

Carbon capture and storage technologies: present scenario and drivers of innovation

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Carbon capture and storage (CCS) technologies are being developed to comply with the intensification of environmental laws and policies. Techniques for carbon capture from exhaust gases include post-combustion, pre-combustion and oxy-combustion. CO₂ separation in gas processing is also a relevant application, employing alternatives commonly used in post-combustion, sharing developments and pulling innovations (additional to innovations pushed by knowledge from basic and applied research). The high volume of exhaust gases and expanding reserves of natural gas defy the state-of-the-art in chemical and physical absorption (the most mature technology). The review identifies technological gaps and drivers of innovation in the CCS chain. In the context of offshore natural gas processing, this work reports a recent and massive technological niche for commercial use of membrane based processes.

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Introduction

Carbon capture and storage (CCS) comprises separation of CO₂ from industrial sources, compression and transportation to a geologic site for storage, or to enhanced oil recovery (EOR). Its uses cover a variety of industrial applications, outstanding abatement of CO₂ from process or exhaust gases and in natural gas (NG) processing. In the former, depending on the technology, CO₂ is separated from H₂ (pre-combustion), N₂ (post-combustion) and H₂O (oxy-combustion, which burns hydrocarbons with pure O₂) [1], while in NG processing CO₂ is separated from CH₄ and light hydrocarbons [2].

Flue gas is released from carbon-fired power plants at moderate temperature (50–100°C) and low pressure (<1.5 bar). Post-combustion with chemical absorption or physical absorption are the technologies closest to full scale realization and preferred for retrofitting [3^{**}]. Although post-combustion and NG processing may employ different technologies, capture of CO₂ by chemical and physical absorption are their leading options, where the solvent loading (α , mol CO₂ per mol of active solvent, *AS*) is a capture response while the capture ratio (*CR*, kg of total solvent per kg of fed CO₂) and the solvent regeneration heat ratio (*HR*, kJ per kg of fed CO₂) are input factors [4^{**}]. In chemical absorption, CO₂ and the *AS* chemically bond giving high selectivity and low hydrocarbon losses (NG processing) with maximum stoichiometric α of 1 molCO₂/mol at *CR* 10–15 kg/kgCO₂ and reversibly requiring high solvent regeneration *HR* (2000–4500 kJ/kgCO₂). In physical absorption, weak physical binding of CO₂ to solvent reduces selectivity, but can give $\alpha > 1$ molCO₂/mol of *AS* at low *CR* (1–5 kg/kgCO₂) and low *HR* (0–500 kJ/kgCO₂) for solvent regeneration. In chemical and physical absorption, the equilibrium α increases with CO₂ fugacity (CO₂ partial pressure) and decreases with increasing temperature [86^{**}].

Pre-combustion firstly reforms fossil fuel to synthesis gas (syngas, H₂ + CO), and, in a second step, to H₂ and CO₂ via water-gas shift (WGS) reaction. H₂ is purified via chemical or physical absorption of CO₂ (easy separation due to high CO₂ partial pressure) and can fuel supercritical boilers, gas turbine (in H₂-fired power plants) or promisingly used in integrated gasification-combined cycle (IGCC) power plants [5]. In H₂-IGCC, high capital expenditure (CAPEX) of syngas, WGS and capture units are drawbacks, and H₂ as fuel requires development of new power machines, another H₂-IGCC risk [6]. In typical coal-fired power plants, the power efficiency reduces from 38.4% without CO₂ capture to 31.2% with CO₂ capture [7], a susceptibility eliminated by changing to full Coal-H₂-IGCC. The capture energy penalty in a carbon-fired power plant is the fraction of power output lost by implementing CO₂ capture.

Oxy-combustion eliminates N₂ in oxidizer of carbon-fired power plants [5], substituting CO₂-N₂ post-combustion separation by O₂-N₂ fractionation via cryogenic distillation, the most cost-effective commercially available route, though with refrigeration energy penalties, in the same range as that for fossil fuel de-carbonization [8]. As oxy-combustion flame temperature [5].

Oxy-combustion is not yet commercial, posing greater technical risks than pre-combustion or post-combustion for large-scales [6]. Porter *et al.* [9] discuss cost and CO₂ purity variations for oxy-combustion and pre-combustion scenarios.

In NG processing, CO₂ must be removed to comply with treated gas specifications. A determinant change in the technological scenario is pulled by offshore NG processing, mainly at ultra-deep waters on FPSO (Floating Production, Storage & Offloading) platforms. Membrane permeation offers advantages over conventional chemical or physical absorption for NG processing: small footprints, modularity and easy scale-up. The treated NG is the membrane permeation retentate at high pressure, which fits the final compression for pipeline dispatch.

Considering the state-of-the-art, Figure 1 depicts the CCS scenario, contemplating CO₂-EOR and other CO₂ sources (*e.g.*, fertilizers, cement and steel production), including bioethanol plants producing food grade CO₂ from fermenters, which can be directed to downstream CCS. This work analyzes the main technologies involved, focusing in identifying technological gaps requiring innovations and technology drivers in the big CCS scenario. Table 1 presents a compilation of state-of-the-art and advanced processes, including those at lower Technology Readiness Level (TRL) [72], from proof-of-concept to small pilot plants.

Carbon capture from exhaust gases

Capture energy penalty on carbon-fired power plants is significant ($\approx 15\text{--}30\%$) [10] representing 65–80% of CCS costs [11,12^{**}]. To retrofit carbon-fired power plants with 33% power efficiency, a decrease of 12% of efficiency represents more than 1/3 of power output [13], with a capital expenditure (CAPEX) increase of $\sim 77\%$ [14]. Carbon-fired power plants face large variations in CO₂ emissions due to differences in efficiency and employed fuel: coal-fired power plants emit 1116 gCO₂/kWh at 30% and 669 gCO₂/kWh at 50% of efficiency [15,5].

Despite coal being the most CO₂ intensive option, capacity expansion plans [67] indicate that carbon mitigation initiatives are insufficient to outweigh the economic incentives of a relatively cheap fuel. Concerning CAPEX, NG-fired power plants configure the best alternative with half CAPEX of coal-fired power plants and 1/5 of nuclear plants [16]. Impacts on operational costs (OPEX) are quantified mainly by simulation [17]. Uncertainties in overall performance are estimated probabilistically [18]. CAPEX estimation uncertainties are high ($\pm 40\%$), though variability has little influence on the levelized cost of energy (LCOE) [19], suggesting that OPEX dominates CCS.

Boot-Handford *et al.* [20^{**}] present extensive review on leading CO₂ capture technologies, available in the short and long term and their maturity. Post-combustion CO₂ capture employing chemical absorption remains the most efficient and cost-effective capture [21], with heat demand (OPEX) for solvent regeneration as main drawback, reducing power capacity (capture energy penalty $\approx 10\text{--}30\%$), despite recent improvements lowering heat ratio (*HR*, energy penalty for solvent regeneration) from 5.5 to 2.6 GJ/tCO₂. Carbon-fired power plant repowering or hybridization using solar-assisted post-combustion may conciliate capture and power plant load targets [22]. Limitations of driving force indicate that state-of-the-art membrane permeation are unlikely to compete with chemical absorption in capturing CO₂ from exhaust gases [21].

The deployment of renewable energy substitutes partially the need of (fossil) carbon-fired power plants, reducing the amount of fossil-fuel burned. However, renewable energy dispatch is intermittent, demanding flexible operation of the capture unit to improve the economics of CCS power plants [23]; flexibility allows exploring this transient pattern to reduce CAPEX up to 28% [73]. With chemical absorption, flexibility can be achieved by solvent storage, exhaust gas venting (decoupling energy generation from CO₂ capture, to meet peak energy prices) and time-varying solvent regeneration (allowing CO₂ to accumulate in the solvent at peak energy prices) [24]. Variable capture aligned to energy demand and dispatch [25] results in temporary reduction of capture energy penalty, increasing net efficiency and capacity [26]. For instance, the absorber sized for a time-average condition costs $\sim 4\%$ less than when sized for peak energy generation [27].

Capture energy penalty can be minimized by new solvents or flowsheet modifications, reducing power losses by 25% [28^{**}], conciliating the tradeoff of sensible heat loss (to raise the temperature of the stripper feed) at high solvent rate (high lean loading) and stripping steam use at low solvent rate (low lean loading) [20^{**}]. Additionally, low solvent thermochemical stability [29^{*}] leads to accumulation of degradation products and toxic emissions [30].

Evolving from the first commercial plant (first of a kind, FOAK) to the *n*th commercial plant (*n*th of a kind, NOAK) reduces OPEX and CAPEX [31]. Alternative technologies are sought, posing greater risk because of their earlier stage of development [6]. Emerging technologies (*e.g.*, new membranes and solvents) with potential for ‘game-changing’ improvements are still scheduled to large-scale testing by 2025 and complete demonstration scale testing by 2030 [64]. Besides low TRL (Technology Readiness Level), a major issue in post-combustion

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