

Selecting Technologies for Sour and Ultra-Sour Gas Treating

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Abstract

As the cleanest fossil fuel, natural gas plays a key role in the path towards renewables. Considering the increasing gas demand, rich CO₂ and H₂S gas reserves, in the past considered economically unviable, are becoming fruitful. However, the non-conventional nature of this kind of gases, in addition to the potentially higher production cost, raises the need of new strategies for their monetization, bypassing the conventional approaches. Starting from the huge number of novel large-scale projects for the exploitation of rich-H₂S gas fields, this paper overviews the current tendencies in sour and ultra-

sour natural gas production, focusing on the removal of sulfur-based compounds, together with H₂S and CO₂. At first, available technologies for ultra-sour gas treatment are discussed. Then, simulations of the absorption processes based on a real case-study are carried out, in order to verify the effectiveness of the proposed alternatives for the removal of mercaptans, as well as CO₂ and H₂S. Results are critically analyzed, in view of providing a practical guide of industrial interest for the selection of the most suitable method.

Keywords: Gas treating, H₂S removal, H₂S+CO₂ removal, Mercaptans removal, Ultra-sour natural gas reserves

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1 Introduction

Energy sustainability is the main concern in controlling climate change. However, an effective alternative to fossil fuels to achieve the decarbonization target [1] needs to be established yet, making unrealistic the scenario of a fast transition to green and renewable energy. Among fossil fuels, natural gas is the cleanest one and is commonly regarded as the key player in the shift towards renewables [1, 2].


In this context, ultra-sour gas reserves, undeveloped or underdeveloped so far due to the prohibitive cost of extraction and conditioning, are becoming profitable [3]. Unlike the conventional natural gas reserves, these kinds of gas show high CO₂ and H₂S contents, which need to be reduced to no more than 2–3 mol % and 4–5 ppm, respectively, for the production of a pipeline-quality natural gas [4].

Sour and ultra-sour natural gas classification by composition is not univocal [5]. According to Rojey and Jaffret [6], sour natural gases typically show a H₂S content > 1 % and a CO₂ > 2 % by volume, even if the industrial practice addresses as sour gases those with a H₂S content > 10 % [7]. TechnipFMC defines as ultra-sour the reservoirs whose total acid gas content is above 20 vol % [8].

Sour and ultra-sour natural gas fields have been estimated to be approximately 40 % of natural gas reservoirs [9], the majority of which is located in Middle East. Middle East gas reserves are predominantly sour. Regarding the high-CO₂-content gases, there are many unexploited gas reserves with a CO₂ frac-

tion ranging from 15 to 80 % [10]. This has led to the development of many low-temperature technologies (i.e., Ryan-Holmes process [11], Controlled Freeze Zone (CFZTM) process [12], Dual Column Cryogenic Distillation (DCCDTM) process [13–21]), that have been proved to be more energy-effective than the conventional ones, i.e., absorption by amine solutions, for CO₂ concentrations above 8–9 mol % [22].

On the other hand, for high H₂S content gas reservoirs, a number of important projects are growing for making these fields fruitful. Among them, the Shah gas plant, officially commissioned in 2016, was the first project to produce and safely process more than 1 bcf of ultra-sour gas, with a hydrogen sulfide content higher than 23 % [23]. Also, Ghasha's reservoirs, located in the West of Abu Dhabi, are layered into compartmentalized reservoirs with high sulfur gas and condensate (10–25 % H₂S). In particular, the Hail reservoir dry sour gas

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content is about 15 % H₂S, while the Dalma zone, comprising three reservoirs (Hair Dalma, Dalma and Bu Jafair), exhibits a sour condensate quantity of 25 % H₂S [24].

China has found 38 large-scale natural gas fields in Sichuan. Sichuan Basin's yield accounts for 80 % of the national total output, and the proved reserves of high sour gas fields of eastern Sichuan represented by the Puguang, Dukouhe, Luoziyazhai, Tieshanpo gas fields are about $6.4 \times 10^{11} \text{ m}^3$ (sulfur content 6–17 %), while the Bohai Bay Zhaolanzhuang gas field sulfur abundance reaches values as high as 60 % [25]. The State of Kuwait is exploiting sour gas reservoirs in the north and the west of the country, known as the Jurassic production facilities (JPF) and West Kuwait (WK), respectively, to meet the country's domestic gas demand [26]. Tab.1 gives a picture of some sour gas fields distribution, currently in production.

When treating sour and ultra-sour gases, there are still significant economic challenges on how these fields can be monetized cost-effectively. The key challenge is related to H₂S removal and conversion while also addressing huge health, safety, and environmental issues [33]. Also, together with hydrogen sulfide, a non-negligible content of sulfur-based compounds is typically present in sour gas reservoirs, like mercaptans, carbon disulfide (CS₂), and carbonyl sulfide (COS). While hydrogen sulfide and carbon dioxide removal has been extensively analyzed with a great variety of processes [34–37], also optimized from the energy consumption point of view [38, 39], natural gas purification from sulfur-based compounds is less discussed in literature. Nevertheless, S-compounds concentration, as for hydrogen sulfide, has to be lowered down to few ppm for natural gas distribution because of severe toxicity issues. The choice of the most suitable solution depends on a number of factors that have to be accounted for: plant size and location, sour gas composition, mercaptans, CS₂ and COS concentration, target-sale gas specification, and desired hydrogen sulfide disposal.

To help the gas-treating technology selection and addressing typical problems of ultra-sour gas management, this work offers an overview of the sour and ultra-sour gas purification

technologies, focusing, together with H₂S and CO₂ removal, on mercaptans abatement. For this purpose, the performances of three different proprietary solvents (chemical, physical, and hybrid) for removing CO₂, H₂S, and mercaptans are compared by means of a ProTreat[®] v8.0 [40] process simulation.

2 Sour and Ultra-Sour Gas Treating

Acid gas removal can be performed by means of different technologies, which can be broadly distinguished into absorption (chemical and physical solvents), cryogenic distillation, membranes, adsorption or a combination of them, named hybrid configurations. For the preliminary selection of the most appropriate acid gas removal method, based on the acid gas concentration in the feed and the desired acid gas partial concentration in the outlet gas, Fig.1 can be used as a reference [41]. According to Kidnay and Parrish [29], three scenarios are considered: H₂S removal with no CO₂ present (this case is representative of natural gas with a very low content of CO₂ in comparison with H₂S, since in nature H₂S is always associated with CO₂ [6]), simultaneous H₂S and CO₂ removal, and selective removal of H₂S. However, considering the great variety of unconventional natural gas reserves, the choice of the most suitable technology always requires a detailed economic assessment.

Note that in Fig.1 “hybrid solvent” denotes a blend of a chemical and a physical solvent, as in Kidnay and Parrish [29], while the term “hybrid technology” is used with reference to the combination of two or more approaches for acid gas removal, to overcome the disadvantages of the standalone technology, as it is typically reported in literature [42, 43].

The technology application ranges of Fig.1 have been validated referring to existing sour gas fields and their associated gas sweetening technologies. Tab.2 summarizes some of these fields: the H₂S and CO₂ removal processes reported in this table match with the application ranges provided in Fig.1. When selective H₂S removal has to be achieved from a H₂S-

Table 1. Examples of sour natural gas composition (vol %) [8, 27–32].

	Ram River (Alberta)	Burnt Timber Facility (Alberta)	Süd-Old- enburg	Tengiz (Kazakh- stan)	Bearberry ^{a)} (Canada)	Lacq (France)	Qilibei (China)	Kashagan (Kazakh- stan)	Puguang, Sichuan Basin (China)	Uthma- niyah (KSA)	Shah ^{b)} (UAE)	Astra- khan (Russia)
N ₂	2.53	0.5	7	0.8	1	0.4	0.50	1.02	–	7–14		3
CO ₂	8.22	9	8	2.6	5	9.6	3.73	5.06	10	0.5–8	10	19
H ₂ S	35.79	8	8	16	90	15.2	16.30	17.69	14	8–14	23	23
CH ₄	52.34	80	77	42	4	69.3	77.90	58.83	76	60–64		55
C ₂ H ₆	0.41	0.8	0.1	8.5	–	3.1	0.50	9.10	–	–		–
C ₃ H ₈	0.14	0.2	–	5.2	–	1.1	–	4.69	–	–		–
C ₄ H ₁₀	0.16	–	–	3.3	–	0.6	–	2.28	–	–		–
C ₅₊	0.410	1.5	–	22	–	0.7	–	0.95	–	–		–

^{a)} For sulfur production only. ^{b)} Information of hydrocarbon composition as well as other impurities not available in literature.

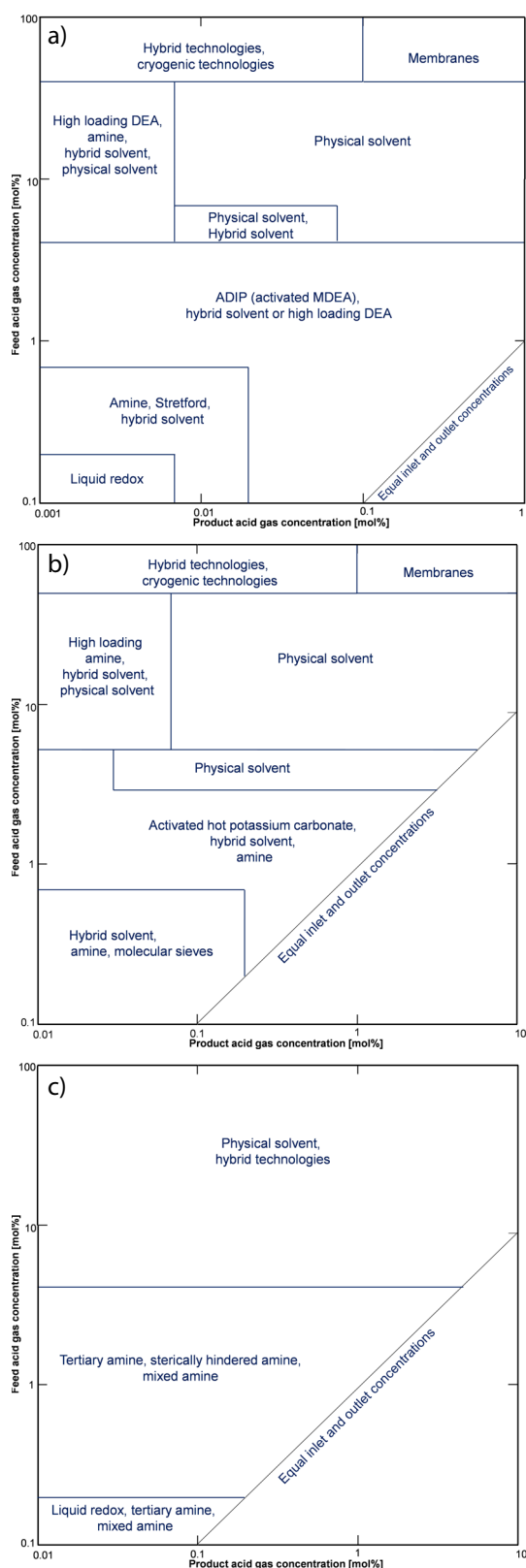


Figure 1. Process selection chart for: (a) H₂S removal with CO₂ present; (b) simultaneous H₂S and CO₂ removal; (c) H₂S removal with CO₂ present (adapted from [29] and [55]).

Table 2. Existing sour gas fields and associated gas sweetening technologies [26, 44–50].

Gas field name	H ₂ S content [mol %]	CO ₂ content [mol %]	Gas sweetening technology	Ref.
Ram River	10–36	2–8	DEA	[32]
Waterton	5–30	5–15	Originally: MEA now: Sulfinol [®]	[51]
Caroline	35		MDEA and Sulfinol [®]	[52]
LaBarge	5	65	Hybrid technologies	[53]
Al Hosn	23	10	DGA	[54]

and CO₂-containing gas, chemical absorption technologies or hybrid technologies can be exploited (Fig. 1c). On the other hand, when both H₂S and CO₂ can be separated in the same acid gas stream, or when mainly H₂S is present, the panorama is wider.

For treating large volumes of ultra-sour gases, cryogenic technologies are proved to process natural gas in a less expensive way, without limitations on CO₂ or H₂S concentration in the inlet stream. Among them, the Dual Column Cryogenic Distillation (DCCDTM) is a new sour gas sweetening process based on cryogenic distillation, available at pilot scale, that allows the separation of acid components as a high-pressure liquid, together with highly pure methane [15]. The distillation is performed in such a way that CO₂ solidification is avoided [56]. The typical process configuration is simplified in Fig. 2 and consists of two distillation columns: the first one is operated at high pressure (50 bar), to avoid CO₂ solid deposits formation, while the second one operates at low pressure (40 bar) below the methane critical pressure.

Unlike the DCCDTM technology, the Controlled Freeze ZoneTM (CFZTM; see Fig. 3), a well-known cryogenic distillation process for acid gas removal, provides for a controlled CO₂ freezing in a specially designed zone within the methane-acid gas distillation column. The process is applicable also when H₂S is present in the feed stream, without limitation on the amount of acid gases in the treated sour gas [57, 58].

With reference to Fig. 1, stand-alone membranes offer the advantage of a simple bulk H₂S removal. Their tolerance or affinity to sulfur-based compounds has to be evaluated case by case. Commercially available membranes solutions include: SourSepTM by Membrane Technology and Research, Inc. (MTR) [59], PEEK-Sep P-Guard technology by Air Liquide [60], GENERON[®] H₂S removal membrane [61], and Integrated Flow Solutions, Inc. (IFS) Membrane Systems [62]. In all these instances, the permeate stream can be either re-injected or flared if its H₂S concentration is not so significant, while it can be sent to a sulfur recovery facility if showing a huge H₂S content. Membranes can be also combined with other sour gas conditioning processes to enhance the overall separation performances and minimize costs. This is the case of hybrid technologies.

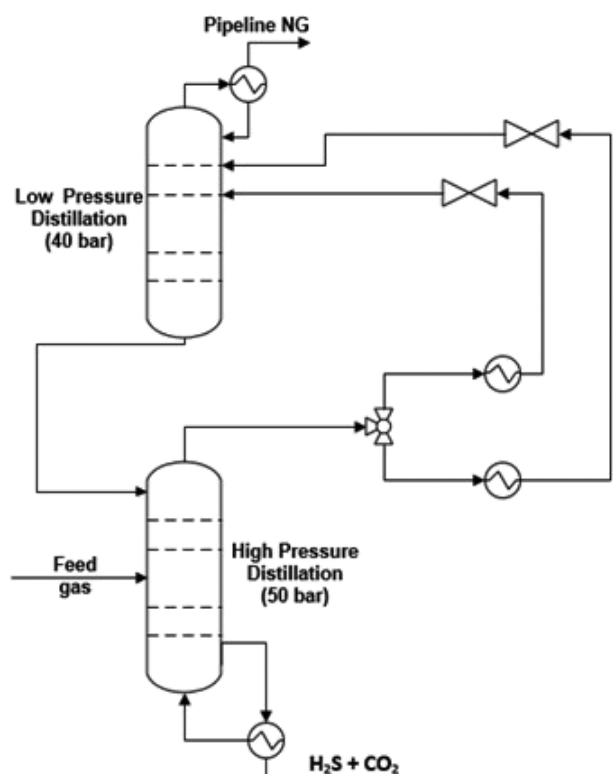


Figure 2. Schematic of the DCCD™.

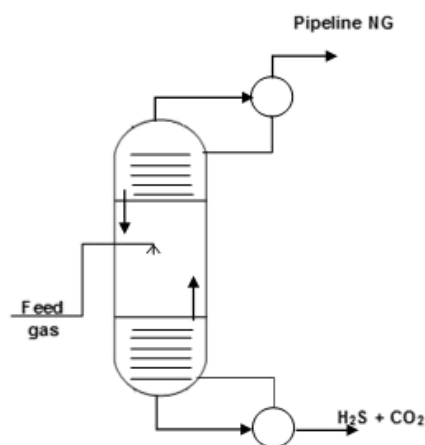


Figure 3. Schematic of the CFZ™.

Hybrid technologies, like cryogenic ones, have the advantage of treating ultra-sour gases in a more cost-effective way than the traditional processing. One of the most applied hybrid technologies is the SPREX® process, whose schematic diagram is depicted in Fig. 4, which was developed by TOTAL and IFP [63]. The technology was initially designed to favor the exploitation of sour gas with a high H₂S content, with values up to 40%, through amine absorption. After that, it was also extended to high-CO₂-content gases (SprexCO₂), to perform the bulk CO₂ removal from natural gas [64]. The SPREX® and the SprexCO₂ processes allow obtaining the acid gases separately,

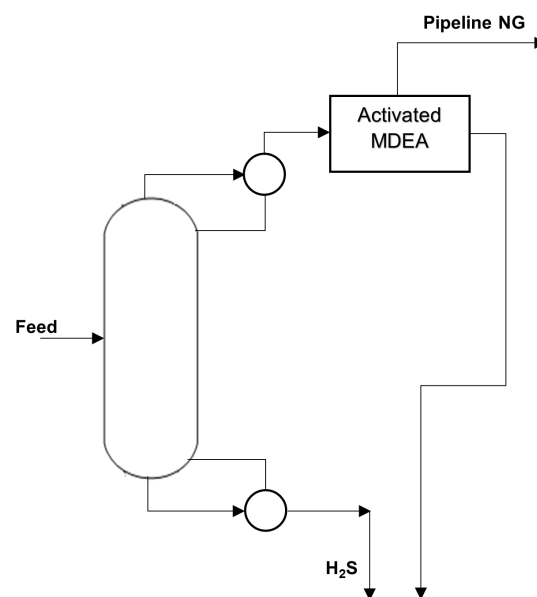


Figure 4. Schematic of the SPREX® process.

for their individual re-injection into gas wells. According to the basic process configuration, the sour natural gas enters the distillation column, operated at high pressure, where H₂S is separated as the bottom liquid phase. The top vapor, containing methane and some residual acid gases, is routed to a methyl diethanol amine (MDEA) absorption stage, where it is further purified to meet the pipeline natural gas specifications [65].

Cryocap™ NG is another example of hybrid technology commercialized by Air Liquide, which combines cryogenic distillation and membranes as reported in Fig. 5. The solution is claimed to be highly efficient for the CO₂ removal, integrating the proprietary Air Liquid membrane technologies PEEK-Sep™ and Medal™, but it is suitable for H₂S and mercaptans gas streams conditioning, too, with 99%+ methane recovery. The small footprint, which makes it suitable for offshore applications, is its main advantage [66].

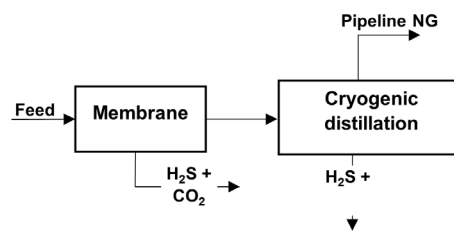


Figure 5. Schematic block flow diagram of the Cryocap™ process.

When the treated gas contains, together with a non-negligible content of acid gases, a significant amount of hydrocarbons C₂₊, the TarT process, developed by 8 Rivers Capital, LCC, and schematically shown in Fig. 6, allows the simultaneous separation of H₂S and CO₂, as well as the C₂₊ fractionation. It uses the liquid CO₂ as the solvent for the separation of H₂S from methane and the subsequent fractionation of CO₂,

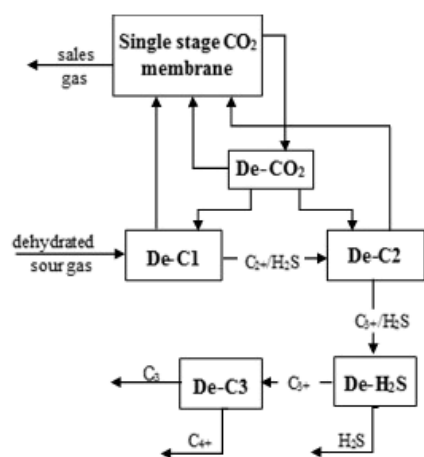


Figure 6. Schematic block flow diagram of the TarT process. Adapted from [67].

H₂S, and C₂₊ within the sour gas. A finishing stage through a single-step membrane allows the methane purification from the residual CO₂. This configuration is proved to reduce the compression energy requirement and the methane loss in the CO₂ product [67].

Regarding adsorption, similarly to absorption, physisorption and chemisorption can be distinguished, depending on the sorbate-sorbent interactions. Adsorption-based processes are best suited to polishing duties [68]. Commercial alternatives for H₂S removal include PURASPEC™ by Johnson Matthey [69] and SULFATREAT and SELECT FAMILY by Schlumberger [70]. PURASPEC and SELECT FAMILY technologies utilize mixed metal oxides to remove H₂S, while SULFATREAT adsorbents are iron oxide-based. Carbonyl sulfide (COS) and mercury removal can be also achieved through PURASPEC™.

When the acid gas inlet concentration is not such that cryogenic or hybrid technologies become advantageous, sweetening of natural gas by absorption is the preferred alternative to focus on [71]. Absorption can take place into traditional packed or trayed equipment, or innovative apparatus, such as the one of the cMIST™ gas treating technology, an inline system that can replace traditional absorbers, whose compact design can be suitable for offshore and remote applications [44].

As the absorption based-technologies application range is quite wide, this work focuses on the different commercially available absorption solvents. Firstly, the most widespread proprietary solvents are reviewed in the following section. Then, to assess the absorbent performance in the sulfur-based contaminants abatement, simulations referred to a real sour gas case study have been performed. Results are critically discussed, in view of providing a practical guide of industrial interest for sour natural gas conditioning.

3 Absorption

The absorption-based processes panorama is extremely wide. As a general remark, high partial pressures of the acid gases in the feed favor physical solvents, whereas low pressures favor

chemical ones. However, the choice of the most suitable solution is not trivial and has to be evaluated referring to the specific case study. The proprietary absorption solvents, classified into chemical, physical or hybrid solvents, are reviewed in the following sections.

3.1 Chemical Solvents

In the case of acid gas removal through chemical absorption, H₂S and CO₂ chemically react with the solvent. Their reactions may be reversible or irreversible. If the reaction is reversible, the solvent can be regenerated in a downstream stage operated at high temperature and low pressure to shift the chemical equilibrium to the left and allow the solvent recirculation. The chemical solvents can be broadly classified into two groups: amine (methyl ethanol amine (MEA), diethanol amine (DEA), diglycol amine (DGA), diisopropyl amine (DIPA), MDEA) and potassium carbonate solutions, nowadays slowly replaced by MDEA solutions. MEA and DEA can remove both H₂S and CO₂ from the inlet gas streams up to sales gas specifications, while, when a selective H₂S removal is required, DIPA or MDEA can be used as solvents. Alongside with these traditional absorbing agents, fully disclosed in literature, a number of new ones have been developed where the amine solution is added with a special chemical activator to meet the final gas specifications. Some of these new solvents and the related technologies are listed in Tab. 3.

The choice of the most suitable solvent for acid gas purification depends on the initial acid gas content and the H₂S/CO₂ ratio of the feed stream. Starting from the feed composition, the absorption process can be optimized in terms of rich and lean solvent loading and solvent circulation rate.

Chemical solvents usually present high acid gas loadings at the expense of high energy requirements for solvent circulation and solvent regeneration. This problem can be partially overcome with hybrid solvents, reported in Sect. 3.3. An advantage, however, is the minimum co-absorption of hydrocarbons, which leads to high hydrocarbon recovery of the overall configuration.

3.2 Physical Solvents

Technologies using physical solvents (e.g., cold methanol or dimethyl ethers of polyethylene glycol) are not directly applicable to high-pressure natural gas due to the significant losses of hydrocarbons in the acid gas. Physical absorption usually takes place at near atmospheric pressure. Some of the patented technologies for acid gas removal through physical solvents are listed in Tab. 4, together with their developer and the solvent involved in the separation process.

As a general remark, the desired selectivity towards H₂S or CO₂ can be adjusted by the proper selection of the operating conditions and through a suitable design of the absorption equipment. Moreover, physical solvents have a better ability to absorb mercaptans rather than chemical ones. For this reason, using a physical absorption step followed by a chemical one can be a smart way to exploit the advantages of both the solvents, despite adding a significant complication of the acid gas removal section.

Table 3. Proprietary solvents for acid gas removal exploiting chemical absorption [26, 44–50].

Process name	Solvent	Developer	Remarks
Flexsorb [®] SE; Flexsorb [®] SE Plus	Sterically hindered amines	ExxonMobil	Lower circulation rate than other amines
OASE [®] sulfexx [™]	Amine-based	ExxonMobil and BASF	Selective removal of H ₂ S while minimizing the co-absorption of CO ₂
OASE [®] purple	Activated MDEA	BASF	Iron content in the solvent < 10 ppm, lower maintenance requirement
HiPACT [®] (High Pressure Acid Gas Capture Technology)	Proprietary	BASF and JGC Corp.	High solvent thermal stability and high absorption performance of CO ₂
AdvAmine [™] (HiLoadDEA, MDEAmax, energizedMDEA)	DEA- and MDEA-based	Prosernat	Tight treated gas specifications on H ₂ S, CO ₂ , but also COS and some mercaptans
Gas/Spec [™] (Gas/Spec SS/CS-1000; Gas/Spec CS-1; Gas/Spec SS/CS-3; Gas/Spec CS-2000; Gas/Spec SG-1060 Gas/Spec CS-1160)	INEOS formulated MDEA	INEOS	Less corrosive and less prone to degradation than MEA, DEA or DIPA. Lower amine circulation rate and energy usage
JEFFTREAT [®] (JEFFTREAT [®] MP, JEFFTREAT [®] MS-100/200/300, JEFFTREAT [®] ULTRA; JEFFTREAT [®] M-500)	MDEA-based	Huntsman	Deep CO ₂ removal with greater acid gas loading capacity, superior chemical stability even at high temperatures, corrosion protection, and longer product life under severe operating conditions.
UCARSOL [™] solvents (UCARSOL [™] HS-101/102/103/ 115; UCARSOL [™] AP 802/804/806/ 810/814)	Specially formulated MDEA	Dow Chemical Company	UCARSOL [™] AP: CO ₂ removal UCARSOL [™] HS: selective H ₂ S removal
Fluor Econamine (Econamine FG; Econamine FG Plus SM)	DGA with side cooler	Fluor Corp.	Lower solution corrosion tendency than MEA. Degradation products formed. (N,N',bis-(hydroxyethoxyethyl) urea – BHEEU, and N,N',bis-(hydroxyethoxyethyl)thiourea – BHEETU), which are not reversible at normal amine regenerator temperature.
AdapT 100	MDEA-based	Eastman Chemical Company	Selective removal of H ₂ S from H ₂ S and CO ₂ containing gas streams.
Benfield-HiPure	DEA-hot carbonate	UOP	Can achieve outlet CO ₂ concentrations as low as 30 ppmv and H ₂ S concentrations of 1 ppmv.
ADIP [®] -X; ADIP-ULTRA	MDEA + piperazine	Shell Global Solutions	CO ₂ removal down to < 50 ppmv for high-pressure process streams.

Table 4. Proprietary solvents for acid gas removal exploiting physical absorption [26, 44, 72–74].

Process name	Solvent	Developer	Remarks
SELEXOL™ SELEXOL™ MAX	Dimethyl ether of polyethylene glycol	UOP	Selective for H ₂ S and sulfur compounds removal
AGR® II Solvent (DEPG)	Dimethyl ether of polyethylene glycol	Coastal AGR	Suitable for hydrogen sulfide, mercaptans, and other sulfur-containing gases removal
EconoSolv	Dimethyl ether of polyethylene glycol	Fluor Corp.	Helpful in eliminating the need for extra tail gas treating units
Fluor Solvent	Propylene carbonate	Fluor Corp.	Suitable for little H ₂ S concentrations, where only CO ₂ removal is required
RECTISOL®	Methanol	Linde AG and Lurgi AG	Either removal of all impurities or selective H ₂ S and sulfur compounds absorption
IFPEXOL™	Methanol	IFP	Simultaneous dehydration and acid gas removal
Purisol	N-Methyl-2-pyrrolidone	Lurgi AG	Selective for H ₂ S removal
Genosorb®	Polyethylene glycol dialkyl ethers	Clariant	–
Morphysorb®	N-Formyl-morpholine (NFM) and N-acetyl-morpholine (NAM)	Gas Technology Institute	Low solubility of C1 to C3 hydrocarbons

3.3 Hybrid Solvents

Another approach for acid gas removal through absorption is to blend the physical and chemical solvents. Most of these mixtures, often defined as “hybrid solvents”, are still at the development stage [75]. A review of these new blends for sour gas applications is available in Pellegrini et al. [76], while the already commercial technologies are reported in Tab. 5. Hybrid solvents try to combine the advantages of physical and chemical solvents, resulting in higher sulfur compounds selectivity and lower energy consumptions compared to the traditional chemical absorption technologies.

3.4 Acid Gas (H₂S, CO₂) and Mercaptans (RSH) Removal: The Kashagan Gas Case Study

To highlight the performances of chemical, physical, and hybrid solvents in sour gases and sulfur-based compounds removal, process simulations have been performed with ProTreat® v8.0 software [40]. This software was selected because of its ability to simulate natural gas treatment using patented solvents, making possible the analysis of case studies of industrial interest. Three solvents, representative of each category and specific for sour-gas applications, have been analyzed: the UCARSOL™ HS-101 by Dow, the dimethyl ether polyethylene glycol (DMEPG) by Coastal AGR, and the Sulfinol®-M by

Table 5. Proprietary solvents for acid gas removal exploiting hybrid solvents [26, 44, 72, 77].

Process name	Solvent	Developer	Remarks
Sulfinol®-D; Sulfinol®-X; Sulfinol®-M	Sulfolane + DIPA/MDEA	Shell Global Solutions	Selective H ₂ S removal over CO ₂ achieved depending on the process modification
Amisol	Methanol + aliphatic alkylamines	Lurgi AG	Can be used for either selective desulfurization or complete removal of CO ₂ , H ₂ S
UCARSOL™ (UCARSOL™ LE 701/702/ 713/777/801)		Dow Chemical Company	UCARSOL™ LE: organic sulfur removal
HySWEET®; HySWEET®DEA; HySWEET®MDEA	Amine solution + ThioDiGlycol	Total	Selective H ₂ S removal over CO ₂ achieved depending on the process modification
Selefining	DMEA + organic solvent	Snamprogetti	Selective H ₂ S removal from gases also containing CO ₂

Shell. UCARSOL™ HS-101 solvent is a specially formulated MDEA-based product, developed to enhance the selective removal of H₂S from contaminated gas streams [78].

DMEPG, also known as Coastal AGR II solvent, is typically used for the removal of H₂S, CO₂, mercaptans, and other sulfur-containing gases from natural gas, synthesis gas, ammonia streams, landfill gas, and Claus feed gas [79]. Sulfinol®-M is a hybrid solvent consisting of a mixture of MDEA (30–45 wt %), sulfolane (40–60 %), and water (5–15 %), particularly suitable for the selective absorption of H₂S and mercaptans, while co-absorbing only part of the CO₂ [80]. For process simulation purposes and to be used as a reference when comparing the disclosed alternatives, a base case with MDEA at 40 wt % as a solvent has been considered.

Process simulations have been performed according to the following basis of design:

- inlet feed available at 25 °C, 52 bar, with a flow rate of 5000 kg h⁻¹, and a composition reported in Tab. 6, which is representative of the Kashagan gas of Tab. 1;
- maximum admissible content of H₂S, CO₂, and mercaptans in the outlet gas of, respectively, 4 ppm vol, 2 vol %, and 6 mg Nm⁻³ [81]. The methane recovery in the sweet gas has been fixed at 99.5 %, while no constraints have been imposed on the residual water content, considering that the sweet gas is routed to a downstream dehydration section.

Only mercaptans have been considered as contaminants since the CS₂ and COS behavior as to absorption is analogous.

The simulated process scheme is a typical absorber-regenerator sequence, as detailed in the following sections, with a proper modification in the case of the physical solvent to reduce the hydrocarbon slip. Both absorption and regeneration columns are packed with Sulzer Mellapak 250X. The operating pressure of the absorption column is fixed at 52 bar, i.e., the pressure of the raw natural gas feed, while the regeneration column operates at about atmospheric pressure (1.7 bar).

3.4.1 Base Case: 40 wt % MDEA

The simulated process scheme for the reference case of chemical absorption with MDEA is reported in Fig. 7. The sour natu-

Table 6. Natural gas composition used in the process simulations. The mercaptans concentration has been calculated according to Cloarec et al. [81], while the water content is determined on the basis of the gas saturation condition.

	Molar composition [mol %]
H ₂ O	0.0870
N ₂	1.0230
CO ₂	5.0740
H ₂ S	17.7380
CH ₄	58.9910
C ₂ H ₆	9.1250
C ₃ H ₈	4.7030
C ₄ H ₁₀ ^{a)}	2.2860
C ₅ H ₁₂ ^{a)}	0.9530
CH ₄ S	0.0150
C ₂ H ₆ S	0.0040
C ₃ H ₈ S	0.0010

^{a)} Both C₄H₁₀ and C₅H₁₂ have to be indented as pseudo C₄ and C₅ fractions, where both *n*- and *i*-isomers are considered. For process simulation purposes, these fractions have been assumed as equivalent *n*-hydrocarbons, taking into account that this simplification should not affect the results.

ral gas, whose conditions and composition are specified in Sect. 3.4, enters the packed absorption column (“Absorber” in Fig. 7), where it is treated by 40 wt % MDEA, to give a pipeline-quality natural gas. The rich solvent, chemically bonded with the removed impurities, is heated in “Lean Rich Exch” ($\Delta T_{app} = 10$ °C) and then routed to a distillation unit, “Regenerator” in Fig. 7, to allow the solvent recovery and its recycle to the absorption column.

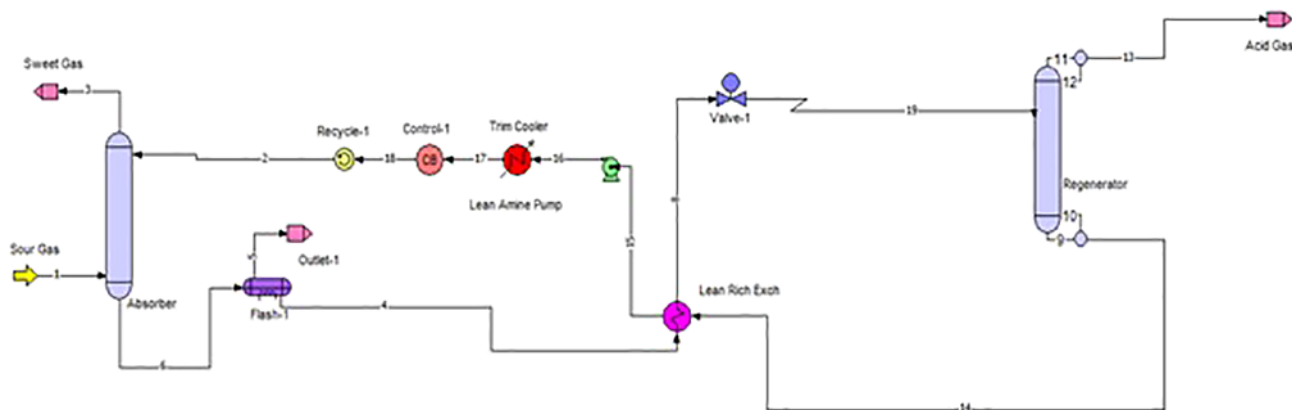


Figure 7. Process scheme of the chemical absorption process. Simulation in ProTreat® v8.0.

3.4.2 Chemical Solvent: UCARSOL™ HS-101

The performance of the chemical solvent UCARSOL™ HS-101 is studied in a process scheme similar to the one reported in Fig. 7, already described for the MDEA reference case.

3.4.3 Physical Solvent: DMEPG by Coastal AGR

For the physical absorption process, the configuration of Fig. 7 is modified to minimize the hydrocarbon slip [82]. As a matter of fact, non-negligible amounts of hydrocarbons can be absorbed in the washing solvent, leading to significant losses within the sour gas removal section. For this reason, downstream the absorption column, three flash chambers, operating at gradually decreasing pressure, have been introduced to favor the hydrocarbon recovery in the vapor phase. The vapor exiting the first chamber (“Flash-1” in Fig. 8) is recycled back to the absorption column, after compression in “Compressor-1” and cooling in “Cooler-1”. The vapor exiting the second and the third flash chambers, “Flash-2” and “Flash-3” in Fig. 8, respectively, are routed to an intermediate absorption column operated at moderate pressure ($P = 13$ bar). The solvent flow rate entering this unit is set to meet the outlet gas specifications. In this way, methane is recovered with a negligible amount of residual H_2S and sulfur-based compounds. Considering this outlet gas to be used as a fuel within the plant, the CO_2 removal performed by this intermediate pressure absorption column is not deep.

3.4.4 Hybrid Solvent: Sulfinol®-M

Regarding the hybrid solvent, the process configuration is the same of the chemical absorption described in Sect. 3.4.1. In this case, the hybrid solvent Sulfinol®-M, not available in ProTreat® simulation software, was introduced as a mixture of sulfolane, methyldiethanolamine, and water, with a weight percent composition of 40, 40, and 20, respectively.

4 Results and Discussion

The process schemes described in Sect. 3.4.1–3.4.3 have been analyzed varying the circulating solvent flow rate as well as the absorption and regeneration column heights to verify the impurity removal efficiency for each solvent type.

Regarding the chemical absorption process with UCARSOL™ HS-101, results are reported in Figs. 9 and 10. In the first simulation set, the absorption column height is varied in the range of 6–14 m while the solvent flow rate is fixed at $17\,000\text{ kg h}^{-1}$. The regeneration column, a distillation unit equipped with a partial condenser (full reflux), is specified fixing the packed height, the molar boilup ratio (i.e., the ratio between the vapor and liquid streams exiting the reboiler), and the top temperature at 40°C to use cooling water as refrigerating medium. As displayed in Fig. 9, the residual H_2S and mercaptans content in the sweet natural gas increases at greater absorber height. This is not the case for CO_2 , whose trend is decreasing at increasing absorber height, as expected.

The results of Figs. 9a and 9b, questionable at a first glance, can be explained considering that, at increasing packed height and a fixed reboiler molar boilup ratio, the solvent is able to pick up slightly more H_2S but, since the reboiler duty is held constant, that extra bit is not being stripped out [83]. On the other hand, CO_2 removal is particularly efficient, since the solvent chemically bonds with CO_2 : its residual content in the sweet natural gas is almost in line with the fixed specification even at the lowest absorber height. The pickup of mercaptans is very sensitive to the lean loading of the amine (all acid gas loading, not just mercaptans loading). Mercaptans are weaker acids compared to CO_2 and H_2S and so the solvent will preferentially pick up those before picking up mercaptans [77]. To lower H_2S and mercaptans concentration, to be consistent with the pipeline-quality natural gas specifications, the solvent flow rate has to be increased.

Results for the UCARSOL™ HS-101 chemical absorption at variable solvent flow rate are reported in Fig. 10. In this case,

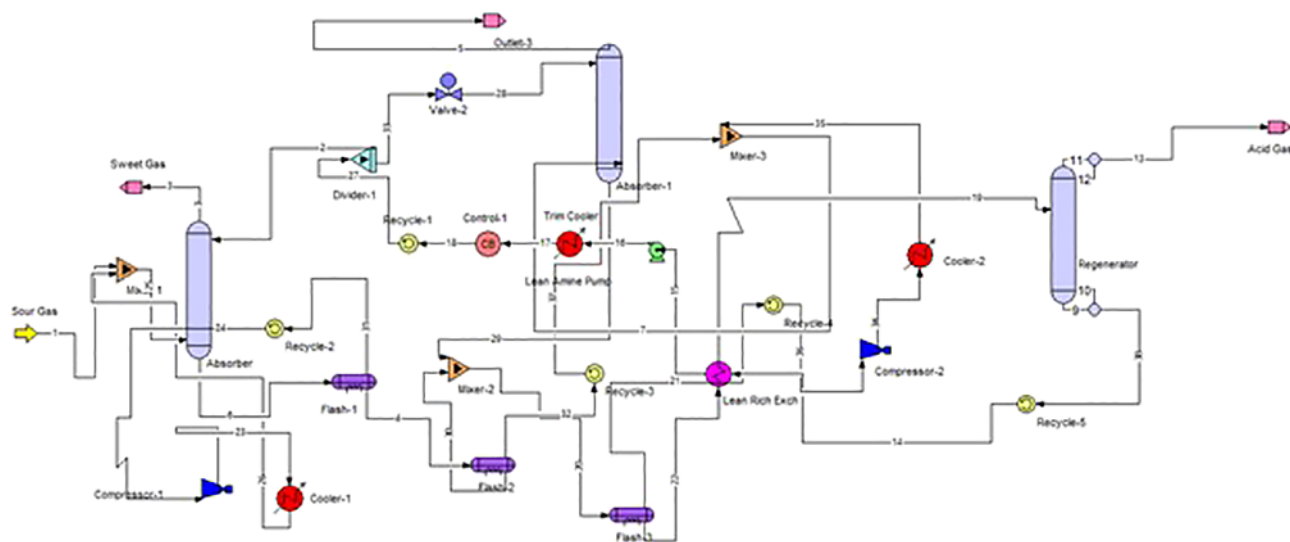


Figure 8. Process scheme of the physical absorption process. Simulation in ProTreat® v8.0.

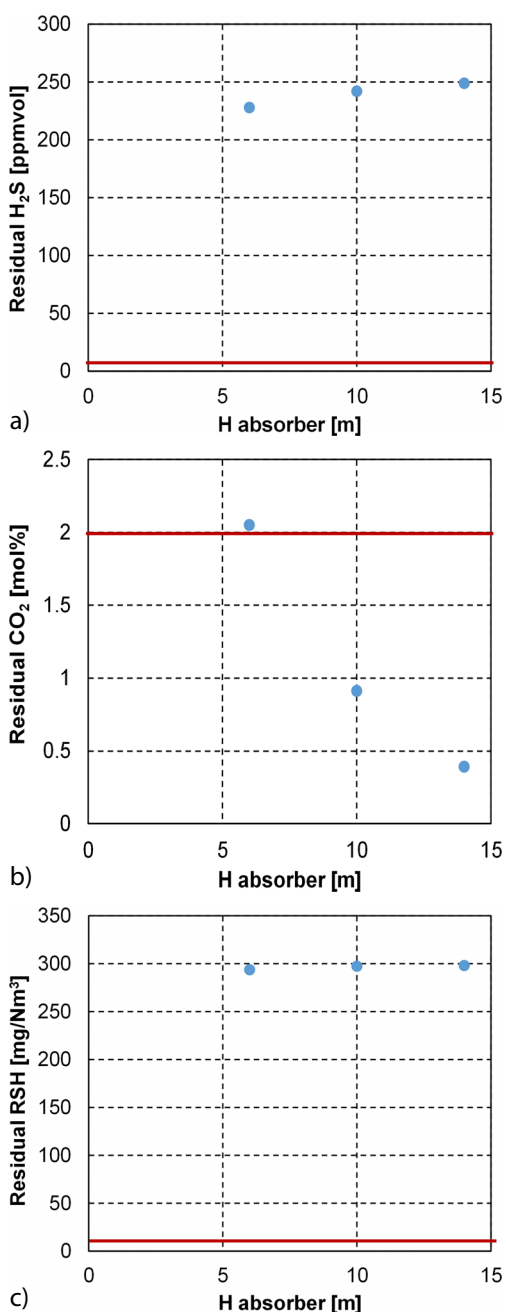


Figure 9. UCARSOL™ HS-101: residual impurities (a) H₂S, (b) CO₂, (c) RSH in the sweet natural gas as a function of the absorber height. The molar boilup ratio of the distillation column is fixed at 0.1, the solvent flow rate at 17 000 kg h⁻¹, and the stripper height at 8 m. Red lines stand for the admissible impurity content in the purified natural gas.

the absorption column packed height is fixed at 14 m. At increasing solvent flow rate, the H₂S and RSH residual content decreases. The controlling component for specifying the solvent flow rate, such that the outlet gas is suitable for gas grid distribution, is H₂S together with sulfur-based compounds. Then, the solvent flow rate has been further reduced acting on the molar boilup ratio of the regeneration unit to decrease the

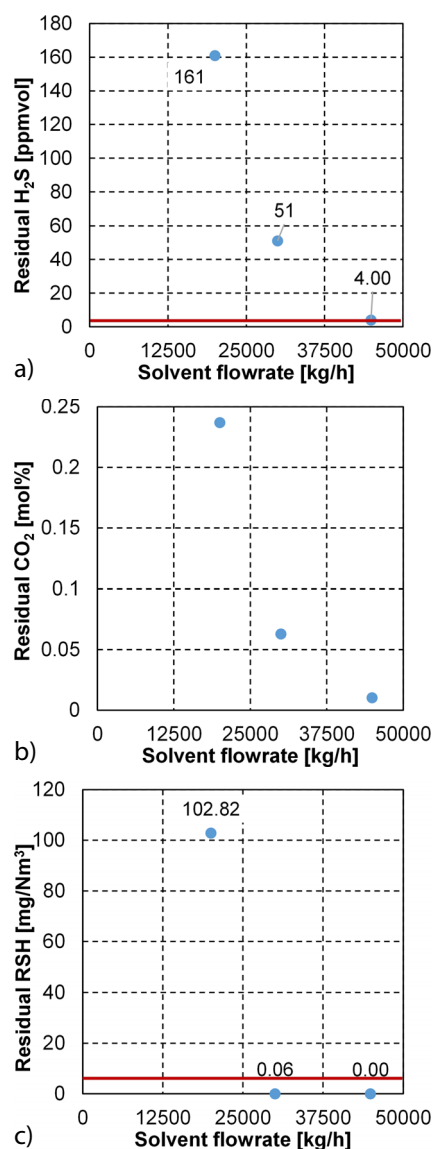


Figure 10. UCARSOL™ HS-101: residual impurities (a) H₂S, (b) CO₂, (c) RSH in the sweet natural gas as a function of the solvent flow rate. The molar boilup ratio of the distillation column is fixed at 0.1, the absorber height at 14 m, and the stripper height at 8 m. Red lines stand for the admissible impurity content in the purified natural gas.

reboiler duty. As a matter of fact, at higher molar boilup ratio, the solvent flow rate needed to accomplish the specified separation decreases because of the lower solvent lean loading. A trade-off between molar boilup ratio and circulating solvent flow rate can be established to guarantee low reboiler heat requirements.

The behavior is different for the DMEPG physical absorption, as demonstrated in Figs. 11 and 12. In Fig. 11, the solvent flow rate is fixed at 43 500 kg h⁻¹, while the absorber height is varied in the range of 6–14 m. As displayed in Fig. 11c, the mercaptans removal is extremely favored: their residual content is below the specification even at the lowest absorber packed

height. As stated in literature, physical solvents are particularly suitable for the sulfur-based compounds abatement. Also CO_2 residual content in the outlet gas fulfils the specification at increasing packing height (Fig. 11b), but this is not the case for H_2S . For this reason, the solvent flow rate has been increased. A huge amount of solvent is needed, much higher compared to the chemical absorption case, as indicated in Fig. 12. Similarly to what done for the chemical solvent, the molar boilup ratio has been varied to reduce as much as possible the reboiler duty.

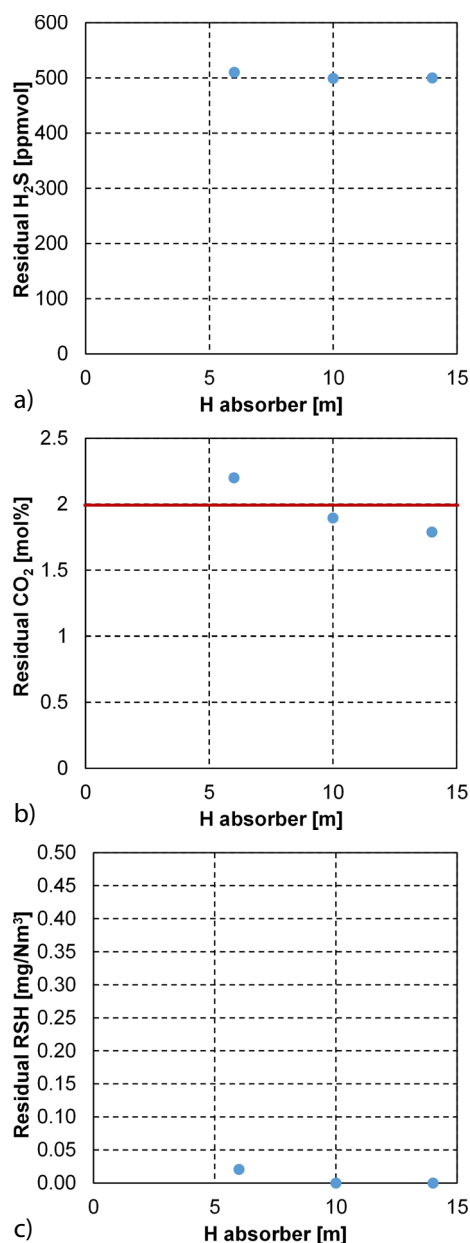


Figure 11. DMEPG: residual impurities (a) H_2S , (b) CO_2 , (c) RSH in the sweet natural gas as a function of the absorber height. The molar boilup ratio of the distillation column is fixed at 0.3, the solvent flow rate is at $43\,500\text{ kg h}^{-1}$, and the stripper height at 8 m. Red lines stand for the admissible impurity content in the purified natural gas.

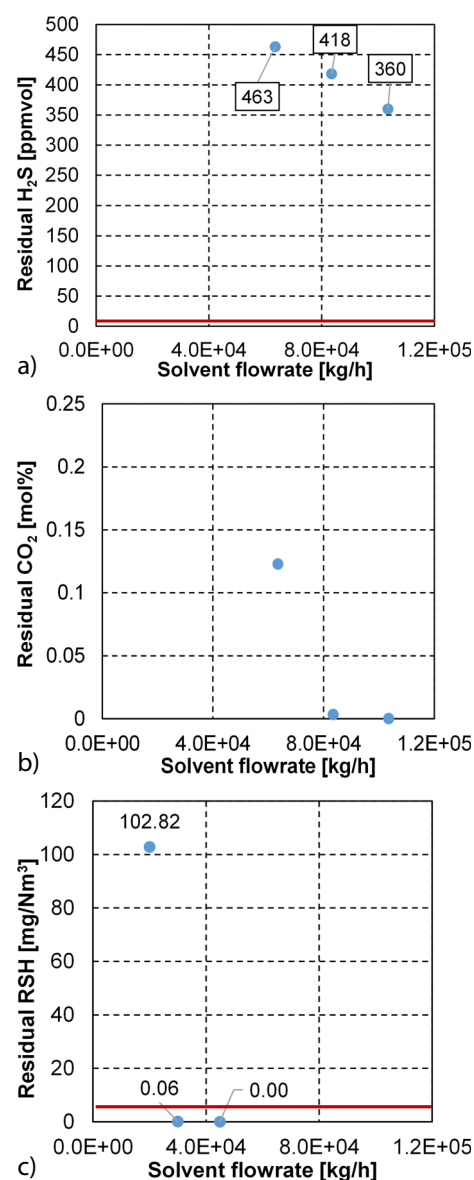


Figure 12. DMEPG: residual impurities (a) H_2S , (b) CO_2 , (c) RSH in the sweet natural gas as a function of the solvent flow rate. The molar boilup ratio of the distillation column is fixed at 0.3, the absorber height at 14 m, and the stripper height at 8 m. Red lines stand for the admissible impurity content in the purified natural gas.

As for the hybrid solvent, results of the process simulations are reported in Figs. 13 and 14. In this case, these outputs can be interpreted as the sum of the chemical and physical solvent behaviors. Mercaptans and CO_2 removal is highly performing in this case: in Figs. 13b and 13c, the CO_2 residual content is below the admissible limit, while the RSH concentration in the purified natural gas is close to the specification.

The solvent flow rate has been increased with respect to the base case of Fig. 8 to further reduce the H_2S concentration in the outlet gas (Fig. 14). When the solvent flow rate rises, the H_2S residual content in the purified natural gas decreases, as

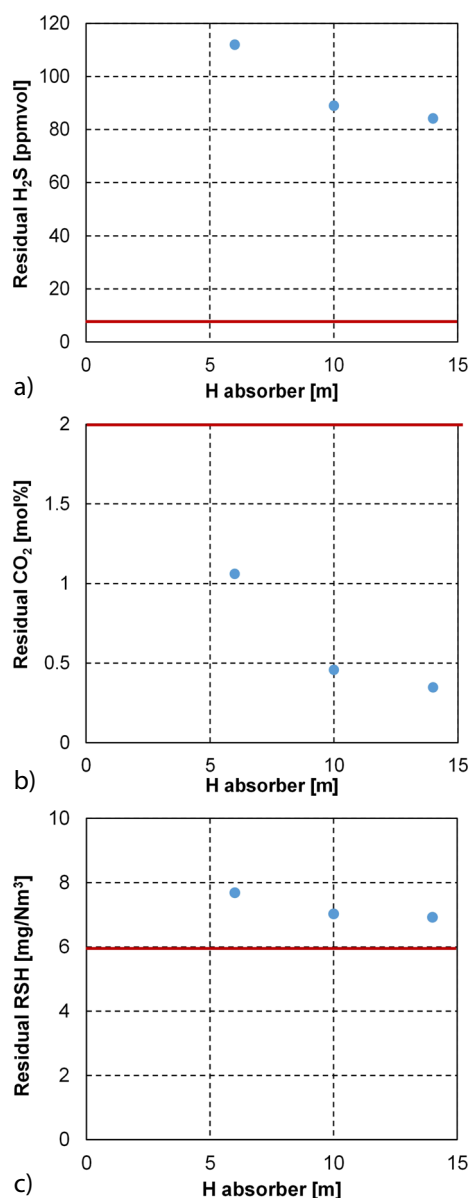


Figure 13. Sulfinol[®]-M: residual impurities (a) H₂S, (b) CO₂, (c) RSH in the sweet natural gas as a function of the absorber height. The molar boilup ratio of the distillation column is fixed at 0.15, the solvent flow rate is fixed at 17 000 kg h⁻¹, and the stripper height at 8 m. Red lines stand for the admissible impurity content in the purified natural gas.

indicated in Fig. 14a. As already observed for the chemical solvent, being H₂S a stronger acid than RSH, it is preferentially absorbed over mercaptans. Mercaptans removal, on the other hand, is highly sensitive to the overall solvent rich loading. For a solvent flow rate of 29 900 kg h⁻¹, all the specifications for the pipeline quality natural gas are met. Also in this case, acting on the boilup guarantees the reduction of the reboiler heat duty.

The dependence of reboiler duty on the molar boilup is reported in Fig. 15 for the sake of example, while in Tab. 7 the corresponding rich and lean loading is provided. As can be observed, as the molar boilup increases, both the H₂S and mer-

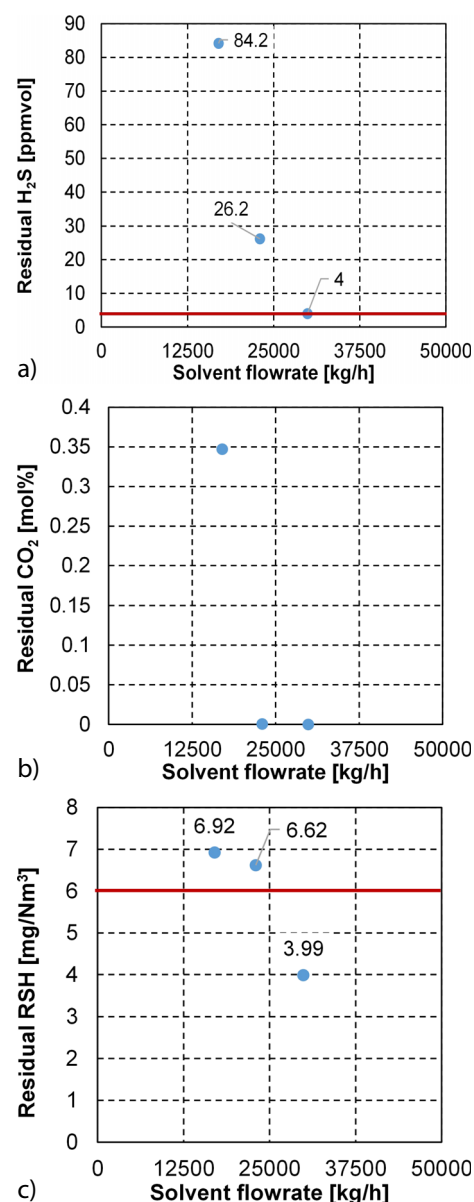


Figure 14. Sulfinol[®]-M: residual impurities (a) H₂S, (b) CO₂, (c) RSH in the sweet natural gas as a function of the solvent flow rate. The molar boilup ratio of the distillation column is fixed at 0.15, the absorber height at 14 m, and the stripper height at 8 m. Red lines stand for the admissible impurity content in the purified natural gas.

captans lean loadings decrease, such that the solvent is more deeply purified.

A comparison between the analyzed configurations is reported in Tab. 8. The physical solvent presents a better ability in the abatement of sulfur-based compounds, but a huge amount of circulating flow rate is needed for CO₂ and H₂S removal, with consequently higher reboiler duty.

On the other hand, the chemical solvent is not as performing as the physical one in separating mercaptans, but, as widely known, it is good for H₂S and CO₂ deep sweetening. The hybrid solvent can combine the advantages of both chemical

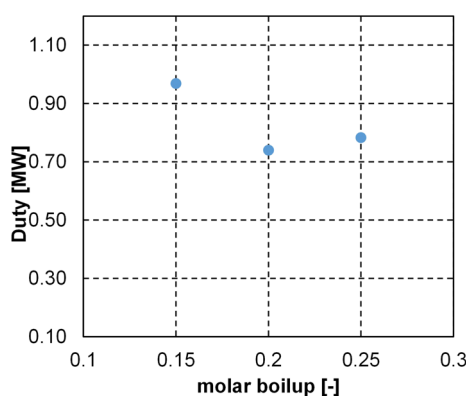


Figure 15. Sulfinol[®]-M: reboiler duty as a function of the regeneration column molar boilup.

Table 7. Rich and lean loading of the Sulfinol[®]-M solvent as a function of the molar boilup.

	Boilup		
	0.15	0.2	0.25
Rich loading [mol _{H₂S} /mol _{amine}]	0.359	0.618	0.713
Rich loading [mol _{CO₂} /mol _{amine}]	0.102	0.174	0.141
Rich loading [mol _{RSH} /mol _{amine}]	4.683×10^{-4}	7.561×10^{-4}	8.059×10^{-4}
Lean loading [mol _{H₂S} /mol _{amine}]	4.24×10^{-3}	3.709×10^{-3}	4.403×10^{-4}
Lean loading [mol _{CO₂} /mol _{amine}]	4.323×10^{-7}	1.328×10^{-6}	3.402×10^{-6}
Lean loading [mol _{RSH} /mol _{amine}]	7.079×10^{-5}	6.374×10^{-5}	4.530×10^{-7}

Table 8. Comparison between solvent performances in removal of acid gas and mercaptans.

	Base case: 40 wt % MDEA	UCARSOL [™] HS-101	DMEPG	Sulfinol [®] -M
Solvent flow rate [kg h ⁻¹]	36 500	23 545	41 900	17 270
Rich loading [mol _{H₂S} /mol _{amine}]	0.299	0.196	–	0.618
Rich loading [mol _{CO₂} /mol _{amine}]	8.292×10^{-2}	5.402×10^{-2}	–	0.174
Rich loading [mol _{RSH} /mol _{amine}]	4.668×10^{-4}	2.139×10^{-4}	–	7.561×10^{-4}
Lean loading [mol _{H₂S} /mol _{amine}]	8.081×10^{-3}	6.789×10^{-3}	–	3.709×10^{-3}
Lean loading [mol _{CO₂} /mol _{amine}]	1.090×10^{-5}	5.470×10^{-6}	–	1.328×10^{-6}
Lean loading [mol _{RSH} /mol _{amine}]	0	6.310×10^{-12}	–	6.374×10^{-5}
H ₂ S [ppm vol]	4	1.628	0.245	4
CO ₂ [mol%]	0.023	0.1	2	0.055
RSH [mg Nm ⁻³]	0.003	6	0	2.329
Reboiler duty [MW]	1.512	1.282	2.045	0.7396

and physical solvent, becoming the preferable alternative when a deep acid gas and sulfur-based compounds removal is required. For these absorption-based processes, operating expenses (OPEX) minimization is critical and is commonly regarded as the reference for cost minimization. Operating expenses are mainly dependent on reboiler duty and its operating temperature. For this reason, the results provided can be reasonably assumed as index of the operating costs.

5 Conclusions

The recent announcements of ultra-sour gas field exploitation worldwide impose the need of viable solutions for their monetization. The removal of H₂S and CO₂ contaminants requires proper technologies for achieving cost-effective processes. Besides the economic issue related to their high treatment cost, environmental concerns of toxic emissions together with the recent sulfur market constraints make their processing even more challenging. For this reason, this work reviews the state-of-the-art for ultra-sour gas management to allow a primary screening of the available technologies. Depending on the inlet acid gas content, the relative H₂S/CO₂ ratio, the sulfur-based compounds concentration, and the processed volumes, the most convenient alternative can be identified for the removal of both acid gases.

Process simulations are provided considering the Kashagan gas composition to compare the performance of hybrid, physical, and chemical solvents on mercaptans removal, together with H₂S and CO₂ abatement.

The hybrid solvent selected is the most effective in accomplishing the gas purification for this specific case study, allowing to reach the sale gas specifications with the lowest duty requirement.

Conflicts of Interest

The authors declare no conflict of interest.



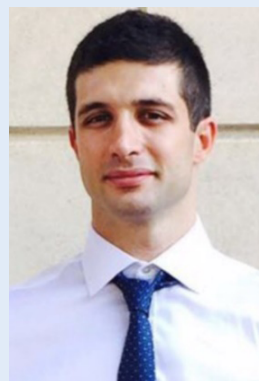
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separations in bioprocesses. The focus is both on the process simulation, with particular attention to energy saving, and on the thermodynamic characterization of the systems.



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Abbreviations

CFZ™	Controlled Freeze Zone™
DCCD™	Dual Colum Cryogenic Distillation™
DEA	diethanol amine
DGA	diglycol amine
DIPA	diisopropyl amine
DMEPG	dimethyl ether polyethylene glycol
IFP	Institut Français du Pétrole
IFS	Integrated Flow Solutions, Inc.
JPF	Jurassic Production Facilities
MDEA	methyl diethanol amine

MEA	methyl ethanol amine
MTR	Membrane Technology and Research, Inc.
OPEX	operating expenses
RSH	mercaptans
WK	West Kuwait

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