EOR) and pipeline applications in the United States, oxygen is limited to less than 10 ppmw (or 10 ppmv) in the bulk CO_2 and liquid CO_2 specifications limit O_2 to 30 ppmv. Some of the concerns with oxygen in CO_2 for CCUS applications are as follows:

- Once CO_2 containing O_2 contacts water, the mixture is more corrosive than CO_2 and water in the absence of O_2
- O_2 can lead to biological growth in the formation
- O_2 can oxidize TEG and glycerol
- O_2 can react with H_2S and SO_2 under certain conditions to form elemental sulfur, sulfuric acid, and / or other sulfur compounds

CO_2 sourced from industrial processes is normally provided at low pressures, often atmospheric pressure. Air ingress in areas of the process that operate below atmospheric pressure will introduce O_2 (and N_2) into the CO_2 gas stream. This is a common problem in recovering CO_2 from ethanol plant sources. During fermenter batch operations including drop, fill, and clean-in-place (CIP), air is sometimes pulled into the headspace of the fermenter, and it can then be routed to the CO_2 header that connects the fermenter gas header to the beer well and then the beer well gas header to the wet scrubber. Some fermentation processes such as yeast propagation also intentionally add air or oxygen that ends up in the captured CO_2. As a result, oxygen is a key component for removal in many ethanol plant CCUS applications.

Oxygen Removal by Liquefaction and Distillation

The most widely applied method for oxygen removal in CO_2 is liquefaction followed by distillation. In this process, the CO_2 is compressed, dehydrated, liquefied (i.e., condensed using refrigeration), and then the oxygen and other low-boiling point components are separated from the liquid CO_2 in a distillation or stripping column. Figure 1 shows an example CO_2 liquefaction process flow diagram.

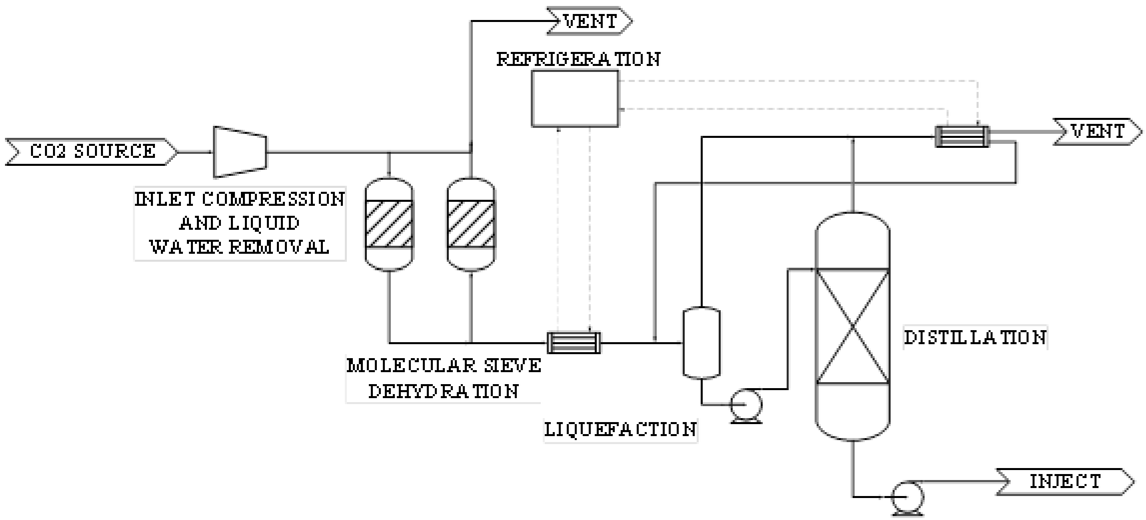

Figure 1 - CO_2 Liquefaction Process Flow Diagram Example

In the CO_2 liquefaction and distillation process, inlet compression is typically achieved using multistage centrifugal blowers followed by oil-flooded screw compressors, which raise the feed gas pressure from 0 to 20 psig and from 18 to approximately 300 psig, respectively. For larger-scale operations, alternative compression technologies such as integral gear centrifugal compressors may provide equivalent performance at a lower cost. Dehydration is commonly performed using molecular sieves, as discussed previously. The dry CO_2 gas is then cooled using refrigeration. Several refrigerants are suitable for use in the liquefaction plant, including anhydrous ammonia and propane, with ammonia being the most prevalent. Liquefaction occurs at pressures between 250 and 300 psig, corresponding to a liquid CO_2 temperature range of -8 to 1 °F. While higher liquefaction pressures are feasible, they necessitate increased feed gas compression, which entails additional capital and operating costs. The distillation column used in this process functions as a stripping column. Liquid CO_2 is pumped to the top of the column, and a reboiler at the bottom generates vapor that travels up the column, stripping oxygen and other low-boiling components from the liquid that is traveling down the column. The purified liquid CO_2 exits from the bottom of the column and is pumped either out of the facility or to storage. For applications requiring high recovery, an optional overhead condenser can be employed to cool the vapor from the top of the stripping column and recover additional CO_2.

This CO_2 liquefaction and distillation process handles moderate fluctuations in gas composition, including air concentrations ranging from 0 to 10%. With proper design, the system maintains high CO_2 recovery rates while ensuring the final product remains within specification. Additionally, hydrocarbons heavier than ethane tend to be condensed into the liquid CO_2 product rather than being lost to the overhead stream in the distillation column. The molecular sieve dryers and downstream equipment may be constructed from carbon steel or low-temperature carbon steel, which reduces capital costs. This process is also well-established in commercial applications, with hundreds of successful installations worldwide. Facilities with capacities under 20 MMSCFD (1,052 tonne/day) are often modular designs, allowing for efficient construction and scalability.

Despite the advantages mentioned above, the CO_2 liquefaction and distillation process has several notable drawbacks. First, the feed gas must be relatively pure; if it contains more than 10% low-boiling components such as N_2, O_2, CH_4, H_2, and/or CO, CO_2 losses vented to the atmosphere through the top of the stripping column increase significantly, potentially making the process economically unviable. Additionally, the process is energy intensive. Oil-flooded screw compressors, which are readily available and have excellent turndown capability when equipped with slide valve controls, are relatively inefficient, and the high volumetric flow rate of low-pressure gas often necessitates multiple compressors in parallel to handle the full CO_2 stream. Refrigeration compressors also consume substantial electricity. For example, a 20 MMSCFD CO_2 liquefaction facility designed to recover 95% of the feed CO_2 typically requires around 8 MW of electrical power. Based on Trimeric project experience, the capital costs for a refrigeration, liquefaction and distillation facility are on the order of 10% higher than a straight compression and TEG dehydration facility (a common choice when oxygen removal is not required), but the operating costs for the refrigeration, liquefaction and distillation facility are on the order of 50% higher primarily due to the energy consumption of the refrigerant compressor(s).

Another concern involves the hazards associated with handling liquid CO_2. Operating at low temperatures increases the risk of dry ice (solid CO_2) formation within equipment. Sublimation of the dry ice or trapped liquid CO_2 can lead to rapid over-pressurization events. If liquid CO_2 is accidentally released into the atmosphere, it immediately flashes into a mixture of dry ice and extremely cold vapor. This cold vapor with a higher molecular weight than air tends to settle in low-lying areas or accumulate in confined spaces and can remain hazardous for an extended period of time before it fully disperses.

Oxygen Removal by Catalytic Oxygen Reduction

Oxygen can be reacted with fuel in the presence of a reducing catalyst to convert the oxygen to water and CO_2. Hydrogen is another fuel choice to consider since it only produces water when it reacts with O_2, but H_2 is expensive and can be difficult to transport and store. In the catalytic oxygen removal process, the feed gas is compressed, pre-treated, and heated before flowing through the catalyst bed to convert the oxygen to water (and CO_2 if fuel other than H_2 is used). Some heat integration may take place downstream of the catalyst bed before the treated gas flows to other parts of the process. Figure 2 shows a process flow diagram of the catalytic oxygen removal process.

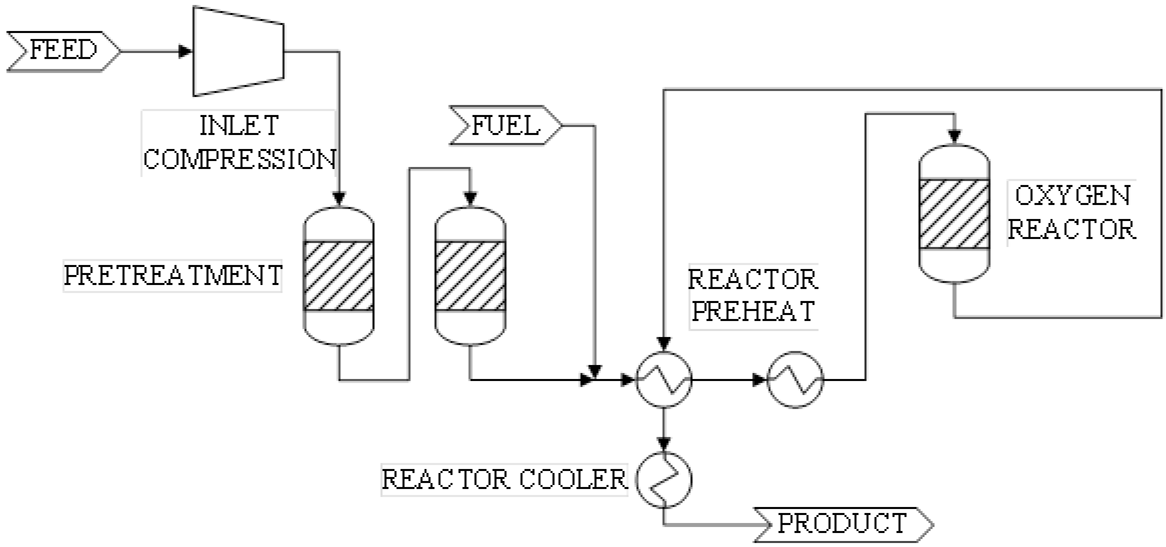

Figure 2 - Process Flow Diagram for Catalytic Oxygen Removal

The catalytic oxygen removal process begins with inlet compression. This can be achieved using a standalone compressor or using a multi-stage compressor system, where the early stages handle feed compression and the later stages compress the product gas. Pretreatment requirements vary depending on the specific CO_2 feed gas composition and may include oil removal filters, carbon beds (for removal of organic contaminants), hydrotreating beds (for converting and/or removing sulfur compounds), and/or sulfur adsorbent beds. The reaction between oxygen and the reducing agent is exothermic and may require the gases to be at elevated temperatures to proceed effectively. As a result, additional heating may be necessary during start-up and may also be required during steady-state operation, particularly when oxygen concentrations are low. The reactor catalyst is often composed of precious metals such as platinum and/or palladium, which can impose more strict pretreatment requirements to avoid catalyst poisoning.

The catalytic process offers several potential advantages over CO_2 liquefaction. It generally has lower operating costs if refrigeration and liquefaction are not already needed for other reasons. The overall electricity demand for catalytic oxygen removal is typically less than that for CO_2 liquefaction due to elimination of the refrigerant compression and despite the higher CO_2 gas compression cost (compared to pumping of liquid CO_2 in the liquefaction process). The process is also less complex, avoiding phase changes and refrigeration systems, which simplifies design and operation. Dehydration requirements are less stringent in applications where liquefaction of the product CO_2 is not required. However, if refrigeration and liquefaction are already required for other reasons (e.g., the capture process produces liquid CO_2, the CO_2 will be transported as a liquid and/or distillated to remove additional light compounds such as N_2 or CH_4), then the benefits of the catalytic process are diminished.

However, there are notable potential drawbacks with catalytic reduction to remove oxygen from CO_2. While oxidation-based versions of this process are proven in CO_2 applications (where hydrocarbons are removed by reacting with excess oxygen), the reduction-based version, where oxygen reacts with excess fuel, has not been commercially demonstrated in CO_2 plants to the knowledge of the authors. A similar reduction process has been demonstrated in argon purification using hydrogen [11]. Proper mixing of gas streams before entering the reactor and control of temperature in the reactor are critical. Furthermore, oxygen concentrations in the feed gas often vary, requiring either rigorous online analysis of O_2 content with real-time adjustment in the fuel injection rate or consistent over-injection of fuel to ensure complete oxygen conversion. Over-injection of fuel can waste valuable hydrogen or increase carbon monoxide emissions and depending on the fuel used, increase methane or VOC emissions. This online analysis adds cost and complexity and may also extend to detection of potential catalyst poisons, especially sulfur compounds. The deactivation of an expensive precious-metal catalyst is also of concern, due to high replacement costs and potentially limited catalyst availability. Spare catalyst systems might be required to ensure reliable plant runtime. Trimeric is aware of several sulfur contamination events in ethanol plant CO_2 capture application that would likely have poisoned catalyst systems if any had been present.

SO_2 Removal

Sulfur dioxide (SO_2) poses two primary issues for post-combustion CO_2 capture: its presence in post-combustion flue gas may result in inefficiencies in CO_2 capture, and its presence in the CO_2 product stream may pose a corrosion or safety risk for the transport system and/or end user. For these reasons and because SO_2 removal from CO_2 is challenging, SO_2 is commonly removed upstream of the carbon capture unit.

For amine-based CO_2 capture processes, SO_2 in the flue gas (feed to CO_2 capture plant) will result in degradation of the solvent and/or sorbent. This in turn will cause increased solvent/sorbent usage and increased solvent/sorbent waste. Solvents tailored for CO_2 capture are typically more expensive than generic MEA (monoethanolamine). Amine-based CO_2 capture technology vendors recommend limiting SO_2 concentrations in the incoming flue gas to <1 ppmv. The amount of SO_2 found in combustion flue gas can vary widely (less than one to hundreds of ppmv) depending on the sulfur content of the fuel and the types of SO_2 controls in place at power plant or other combustion unit, if any.

SO_2 that is not removed upstream of the carbon capture unit will be either absorbed by the solvent or contained in the CO_2 product. In some CO_2 capture technologies including some types of membranes, the SO_2 can be removed selectively along with the CO_2 product resulting in higher SO_2 concentrations in the captured CO_2 than in the flue gas source. SO_2 concentration limits in the CO_2 product are project specific and depend on the other impurities in the CO_2 product, the transport system, and the end use. The limits for SO_2 concentration may be driven by concerns for limiting corrosion and limiting toxic health exposures.

In many cases, the presence of other contaminants in the CO_2 product significantly influences how much SO_2 can be tolerated in the system. While pure SO_2 and dry SO_2/CO_2 mixtures do not cause corrosion in carbon steel piping, the introduction of additional contaminants can lead to compounding corrosion effects. For instance, water in the CO_2 stream can exacerbate corrosion when SO_2 and oxygen are both present, because oxygen can react with SO_2 to form sulfuric acid (H_2SO_4). The presence of SO_2 is particularly concerning in enhanced oil recovery (EOR) applications, as it can come back to the surface in the production well with CO_2 and water (brine) that are co-produced with the oil. The Immediately Dangerous to Life or Health (IDLH) concentration for SO_2 is 100 ppmv, so careful monitoring and control of SO_2 levels are essential to ensure safety and environmental compliance.

SO_2 removal can be achieved upstream of the carbon capture unit by incorporating caustic scrubbing into the direct contact cooler used to cool the flue gas, often achieving less than 1 ppmv SO_2 in the flue gas. The Direct Contact Cooler (DCC) is a counter-current one- or two-stage water contactor which cools the flue gas stream and condenses the water present in the flue gas stream. In addition, caustic (NaOH) may be added to the recirculating water to achieve <1 ppmv SO_2 in the treated flue gas.

H_2S Removal

Hydrogen sulfide (H_2S) is a common contaminant in CO_2 streams. Both H_2S and CO_2 occur naturally in crude oil and natural gas. Therefore, H_2S can be present when CO_2 is captured from these sources – for example by removal of acid gases from sour oil and gas. The amount of H_2S that will be present in the CO_2-rich stream varies depending on the source, and can range from a few parts per million, to a significant mole percentage.

Many technology options exist for removal of H_2S. The most frequently employed technologies are scavenging, liquid redox, and gas treating (e.g., amine, physical solvent) units combined with Claus plants. Appropriate selection will depend primarily on the amount (concentration and total quantity) of H_2S in the feed stream, as well as treated gas specification and emissions limits. These will vary for different CO_2 applications such as pipeline transportation, enhanced oil recovery, or CO_2 sequestration. Limitations on the amount of H_2S that can be vented to the atmosphere will influence the treatment selection for any sulfur-containing off-gas .Figure 3 shows process technology applicability generated by Trimeric that is based on the amount of H_2S in the feed stream.

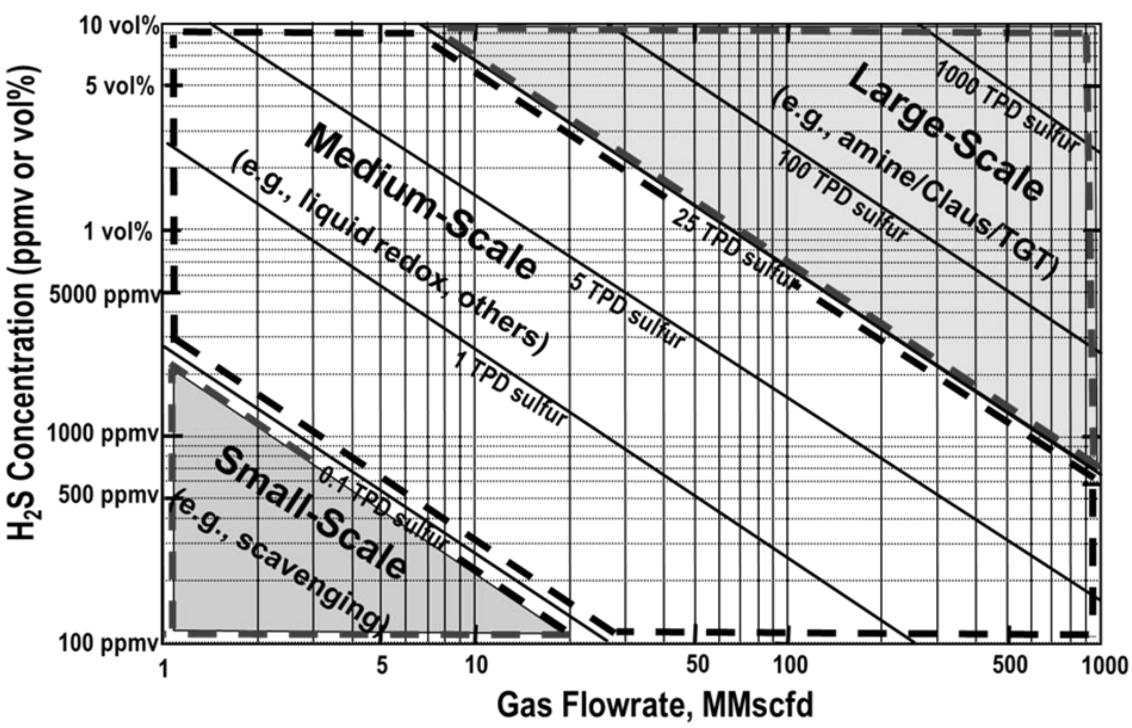

Figure 3 - Process Technology Selection for H_2S Removal

H_2S Scavenging

In the small-scale range, H_2S removal technologies typically focus only on removing the H_2S, without significant regard to the form or quality of the product sulfur. The most common removal method at this scale is H_2S scavengers.

H_2S scavengers are a broad class of compounds that react with H_2S to form non-volatile sulfur-containing compounds before being discarded. Scavengers can be either liquid or solid. Metal oxides on an inert substrate and triazine-based liquids are common examples. Solid scavengers such as iron sponge and SulfaTreat® are widely used for small-scale applications, typically where sulfur removal is less than about 500 pounds per day. These systems operate by passing sour gas over a bed of reactive material, converting H_2S to stable sulfides. While effective and simple, scavenging does not scale up well because frequent replacement and disposal of spent material drive up operating costs and introduce safety concerns. For this reason, scavengers are best employed in situations where only a small amount of H_2S must be removed, as shown in Figure 3.

Liquid Redox and Other Medium-scale Technologies

As the sulfur content of the CO_2 gas increases, treatment methods transition from a focus on removal and disposal to the possibility of recovering sulfur as a saleable product.

Several options exist for medium-scale H_2S removal options. The most common is liquid redox, in which typically an iron-based catalyst facilitates the oxidation of H_2S to elemental sulfur using air as the oxidant. Many licensed processes exist for this application. In addition to liquid redox, biological processes and non-aqueous processes may be used. There are also combinations that use liquid redox processes along with an amine or physical solvent treating unit. Many of these processes are challenging to operate and careful attention to process conditions, chemistry, additive concentrations, and other factors are required by the operator to maintain satisfactory performance and uptime, especially when the gas is processed at elevated pressure. Value of the sulfur product generated by these processes may be minimal.

Several options exist for medium-scale H_2S removal, with liquid redox being the most common and widely proven approach. In these systems, an iron-based catalyst in an aqueous solution facilitates the oxidation of H_2S to elemental sulfur using air as the oxidant [12]. The process operates at near-ambient conditions, making it inherently safer than high-temperature alternatives like Claus. Examples of licensed technologies in this space are LO-CAT® and SulFerox®, and they are typically applied where sulfur loads range from about 0.5 to 20 tons per day.

The liquid redox system consists of an absorber, where H_2S is absorbed and converted to sulfur, and an oxidizer, where the catalyst is regenerated by sparging air. Sulfur is removed as a filter cake that may have significant (30-80 wt%) water content. While the chemical reactions are straightforward, successful operation requires careful control of pH, redox potential, and additive concentrations to prevent foaming, plugging, and excessive salt buildup. Byproducts such as thiosulfate and sulfate can accumulate, sometimes necessitating solution bleed and replacement. Although the sulfur product can occasionally be marketed as fertilizer or soil amendment, its value is generally minimal and therefore it is commonly disposed of in landfills.

Alternatives to liquid redox for medium-scale sulfur recovery include biological processes (e.g., Thiopaq O&G), non-aqueous systems like CrystaSulf®, and hybrid configurations that combine liquid redox with amine or physical solvent treating units. These technologies offer flexibility and high removal efficiency (>99.9%), but they involve higher capital costs than scavengers and require more operator attention to maintain reliability and uptime [12].

Amine/Physical Solvents and Claus Plant Sulfur Recovery

For large-scale H_2S removal, many different commercial options are available. The general process usually involves a solvent-based H_2S removal process, where the solvent is an aqueous alkanolamine (e.g. methyldiethanolamine, or MDEA), a physical solvent, or an alkaline salt solution. Selection of the solvent depends on the composition and flow rate of the feed gas, and target H_2S specification.

In the first step, H_2S is absorbed into the solvent in an absorber column. The rich solvent is then regenerated by heating and stripping, releasing a concentrated acid gas stream that is routed to a Claus-type sulfur recovery unit (SRU). Claus SRUs routinely achieve recovery of more than 97% of the fed H_2S as elemental sulfur. The SRU may or may not be followed by a tail gas treating unit (TGTU) to enable recovery of more than 99% of the H_2S as elemental sulfur.

In the TGTU, the Claus tail gas containing 1-5% H_2S and SO_2 is mixed with hydrogen and reacted across a catalyst bed containing a transition metal sulfide hydrotreating catalyst (commonly Co-Mo/Al_2O_3). This catalyst facilitates the reduction of SO_2 and elemental sulfur to H_2S, which may then in turn be removed in another absorber column and recycled back to the front end of the Claus SRU. This reaction is non-selective and results in all sulfur compounds are converted to H_2S. While this reduction-absorption tail gas treatment process configuration is commonly employed downstream of Claus SRUs, other process configurations may be used to clean up the final 1-5% sulfur compounds in the Claus tail gas.

This Claus and TGTU process technology is the most cost-effective option on a per-pound basis for high sulfur loads, and is considered the industry standard for large, continuous operations. This process comes with significant capital cost, long lead times, and operational complexity, including the need for robust metallurgy and heat management. While the sulfur product is typically saleable, a viable market may not always be nearby, and for smaller-scale projects such as many CCUS applications, the lack of a product sulfur market may impact the technology selection decision.

Nitrogen Oxides (NO_x)

Nitrogen oxides (NO_x) refers to any combination of NO and NO_2 that may be present in flue gas streams generated from combustion processes. NO_x species are formed via multiple pathways during the combustion process (e.g., oxidation of nitrogen species in the fuel, oxidation of nitrogen in the combustion air). The resultant total NO_x content and distribution of NO_x as NO and NO_2 vary based on the specific details of the combustion process (temperature, residence time, fuel utilized, level of excess air and inerts, implementation of NO_x reduction or abatement technologies, etc. In general, post-combustion flue gas may contain total NO_x ranging from tens to hundreds of ppmv with NO_2 representing approximately 5-10% of the total NO_x. When a CO_2 capture process is applied to a post-combustion flue gas stream, the capture process may be impacted by the presence of NO_x in multiple ways, such as degradation of amine or other chemical solvents or inability to selectively separate NO_x from CO_2.

There are multiple well-established commercial approaches to reduce the NO_x content of flue gas. The combustion process and equipment may be modified to reduce the formation of NO_x (e.g., low-NO_x burners or exhaust gas recirculation (EGR)), or treatments which remove the NO_x downstream of the combustion process may be employed (e.g., Selective Catalytic Reduction (SCR) or Selective Non-Catalytic Reduction (SNCR)). The management of NO_x in the combustion process and/or flue gas are not the primary focus of this work; even if these upstream processes are present, they will not completely eliminate NO_x and, therefore, NO_x may ultimately be present in the CO_2 captured from the from the flue gas via a carbon capture process. The focus in this paper is management of the NO_x once it has reached the CO_2 product stream.

NO_x species are known to be corrosive. In addition, according to information in literature [13], the corrosion risk of NO_x may not simply be limited to situations where a free water phase is present. The presence of NO_x and other acid gas species (e.g., SO_x, H_2S) can lead to more complex mechanisms involving reactions between species and/or the formation of a separate acidic aqueous phase that occurs when water content is well below the saturation level in the bulk CO_2. Therefore, it may not be correct to assume that dehydration of CO_2 will fully mitigate risks associated with NO_x in the CO_2 product. Efforts to improve the understanding of the corrosion potential of species such as NO_x in CO_2 streams remain an active area of research. Regardless of the mechanism/risk of NO_x in the CO_2 product, it may be possible that CO_2 product specification will require further removal of NO_x in CO_2 from post-combustion capture processes.

Given that NO_x contamination of CO_2 streams from post-combustion capture processes is expected to occur and that CO_2 product specifications must be met (to appropriately mitigate the risks associated with NO_x contamination), NO_x removal processes may be required for the concentrated CO_2 product. Partial removal of NO_x can be affected by coincidental removal, scrubbing and distillation, or by chemical oxidation.

Coincidental Removal of NO_x in CO_2 processing

As discussed earlier, the CO_2 from a CO_2 capture process will typically undergo additional processing before being ready for transport. For example, the CO_2 will be compressed, with cooling and condensation of water interstage. The CO_2 will also be dehydrated. These additional process steps for CO_2 may remove some of the NO_x present in the CO_2 coincidentally (Table 2). While this incidental removal may not be suitable or sufficient to reliably meet CO_2 product specifications for NO_x, understanding the disposition of NO_x in the CO_2 processing system will be important in creating an overall mitigation approach.

Table 2 - Incidental Removal of NO_x in CO_2 Processing

Process Step	**Reported NO_x Removal** [14]	**Limitations**
NO_x removal in Compressor Condensate	Minimal at 10 psig (0.7 barg). ~ 60% at 216 psig (15 barg).	Limited by what dissolves in the water. NO_2 can be converted to NO and Nitric Acid in the Condensate and NO Can Desorb Back into the Gas.
NO_x removal in Solid Sorbent Dehydration Beds	~ 15%	NO_X ends up in Dryer Regeneration Gas (May be vented or recycled in the process)
NO_x removal in Compressor Oil	High acid number in oil after 850 hours of operation	Increasing oil monitoring requirements. Could impact oil consumption and/or compressor reliability

NO_x Removal via CO_2 Scrubbing and Distillation

In principle, NO_x removal could be accomplished via a multi-step scrubbing and distillation process with CO_2. For species with a higher boiling point than CO_2, such as NO_2, liquid CO_2 can be used directly as a scrubbing agent. In this process, liquid CO_2 counter-currently contacts a CO_2 gas stream contaminated with the NO_2 in a packed or trayed tower. The NO_2 is removed from the CO_2 gas stream while the lighter NO slips in the CO_2 gas to the overhead of the column. The NO could then be removed in a separate distillation tower with other light gases such oxygen and nitrogen to produce the final liquid CO_2 product stream. The liquid CO_2 scrubbing agent leaving the first column will be contaminated with NO_2 and must still be managed in some manner (e.g., via thermal oxidizer with NO_x reduction) as it will not be a suitable CO_2 product.

This approach would require vaporizing all of the CO_2 product if it starts as a CO_2 liquid contaminated with NO_2, followed by condensing it back to a liquid (if required to separate NO from CO_2, for transportation, or other reason). It would also be expected to increase the refrigeration duty and energy requirements by about 20% per tonne of product CO_2 basis and results in a lower net CO_2 production rate. This CO_2 purification technology has some commercial applications for removal of other heavy species, but Trimeric is not aware of any commercial applications for NO_x removal from CO_2.

NO_x Removal via Chemical Oxidants

NO_x removal in general may also be accomplished with a variety of chemical oxidants incorporated as part of an aqueous scrubbing process. In these processes, gas and liquid phase reactions and mass transfer result in NO_x being scrubbed into solution and ultimately converted to a solution of nitric acids and/or nitrates. Ozone [15] and hydrogen peroxide [16] are offered commercially for this purpose. Ozone would be directly added to the gas stream whereas hydrogen peroxide would be added to the scrubbing solution. The specific process configuration and selection of oxidant required to achieve specifications for NO_x (both NO and NO_2) would need to be defined in conjunction with a technology provider with experience in relevant applications. Pilot testing would likely be required to ensure that the process will meet commercial objectives if these approaches are applied to a CO_2 stream.

Table 3 summarizes the two NO_x scrubbing processes reviewed in this paper. Both processes are currently at lower technology readiness level.

Table 3 - NO_x Scrubbing and Distillation Process Options

Process	Advantages	Limitations
CO_2 Scrubbing and Distillation	No additional chemical usage, no wastewater stream	May not be fully commercially demonstrated for CO_2 applications, increases energy requirements to produce liquid CO_2 scrubbing agent, higher CO_2 losses than distillation alone, waste CO_2 stream requires disposition, multiple towers.
NO_x removal via Chemical Oxidants	Can be designed to selectively remove NO_x while minimizing CO_2 losses, minimal impact on energy use.	May not be fully commercially demonstrated for CO_2 applications. Potentially significant new chemical consumption requirement, aqueous waste stream that requires disposition (will have nitrates present).

Other Contaminants in CCUS

Ammonia

Ammonia can react with CO_2 to form ammonium carbamate salts under some conditions. This has been observed in one instance where an economizer (ammonia / CO_2 heat exchanger with ammonia on the tube side and CO_2 on the shell side) had a tube leak into the CO_2 that was captured from a fertilizer plant, and it could be an issue in post-combustion flue gas that has SCR or SNCR with ammonia injection that leads to some ammonia slip.

Sulfur Contaminants

There have been a handful of incidents that Trimeric is aware of at ethanol plant CO_2 capture facilities which led to one or more types of sulfur contaminant accumulation. Impellers on multistage centrifugal blowers at the inlet of ethanol plant CO_2 capture facilities may need to be cleaned or replaced approximately every five years due to contamination buildup.

Compressor Oils

Screw compressors and reciprocating compressors use lubrication oil in contact with the CO_2 that can end up in produced water (wastewater) from ethanol plant capture facilities and in the product CO_2. Primary and secondary coalescers are often used following oil-flooded screw compressors to reduce oil contamination to single digit ppm levels (difficult to measure). A change from mineral oil to synthetic oil for cylinder lubrication in a reciprocating compressor and making sure that the oil injection rate is selected for the proper oil type and proper maximum operating (not design) compressor speed (rpm) can reduce the buildup of asphaltenes in piping and injection wells downstream of CO_2 capture facilities.

Glycol

Glycol carryover from the dehydration unit can also contaminate product CO_2. Maintaining proper CO_2 and glycol flow rates and using a dehydration unit discharge coalescer can reduce this problem.

Sulfur oxides

In flue gas streams, a portion of SO_2 oxidizes to SO_3, which subsequently reacts with water to form liquid H_2SO_4 droplets in the direct contact cooler (DCC). These droplets can serve as nucleation sites for additional aerosol formation, creating fine particles that are difficult to remove. Several technologies are available for aerosol mitigation. Alkaline sorbent injection upstream of particulate matter control devices can remove up to 99% of H_2SO_4. Wet electrostatic precipitators (WESP) are highly effective for capturing sulfuric acid and non-sulfuric acid aerosols, achieving approximately 90% removal with one field and up to 99% with two fields. Absorber water wash systems can also be designed to enhance aerosol removal efficiency. Combinations of these technologies are often employed to achieve stringent aerosol control requirements.

Mercury

Mercury (Hg) is a hazardous air pollutant commonly present in flue gas streams containing CO_2. Its presence is particularly problematic for brazed aluminum components used in "cold box" heat exchangers. Several strategies are available for mercury removal. One approach involves "co-benefit" removal through other emission control systems, such as selective catalytic reduction (SCR) and flue gas desulfurization (FGD), which inherently reduce mercury concentrations. Another method is the injection of activated carbon upstream of the particulate matter (PM) removal device to adsorb mercury from the gas stream. Additionally, halogen compounds can be introduced into coal in coal-fired power plants to promote mercury oxidation, facilitating its subsequent removal in the FGD system.

Radon

Radon has also been found in some naturally occurring sources of CO_2. Published studies indicate that zeolites [17], activated carbon [18], and metal oxides [19] have all been proposed for radon removal.

Conclusions

The variety of contaminants found in captured CO_2 is extensive, and the potential applications for the recovered CO_2 product are numerous as well. Each combination of a CO_2 source and a CO_2 use or sink may come with its own unique specifications and purification technologies. An important consideration which is consistent across all applications is that the selection of CO_2 purification technologies is a complex decision and each individual application requires careful selection of specifications and purification technologies in order to be successful from economic and operability perspectives.

In this paper, Trimeric has provided an overview of purification technologies for removal of key and common contaminants including H_2O, O_2, SO_2, H_2S, and NO_x. There are multiple proven options for water removal (dehydration) of CO_2 and selection depends on the specification and operator preferences. Oxygen removal from CO_2 is usually done using CO_2 liquefaction and distillation, but the additional costs of this technology, particularly when the concentrations of O_2 in the feed gas are <100 ppmv, are driving the emergence of catalytic reduction technologies which have been demonstrated in other applications, but not yet for CCUS applications based on Trimeric current industry knowledge that is informed by frequent searching and discussions with customers and suppliers. If it is possible to prevent air ingress from contaminating CO_2 with oxygen (and nitrogen), this is often preferred over oxygen removal. Removal of NO_x from CO_2 is complex, with different strategies required for the NO vs. the NO_2 component. The best option may be to reduce the NO_x levels in the source gas with upstream equipment such as Low-NO_x burners and SCR / SNCR. Emerging distillation and scrubbing technologies for NO_x removal are discussed and compared. SO_2 is frequently removed to < 1 ppmv by injecting caustic to the circulating water based on pH control in a direct contact cooler (DCC) that is often needed to cool and remove excess water from flue gas CO_2 sources as the first step in most post-combustion CO_2 capture facilities. H_2S removal technology selection is primarily driven by the amount of sulfur compounds present, with scavenger applications being favored in many CCUS projects due to the relative simplicity of the process and feasible due to lower inlet H_2S concentrations in the feed gas and lower overall treated gas flow rates (relative to applications such as refinery streams). Liquid redox and Claus are other options for higher sulfur loading applications. Other sources of CO_2 contaminants were discussed including ammonia, other sulfur compounds including SO_3 and other aerosols, compressor oils, dehydration solvents (e.g., glycol), mercury, and radon.

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