



# Upgrading of natural gas ultra-rich in carbon dioxide: Optimal arrangement of membrane skids and polishing with chemical absorption



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## ABSTRACT

Water depths in oil and gas offshore production are moving from shallow to deep and ultra-deepwaters. Floating Production Storage and Offloading platforms are preferred in such frontier offshore enterprises. However, ultra-deepwaters natural gas processing imposes challenges in the design of floating units, limited in area and weight of processing equipment. Oil reserves with high gas/oil ratio ( $>250 \text{ sm}^3/\text{m}^3$ ) and high carbon dioxide content create additional challenges due to the impacts in the deck area of the gas plant. Among other natural gas processing operations, removal of carbon dioxide is required to meet sales gas specification, being skid-mounted membrane modules well suited for this purpose. To avoid emissions and to increase oil production, the separated acid gas is injected for enhanced oil recovery, creating a changing scenario due to increasing carbon dioxide content in the reservoir along production lifetime. In this context, this work optimizes arrangements of membranes modules and operational conditions via nonlinear programming formulations to optimize total membrane area for minimum footprint of membrane skids, with either carbon dioxide content in the treated natural gas less than 3% mol (Type 1 Constraint) or methane losses in the injection gas limited by imposing carbon dioxide content in the injection gas greater than 75%mol (Type 2 Constraint). To reduce computational effort, surface response models are employed, regressed from data simulated with a rigorous phenomenological model of spiral-wound membrane modules. Optimization results – total and stage area, carbon dioxide contents in retentates and permeates and natural gas production – are obtained for three feed scenarios: 10%mol, 30%mol and 50%mol carbon dioxide in raw natural gas. Type 1 Constraint leads to higher methane losses while Type 2 Constraint demands a polishing Chemical Absorption to comply with carbon dioxide specification of sales gas, configuring a hybrid process. Life cycle costs and total footprint area point to superiority of carbon dioxide separation design resulting from Type 2 Constraint, with optimal service distribution between bulk removal in Membrane Permeation and polishing operation in Chemical Absorption, considering time-varying composition of the raw natural gas.

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## 1. Introduction

Water depths in oil and gas exploration and production (E&P) are expanding to frontier offshore regions (ultra-deepwaters) (EIA, 2016), where Floating Production Storage and Offloading (FPSO)

platforms are preferred. For instance, Libra Field, is located in the Brazilian pre-salt layer, ~200 km south offshore of Rio de Janeiro city and approximately 2000 m deep, and has associated gas with carbon dioxide (CO<sub>2</sub>) content in the order of 40mol% CO<sub>2</sub> (Arinelli et al., 2015). Design of FPSO is challenged by high gas/oil Ratio ( $>250 \text{ sm}^3/\text{m}^3$ ) and natural gas with high CO<sub>2</sub> contents (Gaffney et al., 2010), due to area and weight of gas processing equipment (Andrade et al., 2015; Araújo et al., 2017).

CO<sub>2</sub> removal from natural gas reduces the gas volume to be transported, increases the heating value of sale gas and avoids emissions (Peters et al., 2011). Such benefits, among other drivers,

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results in limits to sale natural gas in Brazil to 3% CO<sub>2</sub> (ANP, 2008). Consequently, CO<sub>2</sub> separation and destination become two major technological challenges for producing natural gas in Brazilian ultra-deepwaters. Membrane Permeation stands out for being capable of processing CO<sub>2</sub> rich feeds, having good weight/space efficiency, being ideally used in remote locations. Since the driving force for Membrane Permeation mass transfer is the trans-membrane difference of partial pressures (fugacities), the main membrane permeation disadvantage is the power required for compression to provide the partial pressure difference (Beggel et al., 2010). It also has other advantages, such as lower capital and operational costs, simplicity and high reliability (Cnop et al., 2007) and, thus, is the leading CO<sub>2</sub> separation technology in Brazilian ultra-deepwaters E&P.

It is relevant to offshore CO<sub>2</sub> separation from natural gas that a correct destination is required, with Enhanced Oil Recovery (CO<sub>2</sub>-EOR) being the best choice as it increases well productivity and, therefore, monetizes CO<sub>2</sub>. Although Gazalpour et al. (2005) consider low technical challenges for CO<sub>2</sub>-EOR projects for onshore operations, in offshore applications, reinjection of CO<sub>2</sub> rich gas faces the scenario of high cost CO<sub>2</sub> separation and low oil price as the main barrier for oil producers to apply the technology. Nevertheless, the high CO<sub>2</sub> content in natural gas in the Brazilian pre-salt, induced early CO<sub>2</sub>-EOR (Malone et al., 2014). It is worth mentioning that the continuous reinjection of CO<sub>2</sub> contributes to the gradual increase in CO<sub>2</sub> content (%) in the reservoir, as more than 50% of injected CO<sub>2</sub> – injected gas (IG) – returns to the surface with the oil and gas products (Kwak et al., 2014). Consequently, along operation time, the CO<sub>2</sub> content in the produced natural gas is expected to rise, imposing the need of flexibility in offshore CO<sub>2</sub> separation technology.

Facing these challenges, Membrane Permeation systems have competitive advantage due to its modularity. By connecting membrane modules, one can promptly adapt the membrane plant to changing flow rates and compositions. Fig. 1 displays the configuration of offshore natural gas processing in the Brazilian pre-salt, highlighting the CO<sub>2</sub> separation step approached in this work.

Specification on CO<sub>2</sub> in sales natural gas and high flow rates, typical of offshore E&P in Brazilian Pre-Salt, demand more complex approaches than the conventional single-stage applications that dominate the scene. The literature on Membrane Permeation is dedicated to analyses of single stage membrane units, evaluating the conflict flow rate – selectivity in response to design parameters such as membrane area (Lindqvist et al., 2014). Although the natural gas processing literature offers a wide range of studies for natural gas sweetening, very few approach the scenario of CO<sub>2</sub> rich natural gas at high capacities. Additionally, hybrid (single-stage) Membrane Permeation plus Chemical Absorption with amines, is a promising alternative for facing ultra-deepwaters requirements (Araújo et al., 2017).

Cook and Echt (2006) reported operation of membrane modules which suffered a 35% increase in process feed with respect to design conditions, with raw natural gas approaching 15%mol CO<sub>2</sub> and treated natural gas with <2%mol CO<sub>2</sub>. The increase of capacity, with fixed membrane area resulted in decreased loss of hydrocarbon (CH<sub>4</sub>) to the permeate. Their result evidences that oversized membrane area occurred for low feed flow rate operation, and suggests the need of exploring module number (increasing modules in operation as capacity increases) as a strategy of attributing operational flexibility.

Alkatheri et al. (2016) investigated, for a fixed content of CO<sub>2</sub> in natural gas (10%mol), the impact of varying the CO<sub>2</sub> removal fraction in an upstream Membrane Permeation module (one stage configuration), responsible for bulk CO<sub>2</sub> removal, and a secondary

polishing step via Chemical Absorption. Araújo et al. (2017) used one-stage Membrane Permeation to reduce CO<sub>2</sub> content to 4%mol and achieved sales natural gas specification (<3%mol) in a downstream Chemical Absorption operation under three CO<sub>2</sub> content levels in the gas feed: 10%mol, 30%mol and 50%mol CO<sub>2</sub>.

A gap in the literature was identified concerning the impact of CO<sub>2</sub> content in the raw natural gas in terms of membrane arrangement performances (single and multiple modules) as the only CO<sub>2</sub> separation process or as part of a hybrid process combining Membrane Permeation arrangements and Chemical Absorption. Furthermore, absence of accounting hydrocarbon losses in the separation design was identified, being most design procedures exclusively oriented towards specification of CO<sub>2</sub> in sales natural gas.

The arrangement of Membrane Permeation modules with varying CO<sub>2</sub> content and gas flow rates, with impact in total footprint area, poses a nonlinear optimization design problem. In this context, this work aims to optimize the total membrane area, resulting in minimum footprint and weight of the Membrane Permeation plant considering three membrane arrangements – 1 stage, 2 stages and 3 stages of permeation, referred as 1S, 2S and 3S – under two types of product constraint: (i) Type 1 Constraint – upper bound of CO<sub>2</sub> content in the treated natural gas; and (ii) Type 2 Constraint – lower bound of CO<sub>2</sub> content in the injection gas, indirectly imposing upper bound of CH<sub>4</sub> losses in the injection gas. The comparison of performances of the designs with optimized total membrane area having stage areas as decision variables for 1S, 2S and 3S arrangements and the two types of product constraints, allows defining guidelines for designing ultra-deepwaters gas processing units.

The design of the CO<sub>2</sub> removal plant is subjected to three levels of CO<sub>2</sub> content in the gas feed – 10%, 30% and 50%mol – and three membrane arrangements – 1S, 2S and 3S – totaling 9 combinations. Considering the two types of product constraints that can be applied, results a grand-total of 18 nonlinear programming optimization design problems, all solved with CONOPT solver of GAMS - General Algebraic Modeling System.

As designs subjected to Type 2 Constraint cannot guarantee a sufficiently low CO<sub>2</sub> content in the treated natural gas, there is a gap to be fulfilled by some finishing step. Therefore, such designs result in hybrid Membrane Permeation – Chemical Absorption processes, with the Chemical Absorption operation evaluated by process simulation with Aspen-HYSYS. Optimized design alternatives are compared in terms of achieved footprint, life cycle costs (LCC, obtained from mass and energy balances and resulting equipment sizing) and hydrocarbon losses.

It is relevant to mention that, to the authors' best knowledge, the evaluation of hybrid Membrane Permeation – Chemical Absorption technology has been limited in the literature to one Membrane Permeation stage followed by Chemical Absorption, exclusively varying the fraction of the CO<sub>2</sub> removal service performed in the membrane, frequently deprived of hydrocarbon loss control. In contrast to this, this work investigates Membrane Permeation configurations – 1S, 2S and 3S – for CO<sub>2</sub> removal aiming at the two end-products: specified sales natural gas and CO<sub>2</sub> rich fluid to EOR. The results of this work contribute to the field of CO<sub>2</sub> separation from CO<sub>2</sub>-rich raw natural gas at high gas/oil ratio, implying high capacity natural gas processing and CO<sub>2</sub> removal on the topside of FPSOs.

## 2. Theory of high capacity CO<sub>2</sub> removal from natural gas

Only a few technologies are commercially available for high capacity services of removal of CO<sub>2</sub> from natural gas on offshore platforms like FPSOs. Excluding cryogenic distillation, which is

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