

Acid Gas Removal from Natural Gas by Water Washing

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Projections in the future energy scenario outline an important role played by fossil fuels to meet the increasing global energy demand. A "golden age" has been recently outlined for natural gas, in particular, as the fastest growing and the cleanest of all fossil fuels. Although natural gas is mostly considered to be a "clean" fuel with respect to the emission of pollutants from its combustion, the raw natural gas found in reservoir deposits is not free of contaminants. Among the others, hydrogen sulphide and carbon dioxide are two undesired compounds, which are responsible for the sour or acidic nature of natural gas and must be removed for operational and safety reasons. Acid gas treating is typically performed in facilities built at surface locations, mainly by means of chemical absorption into aqueous amine solutions. However, subsurface technologies may allow to possibly separate the gas undesired compounds directly downhole. The high pressure encountered in this environment makes the use of water as liquid absorbent worth considering. This work investigates the possibility of acid gas removal from natural gas by downhole water washing and presents a preliminary evaluation of the performances of the process, which is assumed to be carried out in the gas production casing that can be represented as a bubble column. A previously proposed correlation for the gas holdup in this type of contacting device operated counter-currently has been used to determine the volumetric mass transfer coefficient for design purposes, considering different raw gas flow rates and inlet acid gas concentrations. By solving a simplified model of a bubble column and by using water flow rates compatible with reinjection into the reservoir, it has been found that it is possible to reduce the H₂S content from the inlet concentration to the commonly accepted value to meet pipeline specifications and, depending on the inlet CO₂ concentration, to perform a bulk removal of it.

1. Introduction

Conventional natural gas exists in nature under pressure in deep reservoirs. After the raw natural gas is brought to the surface, it must be further processed to be suitable for pipeline transportation or commercial use. Among the other constituents which can be found in the raw natural gas, carbon dioxide (CO₂) and hydrogen sulphide (H₂S) must be taken into account, since they require complete or partial removal. These compounds are collectively known as "acid gases" and they are very undesirable, as they cause corrosion and present major safety risks. Because of the toxicity of H₂S and the lack of heating value of CO₂, natural gas prepared for sales is required to contain no more than 5 ppm of H₂S and 2-3 mol% maximum of CO₂ (Mokhatab et al., 2006). The actual specifications are dictated by the use and by the country where the gas is employed.

Since natural gas has a wide range of composition, including the concentration of the two acid gases, a number of processes have been proposed and practiced on a commercial scale to cope with their removal. They can be categorized as: absorption (chemical, physical and hybrid), adsorption, membrane permeation, low-temperature separation. Decisions in selecting a gas treating process can be taken considering gas composition and operating conditions. For instance, physical solvent processes are generally most efficient when the partial pressures of the acid gases are relatively high, because the solubility of a compound in a solvent is directly proportional to its partial pressure in the gas phase, which is the driving force for the absorption. Energy requirements for regeneration of the solvent are lower than in systems which involve

chemical reactions, since most of the solvent can be regenerated by a simple pressure letdown (Speight, 2007).

Historically, methanol was the first commercial organic physical solvent and it was used in the development of the Rectisol[®] process (Mokhatab et al., 2006). Water is also used as an absorbent for gas impurities since it offers some advantages: it is available at low cost, it can be used in simple scrubbing units with less concern over leakage than for organic solvents and it can be used on a once-through basis (Kohl and Nielsen, 1997). Moreover, water-wash does not require heat loads, even if very high pumping loads are needed. Water may also be applicable to the washing of high-pressure gases, where the high partial pressure of the impurity, which is only sparingly soluble at low pressure (e.g., CO₂), brings its solubility up to a level high enough to make the process economically viable. As far as CO₂ is concerned, the absorption in water at elevated pressure was formerly an important industrial process (particularly for the purification of synthesis gas for ammonia production). However, the process has now been replaced by more efficient systems, which employ chemical or physical solvents having a higher capacity for CO₂ than water (Kohl and Nielsen, 1997). Water scrubbing has been also proposed for the removal of CO₂ from methane produced by anaerobic digestion (i.e., from biogas, which can be conceived as a peculiar natural gas with a high CO₂ content). Water scrubbing is considered attractive for this type of application because of its relatively low capital cost in small sizes, simplicity of operation and maintenance and use of a readily available nonhazardous absorbent (Kohl and Nielsen, 1997), even if biogas upgrading by water scrubbing typically involves larger electricity consumptions in comparison with chemical absorption by amine solutions (Pellegrini et al., 2015). As far as H₂S is concerned, several attempts have been made to commercialize processes based on the use of water for removing this impurity from gas streams and, especially, for treating natural gases of very high H₂S content (Kohl and Nielsen, 1997). Despite that, the process has not become commercially important since the savings with respect to conventionally used alkanolamine processes resulting from the reduced energy requirements have turned out to be not as high as expected because of the large amounts of steam generated in the conversion of H₂S to elemental sulfur in downstream Claus type units, which can be used to satisfy the heat requirements for solvent regeneration in an amine plant. In spite of these considerations, the absorption of H₂S in water may be economical in special cases and it has been demonstrated to be a technologically feasible operation (Kohl and Nielsen, 1997).

This work contributes to the existing discussion on acid gas removal technologies and, in particular, it deals with a preliminary evaluation of the performances of the water washing process applied to the removal of H₂S or CO₂ from a CH₄-rich stream, considering high pressures which enhance the solubility of the acid gases in water and are typically encountered in downhole environments. Some patents (Kjos, 1999; Koelmel et al., 2002; Thomas et al., 2003; Verma and Ramakrishnan, 2009) can be found in the literature concerning acid gas removal processes applied to natural gas directly in the well hole, which mainly involve the use of membranes. Contrarily, the absorption system considered in this work can be schematized as a bubble column having geometric features as close as possible to the real gas production casing. In particular, a large-diameter (inner diameter, $d_c = 0.24$ m) vertical pipe has been taken into account, for which the gas-liquid flow has been experimentally studied using gas and liquid velocities corresponding to gas and liquid flow rates in line with typical gas throughputs and with liquid flow rates compatible with reinjection into the reservoir and with the establishment of a counter-current flow.

2. Methods

The following two sections describe how the system under investigation has been modelled and explain the criteria that have been taken into account to select the most appropriate operating conditions.

2.1 Modelling approach

The absorption system considered in this work has been modelled as a bubble-type water scrubber (i.e., a bubble column) and, thus, it can be considered as a continuous-contact equipment, with the fluids that come into contact continuously in their path through the tower, rather than intermittently, as it occurs in plate towers (Foust et al., 1967). Bubble columns offer many advantages, including simplicity of construction (Besagni et al., 2017a). The system under study has been modelled considering that only one component (H₂S or CO₂) is transferred from the gas to the liquid phase. Methods for estimating the height of the active section, H_a , of counter-current flow differential contactors are typically based on rate expressions representing mass transfer at a point on the gas-liquid interface and on material balances representing the changes in bulk composition in the two phases that flow past each other. The rate expression can be obtained referring to a differential section, by equating the rate at which the solute is absorbed by the solvent to the rate at which it is transferred from the interface through the liquid phase. This leads to Eq(1) for the height of the active section of the column (Perry and Green, 2008).

$$H_i = \frac{L}{c_i} \int_{c_i}^1 \frac{(1+X_i)(1+X)}{k_L a (X_i - X)} dX \quad (1)$$

In Eq(1), L denotes the molar flow rate of the inert-liquid component, c_i stands for the total concentration, X denotes the mole ratio in the liquid phase, and subscript i denotes the interface between the two phases. Eq(1) suggests that H_i is inversely proportional to the volumetric mass transfer coefficient, $k_L a$, which has been evaluated in this work according to the relation found in the work by Seno et al. (Seno et al., 1990). These authors have studied the hydrodynamics and mass transfer characteristics in counter-current, co-current and liquid batch operation in a bubble column, and they have concluded that the experimental correlation of Akita and Yoshida (Akita and Yoshida, 1973), Eq(2), could be applied not only to the liquid batch but also to the counter-current operation.

$$\frac{k_L a \cdot d_c^2}{D_L} = 0.6 \left(\frac{\mu_L}{\rho_L D_L} \right)^{0.5} \left(\frac{g D^2 \rho_L}{\sigma} \right)^{0.62} \left(\frac{g D^3 \rho_L^2}{\mu_L^2} \right)^{0.31} \varepsilon_G^{1.1} \quad (2)$$

Eq(2) gives the dependence of the volumetric mass transfer coefficient on the gas holdup, ε_G , which has been computed according to the relation proposed by De Guido et al. (De Guido et al., 2016). This correlation has been obtained using the data (Besagni et al., 2017b) collected for a large-diameter and large-scale bubble column (Besagni et al., 2016, 2017a) to also account for the effect of a counter-current liquid flow ($U_L < 0$) (Besagni and Inzoli, 2016), which is less investigated in the literature and mainly in small-diameter columns. The gas hold-up correlation allows to relate the gas hold-up measured with no liquid flow (batch operation) to that measured in the presence of a counter-current liquid flow. Both the relations for the gas holdup and the volumetric mass transfer coefficient (Eq(2)) have been proven to hold at atmospheric pressure. However, it is reported in the literature (Krishna et al., 2000; Wilkinson et al., 1994) that these parameters increase with pressure. Thus, as for the description of the hydrodynamics of the column, the most conservative choice has been made. In Eq(2) the physical properties of water (viscosity, density and surface tension) have been taken from the literature (Kestin et al., 1978; Perry and Green, 2008; Vargaftik et al., 1983): $\mu_L = 719.5 \text{e-}6 \text{ Pa}\cdot\text{s}$, $\rho_L = 994.032 \text{ kg/m}^3$, $\sigma = 70.41 \text{e-}3 \text{ N/m}$. The diffusion coefficients for H_2S and CO_2 in pure water have been taken from Tamimi et al. (Tamimi et al., 1994). In this work, the Henry's law has been used to express the equilibrium relation: the Henry's law coefficients have been taken from Perry and Green (Perry and Green, 2008) and from Sander (Sander, 1999), respectively for dissolved H_2S in water and CO_2 in water. Concentrations for H_2S of 5, 10, and 15 mol% in the inlet gas have been taken into account, since they are typical H_2S concentrations that can be found in many natural gases, as reported by Katz (Katz, 1959). The same values have been also considered for CO_2 for comparison purposes. Moreover, a value of 0.0004 mol% and of 2 mol% has been considered as the desired mole fraction, respectively, of H_2S and of CO_2 in the outlet gas, assuming that the investigated process aims at bringing their content to the value commonly accepted for transport via pipelines. The properties of the gas phase (Table 1) have been kept constant along the column and equal to those of the inlet gas mixture, which have been taken from REFPROP V9.0 (Lemmon et al., 2010).

2.2 Operating conditions

Since both H_2S and CO_2 are hydrate formers and they can also form a second liquid phase in addition to the H_2O -rich liquid phase under certain temperature and pressure conditions, some considerations have been made to select proper operating conditions such to avoid the formation of hydrates and of a second liquid phase, which would cause a change to flow phenomena occurring within the column with respect to two-phase flow phenomena that establish in the presence of only the gas and H_2O -rich liquid phase.

Table 1: Properties of the gas phase ($\text{CH}_4+\text{H}_2\text{S}$ or CH_4+CO_2 , respectively for H_2S removal and CO_2 removal) taken from REFPROP V9.0 (Lemmon et al., 2010).

y_o [%]	P [MPa]	H ₂ S Removal		CO ₂ Removal	
		ρ_G [kg/m ³]	$\mu_G \cdot 10^6$ [Pa·s]	ρ_G [kg/m ³]	$\mu_G \cdot 10^6$ [Pa·s]
5	15	120.89	16.65	122.92	16.707
10		131.35	17.224	135.03	17.36
15		143.06	17.835	147.83	18.036
5	20	160.87	19.574	163.34	19.525
10		175.31	20.505	179.75	20.411
15		191.43	21.54	197.16	21.359

As for the removal of CO₂ from methane, the pressure-temperature diagram for the system CH₄-CO₂-H₂O illustrated in Figure 1 has been taken into account, which shows the vapour-liquid-liquid equilibrium (VLLE) region defined with the data reported by Al-Ghafri (Al Ghafri, 2014). This region is bounded by the quadruple points (at which the four phases hydrate (H), H₂O-rich liquid (L_w), CO₂-rich liquid (L_c) and vapour (V) coexist) and the upper critical end point (UCEP) data reported by the author and by the CO₂ saturated vapour pressure curve, which has been computed using the correlation reported by Perry and Green (Perry and Green, 2008). All the other lines (*i.e.*, the L_w-H-V three phase equilibrium line for CH₄ and CO₂, and the L_w-H-L_c three phase line for CO₂) have been drawn using the correlations reported by Carroll (Carroll, 2009). Based on this diagram, a temperature higher than *ca.* 290.15 K may be reasonable for avoiding hydrate formation and, at these temperatures, pressures higher than *ca.* 8 MPa are a good choice for avoiding the formation of a second CO₂-rich liquid phase in equilibrium with a H₂O-rich liquid phase and with the vapour phase. As for the system CH₄-H₂S-H₂O, which has been less investigated than the one with CO₂, the results reported by De Guido (De Guido, 2016) have been taken into account, which suggest that the temperature should be higher than *ca.* 308.15 K to avoid hydrate formation and that, at these temperatures, pressures should be higher than about 15 MPa to avoid the existence of a second H₂S-rich liquid phase. For all these reasons, calculations have been performed at 308.15 K and at pressures of 15 and 20 MPa.

3. Results and discussion

In order to assess the feasibility of the process, the minimum liquid rate, L_{min} , corresponding to an infinite column height, should be calculated to get an idea of the amount of water needed to achieve the desired purification level and to check for its compatibility with hydrodynamic constraints. L_{min} can be calculated from the composition of the entering gas and the solubility of the solute in the exit solvent, saturation being assumed. The actual liquid-to-gas ratio (L/G) has been then determined by multiplying the minimum value by 1.5, following the general rule according to which it is normally greater than the minimum by as much as 25 to 100 percent (Perry and Green, 2008).

Different gas flow rates have been considered to understand which type of gas reservoirs may be considered for the application of the investigated process. Indeed, for high gas flow rates the amount of solvent to be used to meet the desired specification on the outlet gas may be too high and not suitable to ensure a counter-current flow and with reinjection purposes.

The results reported in Table 2 (for a gas throughput of 30,000 Nm³/day) confirm that, as pressure increases, keeping the same inlet conditions, a lower amount of solvent is required to reach the desired specification. At a given pressure, the minimum liquid-to-gas ratio, $(L/G)_{min}$, increases as the inlet acid gas content increases, as expected. However, the trend for the minimum solvent flow rate with respect to the acid gas mole fraction in the inlet gas, y_o , is different for the two cases: as for the removal of H₂S, L_{min} decreases as y_o increases, whereas it increases when the removal of CO₂ is considered.

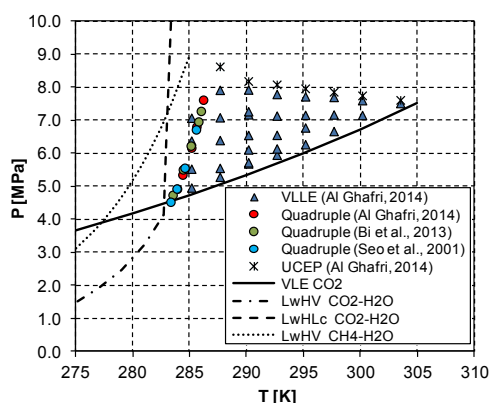


Figure 1: Pressure-temperature diagram for the system CH₄-CO₂-H₂O showing the VLLE region and its boundaries. Symbols represent: ▲, VLE data reported by Al Ghafri (Al Ghafri, 2014); ●, quadruple data reported by Al Ghafri (Al Ghafri, 2014); ●, quadruple data reported by Bi et al. (Bi et al., 2013); ●, quadruple data reported by Seo et al. (Seo et al., 2001); *, UCEP data reported by Al Ghafri (Al Ghafri, 2014). The solid line represents CO₂ saturated vapour pressure (Perry and Green, 2008). The following lines calculated by Carroll's correlation (Carroll, 2009) represent: ···, three phase region L_w-H-V for pure CH₄; ---, three phase region L_w-H-V for pure CO₂; - - -, three phase region L_w-H-L_c for pure CO₂.

Table 2: Results at 308.15 K for a gas flow rate of 30,000 Nm³/day (0.0004 mol% and 2 mol% specified for the H₂S and for the CO₂ mole fraction in the outlet gas).

y_o [%]	P [MPa]	H ₂ S Removal				CO ₂ Removal			
		$(L/G)_{min}$ [mol/s]	L_{min} [mol/s]	L [mol/s]	H_t [m]	$(L/G)_{min}$ [mol/s]	L_{min} [mol/s]	L [mol/s]	H_t [m]
5	15	4.75	69.96	104.94	13.60	5.44	80.08	120.11	1.86
10		4.96	69.19	103.78	13.65	7.61	106.14	159.20	3.30
15		5.20	68.41	102.62	13.22	8.68	114.30	171.44	4.11
5	20	3.55	52.28	78.41	12.14	4.07	59.94	89.91	1.65
10		3.69	51.50	77.25	12.24	5.69	79.29	118.93	2.95
15		3.85	50.73	76.09	11.90	6.47	85.21	127.81	3.69

This is due to the fact that computations have been carried out at constant total gas flow rate and, thus, the amount of inert-gas component (G) decreases as the acid gas mole fraction in the inlet gas increases from 5 to 15 mol%. The decrease in G at increasing y_o prevails on the increase of $(L/G)_{min}$ when CO₂ is the acid gas to be removed, whereas the opposite occurs when the undesired acid gas is H₂S. In Table 2 the column height is also reported: the values obtained for the removal of H₂S are higher than those obtained in the case of CO₂ removal because of the much lower acid gas concentration that has been required to be reached in the outlet gas stream. If a lower active section is actually available, a possible process solution may also consist in performing a downhole bulk removal of the undesired acid gas that may be followed by a finishing treatment. The fact that the results obtained for the column height for the cases related to the removal of H₂S at the same pressure for different y_o are very similar can be explained considering that the operating lines are very close to each other (due to the low specified H₂S content in the outlet gas). Considering a higher gas flow rate of 50,000 Nm³/day, the removal of H₂S or of CO₂ from methane by water washing at high pressure requires higher column heights to achieve the same gas outlet conditions. However, when CO₂ is the acid gas to be removed, especially as its inlet concentration in raw natural gas increases, high liquid flow rates are necessary, which may be not compatible with reinjection into the reservoir and may avoid the establishment of a counter-current two-phase flow (in this work, the maximum acceptable liquid flow rate has been considered to be 200 mol/s, given typical natural gas flow rates and the dimension of the equipment - a bubble column having $d_c = 0.24$ m- where absorption takes place). In such cases, a bulk removal may be performed, which allows to make use of a lower solvent rate to achieve the less strict CO₂ content in the gas outlet stream.

4. Conclusions

In this work, the possibility of removing acid gases (either H₂S or CO₂) from a natural gas stream has been investigated by means of water washing at high pressure. This type of process is not much studied in the literature since other purification technologies, such as amine scrubbing, are typically preferred to it. However, when considering the possibility of performing gas treating directly downhole so to avoid building surface facilities, the use of chemicals may be not appropriate and water can be used instead, exploiting the high pressures encountered in downhole environments, which enhance the acid gas solubility into it. The operating conditions in this case would be different from the typical ones at which acid gas removal is currently performed, suggesting some considerations for assessing the feasibility of the process. In this work, the operating temperatures and pressures have been defined considering the thermodynamic behaviour of the two systems of interest, namely CH₄-CO₂-H₂O and CH₄-H₂S-H₂O. With the aid of the pressure-temperature diagram showing the VLLE region and lines of equilibrium conditions involving the hydrate phase for these systems, a temperature of 308.15 K and pressures of 15 and 20 MPa have been chosen to avoid some problems like the formation of hydrates and of a second non-aqueous liquid phase. In the end, a preliminary evaluation of the feasibility of the process has been made considering it to be carried out in a bubble column, which can resemble the gas production casing. A previously proposed correlation for the gas holdup in this type of contacting device operated counter-currently has been used to determine the volumetric mass transfer coefficient for design purposes, considering different raw gas flow rates and inlet acid gas concentrations. It has been found that it is possible to reduce the H₂S content from the inlet concentration to the commonly accepted value to meet pipeline specifications using liquid flow rates compatible with reinjection into the reservoir, even if a high column height is necessary due to the very low H₂S content required in the outlet gas stream. The removal of CO₂ is also possible, although at high gas throughputs the liquid flow rate required to reduce its content to the desired value may be not compatible with the one that can be reinjected into the reservoir. Thus, depending on the inlet CO₂ concentration, a possible alternative may consist in using the same process for performing only a bulk removal of it.

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