

Condensed rotational separation

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Condensed Rotational Separation

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Abstract

Condensed Rotational Separation is based on partial condensation of components of gas-gas mixtures. Condensation is induced by flash evaporation or pressure distillation. The rotational particle separator removes the micron sized particles formed by condensation from the gas. Yields and purities are enhanced by adding a next stage of liquid flash and relooping the gas to the first stage. A great improvement in separation performance, in both yields and purities, is achieved by allowance for operation of CRS within the vapour-liquid-solid region. Condensed rotational separation is shown to be an economically attractive process for upgrading sour gas fields contaminated with CO₂ and/or H₂S.

1 Introduction

Separation of gas mixtures is an issue in various energy related processes. Examples are the separation of H₂S and CO₂ from methane to upgrade sour gas fields and the capture of CO₂ from coal fired power plants to reduce the emissions of greenhouse gases. The mature industrial method for these applications has been the amine absorption technology. The contaminated gas is bubbled through solvent in an absorber column where the solvent absorbs the contaminant. Afterwards the solvent is regenerated by heating and/or flashing to a lower pressure. The process becomes increasingly uneconomical at higher contamination levels due to the intensive energy requirements and large equipment size [1]. An alternative method is to separate on the basis of phase separation. The mixture is cooled to semi-cryogenic temperatures, whereby the contaminant condenses partially and after which the condensate is separated. Several processes have been developed on this principle, for example the Total Sorex process to remove H₂S from natural gas [2], Cryocell by CoolEnergy to remove CO₂ from natural gas [3], Controlled Freeze Zone by Exxon for sour gas fields [4] and the Alstom anti-sublimation process [5]. The energy consumption involved with these processes is limited and makes them useful for high contaminant levels. Drawback is the large size of the installations and capital costs involved.

Instead of fractional distillation employing temperature differences, one can separate by flash evaporation or pressure distillation. The advantage is simplicity and short residence time, i.e. small equipment and limited investment costs. A technological challenge is however posed by the feature that rapid cooling of binary or multi component mixtures of gases to temperatures where one, or some of the components preferentially condense, leads to a mist of very small droplets with diameters of 1 to 10 micron [6]. The phenomenon is known to occur by aerosol formation in flue gases of biomass combustion installations [7], condensate droplets resulting from cooling of wet natural gas [8] and has also been measured in experiments with CH₄/CO₂ mixtures [9]. As the micron sized droplets are difficult to separate from the gas stream [10], the size advantage of pressure distillation is often lost in the required separator. For a process which relies on fast phase change as a means

of separation to be economical and practical, it is necessary to have a device capable of capturing micron-sized droplets with high collection efficiency at low pressure drop and a small footprint.

Cyclones are standard for liquid/gas separation in hydrocarbon processing plants [11, 12]. These cyclones are used for water and condensate removal but have not been applied for removing condensed contaminants, such as CO_2 or H_2S . This is because cyclones can only handle condensing droplet sizes above $15\ \mu\text{m}$ [13–15]. To achieve such droplet sizes requires unrealistically large extended highly cooled droplet growth pipes. It is well known in laboratory chemical applications that microcyclones can separate micron-sized droplets having diameters as small as one micron, but then the flow is very small and orders of magnitude less than the flow in gas well applications. Alternatively one can improve separation efficiency by increasing swirl velocity to supersonic velocity but at the costs of large pressure drops [16].

A more feasible solution is provided by the rotational particle separator, abbreviated by RPS [34]. The RPS consists of a cylindrical pipe wherein a rotating element is placed. The rotating element is a simple rotating body consisting of a very large number of axial channels of a few millimeters in diameter. In such channel the micron-sized droplets are centrifuged to form a liquid film at the channel-wall, which is ripped off at the exit of the channel in the form of droplets; typically $20\ \mu\text{m}$ or larger. These droplets are separated according the working principles of ordinary axial cyclones. The rotating element can receive its momentum for rotation by pre-rotation of the gas entering the rotating element, and/or by external drive through an electrical motor which is indirectly connected through a magnetic field.

In the CRS process, the RPS is used to separate the micron sized droplets. The contaminated gas is rapidly lowered in temperature ($-50/-100^\circ\text{C}$) and reduced in pressure (30 to 40 bar). A mixture forms which consists of predominantly the gaseous phase with a mist of small micron-sized droplets of liquid/solid contaminant. The separation process is further enhanced by introducing a second step. The collected liquid/solid slurry is flashed to 10 to a liquid/gas mixture at 20 bar. The liquid is purified, while most of the gas which was dissolved in the liquid/solid slurry evaporates. This gas is re-fed into the gas stream in the first part of the process.

In this paper we shall address the important aspects of CRS. In section 2 the CRS process is compared to fractional distillation on thermodynamic principles. In section 3 the design considerations are applied to a fractionation scheme for sour gas fields. Case studies are presented for contaminated gas fields. Other potential applications of the CRS technology are shortly discussed in section 4, conclusions are presented in section 5.

2 Process thermodynamics

In Fig.1 the phase diagram of the $\text{CH}_4\text{-CO}_2$ system is shown. The solid-phase boundaries are shown by the dash-dotted lines. These lines represents the limit of CRS in the binary system, as below and right of these lines there exists only vapor and pure solid CO_2 or liquid and pure solid CO_2 . Also depicted in Fig.1 is the principle of condensed rotational separation. The incoming mixture with methane concentration X_{in} is chilled by a combination of cooling and expansion to a point close to the solid boundary of the vapour-liquid phase (within 5°C), where the recovery of methane is maximal [17]. This point is indicated by A in Fig.1. Because of binary condensation a mist of small droplets is formed with composition X_{Al} . The vapour phase has a composition X_{Av} and is separated. The droplets are separated by the RPS to obtain a liquid stream. This liquid stream is subsequently heated and expanded to obtain liquid droplets with composition X_{Bl} and a gas with composition X_{Bv} , at a point (p, T) even closer to the solids boundary. The resulting gas has a composition that is not far from the original untreated feed gas and is re-fed to the original gas entering the installation. In case of the $\text{CH}_4\text{-CO}_2$ system, CRS has the potential to reach a purity of about 85% on the methane side and 95% on the CO_2 side. This compares to fractional distillation at

40 bar between the temperatures T_1 and T_2 as is shown in Fig.2.

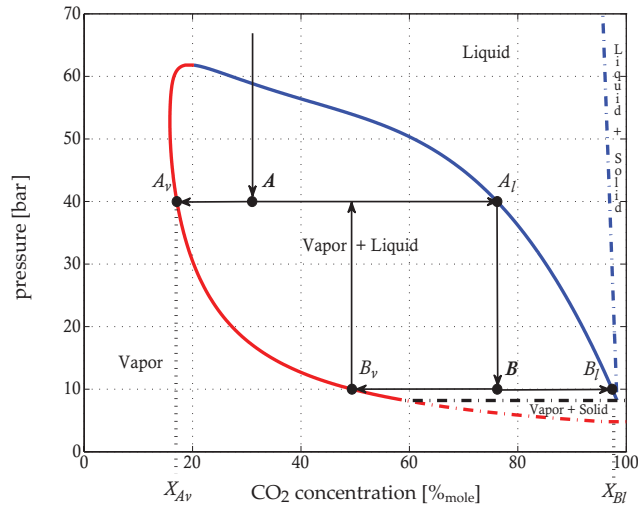


figure 1: $P - X$ diagram for the $\text{CH}_4\text{-CO}_2$ system. The incoming gas mixture is cooled and expanded to point A in the phase diagram where it is separated in a gas stream with composition X_{A_v} and a liquid stream with composition X_{A_l} . In the second flash the liquid is separated into a liquid with composition X_{B_l} and a gas X_{B_v} . This gas is mixed with the incoming gas on line $A_v - A_l$.

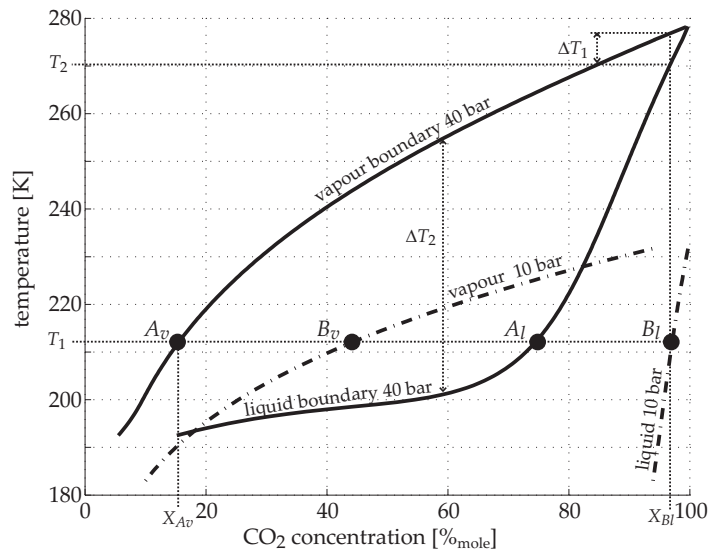


figure 2: $T - X$ diagram for the $\text{CH}_4\text{-CO}_2$ system.

In reality raw natural gas is not a binary mixture, but contains traces of N_2 , C_2H_6 and H_2 . The presence of H_2S in a $\text{CO}_2\text{-CH}_4$ system lowers the temperature at which solids are formed. In that case significantly higher purities of methane in the gas phase can be reached while operating in the vapor-liquid regime: see also section 3. There is however a more important benefit of ternary or

multi-component mixtures, like the system CH₄-CO₂-H₂S. At temperatures below the solid boundary, multi-component mixtures have a vapour-liquid-solid region instead of a vapour-solid region. This makes operation of CRS possible in this region, as long as the solid fraction does not become dominant. By operating CRS in the VLS-region, much higher purities of methane on the gas phase can be achieved, compared to operation of CRS in the vapour-liquid region. This is illustrated in section 3, for the light contaminated Arabiyah and Hasbah gas fields in Saudi-Arabia.

2.1 Energy considerations

The lost work by cryogenic separation can be calculated as follows [18]: condensation starts at the temperature where the partial pressure of the condensing species in the feed mixture reaches the saturation pressure. This temperature is below the bubble point line of the separated liquid. If we take an arbitrary control volume where the vapour and non-condensable gas temperatures are the same in the in- and the outlet: $T_{v1} = T_{g1}$ and $T_{v2} = T_{g2}$, the difference between the inlet enthalpy of the vapour and the condensate enthalpy can be described as:

$$h_{v1} - h_{cond} = c_{p,v}(T_{v1} - T_{cond}) + h_{fg}, \quad (1)$$

where h_{fg} denotes the condensation enthalpy. The first right hand term is due to cooling and is generally small compared to the heat of condensation. Using standard thermodynamic relations for an ideal gas at constant pressure [19], it can be shown that the difference in entropy is given by

$$s_{v1} - s_{cond} = c_{p,v} \ln \left(\frac{T_{v1}}{T_{cond}} \right) + s_{fg}. \quad (2)$$

Again the term due to cooling will be small compared to the condensation entropy s_{fg} . Using the concept of availability (exergy) with the evaporation temperature T_{evap} of the liquid as reference, the change in availability can be written as

$$b_{v1} - b_{cond} = h_{v1} - h_{cond} - T_{evap}(s_{v1} - s_{cond}). \quad (3)$$

Substituting equation (1) and (2) in (3) yields

$$b_{v1} - b_{cond} = c_{p,v} \left[(T_{v1} - T_{cond}) - T_{evap} \ln \left(\frac{T_{v1}}{T_{cond}} \right) \right] + h_{fg} - T_{evap}s_{fg}. \quad (4)$$

In the limit of an infinite contact surface $T_{v1} = T_{cond}$ and equation (4) reduces to

$$b_{v1} - b_{cond} = h_{fg} - T_{evap}s_{fg}. \quad (5)$$

In thermodynamic equilibrium the change in Gibbs free energy is zero and $h_{fg} = T_{cond}s_{fg}$. In that case equation (5) can be written as

$$b_{v1} - b_{cond} = \left(1 - \frac{T_{evap}}{T_{cond}} \right) h_{fg} = \eta_{carnot} h_{fg}. \quad (6)$$

In other words: even in the case of a thermodynamically perfect equipment work $w = b_{v1} - b_{cond}$ has to be provided to the process by a thermodynamic engine. Integration of equation (6) over the relevant bubble - dew point line allows direct calculation of the work lost. Cryogenic separation to certain specifications by the route of distillation or CRS requires the same amount of work. Typical values for energy consumption will be presented in section 3.

2.2 Equipment size

The basic idea behind the CRS process is that condensation by expansion is faster compared to condensation by adiabatic cooling and therefore has an overall shorter residence time and equipment size compared to distillation. As the contaminated natural gas expands, it cools down. At a certain point during this cooling process the saturation pressure becomes, due to the lower temperatures, lower than the partial vapour pressure of the contaminants. When this happens part of the gas starts to condense. Condensable matter can form new droplets or so-called nuclei which is the homogeneous nucleation process, or the condensable matter diffuses to and condenses on existing droplets or particles. Homogeneous condensation occurs very fast [7, 17]. As soon as the number of particles and nuclei formed by homogeneous condensation becomes of the order 10^{12} , the condensation process becomes dominated by heterogeneous condensation. The droplet formation during expansion cooling is therefore limited by the heterogeneous condensation process.

Condensation is a combined heat and mass transfer process. The ratio between thermal diffusivity to mass diffusivity (Lewis number) determines which process is dominant. As both processes are analogous diffusion processes, they can be described in the same manner [20].

The total droplet surface area in a tube per unit length of pipe A'_p is defined as:

$$A'_p = \pi d_p^2 N \frac{\pi}{4} D_{tube}^2, \quad (7)$$

where the term πd_p^2 represents the surface area of one nucleus and N represents the number of nuclei per cubic metre, whereas $\frac{\pi}{4} D_{tube}^2$ represents the volume per unit length. Therefore, the vapour flux towards the particles in the tube per unit length can be defined as:

$$j'_{\alpha,p} = \frac{N d_p \text{Sh}(d_p) \pi^2 D_{tube}^3 \mathbb{D} M_\alpha}{4 R_u T_g} (p_{\alpha,p} - p_{\alpha,g}). \quad (8)$$

where M_α represents the molar mass of the aerosol forming compound, R_u is the universal gas constant, T_g is the flue gas temperature, p is the partial vapour pressure and \mathbb{D} represents the diffusion coefficient of the condensing vapour. Equation (8) is based on diffusion of molecules to the surface of the particle in the continuum limit.

Inertia effects can be neglected for particles in micrometre range [21], implying that the particles follow the streamlines and the net flow around the particle is zero. Therefore, the ratio of convective to diffusive mass transport (Sherwood number) $\text{Sh}(d_p)$ as mentioned in Eq. 8 equals 2 in the case of small net mass-transfer rates and zero flow around a spherical particle.

In contact devices, such as distillation columns and two phase heat exchangers, the condensing molecules have to diffuse through the gas phase to the gas-liquid. To compare we assume that this interface coincides with the tube diameter D_{tube} . The mass vapour flux towards the walls of the heat exchanger per unit length $j'_{\alpha,w}$ can be written as [20]:

$$j'_{\alpha,w} = \frac{\text{Sh}(D_{tube}) \pi D_{tube} \mathbb{D} M_\alpha}{R_u T_g} (p_{\alpha,w} - p_{\alpha,g}), \quad (9)$$

The tube diameter at which condensation to the surface equals condensation to nuclei can be found by equalling equation (9) to (8)

$$N d_p D_{tube}^2 = \frac{4 \text{Sh}(D_{tube})}{\pi \phi \text{Sh}(d_p)} = \mathcal{O}(1) \quad (10)$$

The reported number concentration under typical CRS conditions varies between 10^{12}m^{-3} for condensation on foreign nuclei [7, 22] and 10^{20}m^{-3} for homogeneous condensation [17, 22]. The corresponding droplet sizes are 10^{-5}m and 10^{-7}m . As a lower limit for the product $N d_p$ we expect a value of 10^7m^{-2} . The consequence is that the contact device must have typical dimensions below

1 mm to obtain the same condensation rate in the same volume. As practical devices have typical dimensions of 10 mm or larger, the potential for size reduction is about 10^2 , provided that the contact devices in the CRS process the heat exchangers and separators are also significantly smaller.

The design goal for a distillation column is maximum selectivity, or local thermodynamic equilibrium between the gas and liquid phase. The consequence is that the maximum temperature difference that can be used for heat transfer is ΔT_1 in Fig.2. As the heat exchangers in the CRS process are not used for separation, the full temperature difference ΔT_2 between the bubble and dew point line can be used for heat transfer. For an equal design, the size of the heat exchangers in CRS are at least $\Delta T_1 / \Delta T_2$ smaller than a distillation column. In practical circumstances of methane production this amounts to a factor 0.1 or less. This is also the factor by which CRS as a whole is smaller when compared to a complete distillation process with the same separation performance.

3 CRS as a bulk sweetening process

As the energy demand of the classic chemical absorption process increases rapidly with the contamination level, processes relying on a combination of chemical and physical absorption are being developed for moderately contaminated ($\sim 10\%$) gas fields [24]. Under those conditions CRS can be applied as a bulk separation process prior to the chemical/physical absorption process to reduce the size and energy consumption of the latter.

3.1 Moderately contaminated gas field

The case study presented in this section is based on the data for the combined stream of the recently discovered (2008) Arabiyah and Hasbah gas fields in Saudi-Arabia [25]. The main components of the gas are 77 %_{mole} CH₄, 8 %_{mole} CO₂, 4 %_{mole} H₂S and 1 %_{mole} C₂H₆. One of the reported options for the amine based gas sweetening process is the Shell Sulphinol process.

The sour gas is fed at elevated pressure (70 bar) to an absorption tower where it is contacted with the solvent to absorb the acid gas components. The rich solvent is subsequently flashed to a lower pressure to regain most of the absorbed hydrocarbons. The rich solvent is then regenerated by steam heating in the stripper column. The energy demand associated with the solvent regeneration of this process is given as 3.4 MJ thermal per kg of treated gas, equivalent to 9% of the heating value of the incoming gas. To obtain this value a thermal to work efficiency of 0.4 is assumed. As the circulation rate of the solvent and therefore the energy demand and size of the stripper column is proportional to mole percentage of contaminant present in the gas [26], application of CRS before the amine treatment can result in considerable savings on both size and energy demand of the process.

The flow diagram for a condensed rotational separation process with recirculation as bulk separator before the amine process is given in Fig.3. To obtain this diagram an extended equation of state program is used based on a cubic equation of state of the Soave-Redlich-Kwong type with pure component parameters fitted to vapor pressures and liquid densities along with a composition dependent mixing rule. A freeze out model to predict the boundary of solid CO₂ formation is incorporated. The motivation for developing an in-house code instead of using commercially available programs is that flexibility in the optimization studies was needed. The final results for operation within the vapour-liquid region are double-checked against a commercial available computer package; Aspen-Plus.

To reach the desired temperature of -80 °C, the feed gas is first cooled (HE1) to -46 °C at 70 bar and subsequently expanded to 40 bar. The pre-treated gas leaves the first separator (RPS1) at -80 °C and this cooling energy is recovered in HE4. After the RPS the contaminant level of CO₂+H₂S is down from 12 to 5 %. The liquid leaving RPS1 is heated to -52 °C (HE2) and further throttled to 10 bar at -80 °C before entering the second separator (RPS2). The methane content of the gas leaving RPS2 is approximately equal to that of the incoming feed gas and can be mixed with this stream

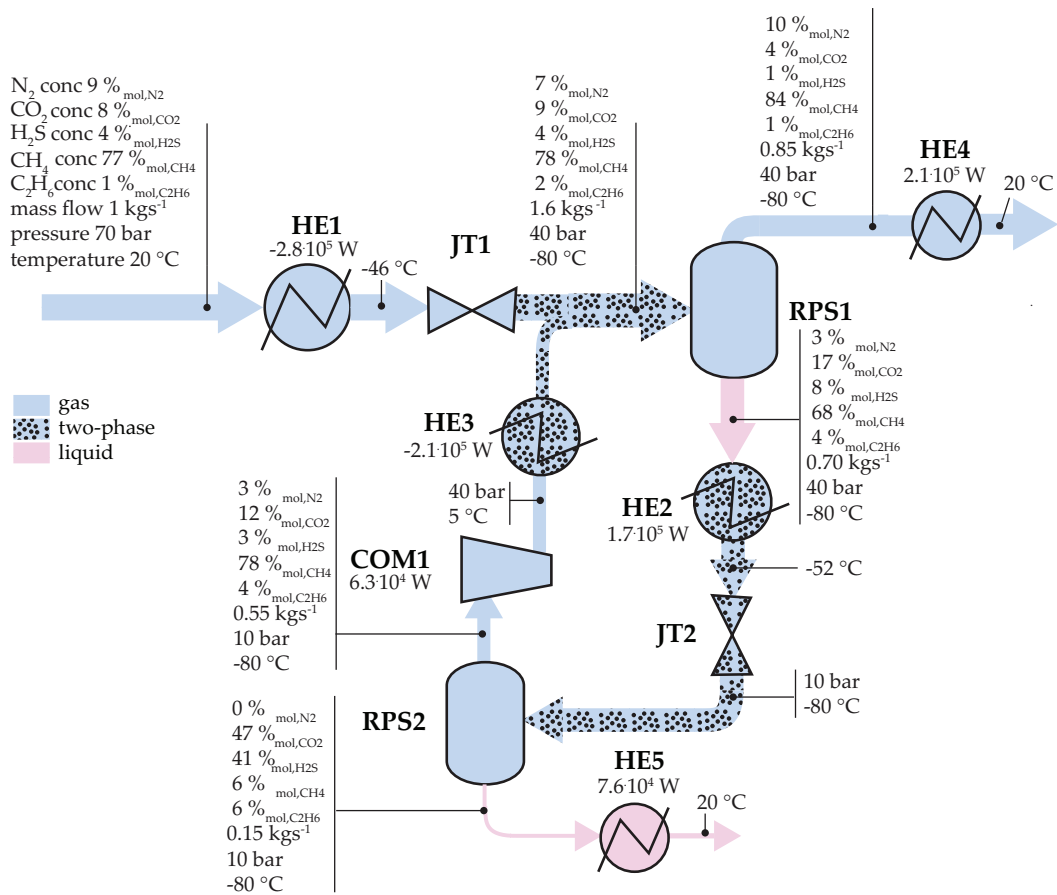


figure 3: Flow diagram of the two-stage separation process for a moderately contaminated gas field

after recompression to 40 bar and cooling to $-80\text{ }^{\circ}\text{C}$. The methane content leaving RPS2 in the liquid is about 7%.

The cooling requirement of heat exchanger 2 can be balanced by heat exchanger 4 in a single heat exchanger. Heat recovery from the product streams is almost sufficient to cool down the incoming gas. The remainder (0.04 MJkg^{-1}) has to be supplied by an external cooler. Assuming a heat to power efficiency of 0.4 and a compressor efficiency 0.85, the energy required by the cooler and compressor amounts to only 0.2 % of the heating value of the incoming gas. The hydrocarbons dissolved in the liquid $\text{CO}_2/\text{H}_2\text{S}$ represent 4.5 % of the total heating value. This energy can be used to generate steam for the the reboiler of the Sulphinol process. The feed gas flow after the CRS process is lowered by 15 % and the contamination level decreases from 8 to 4 % $_{mole}$ CO_2 and from 4 to 1 % $_{mole}$ H_2S . As a result the required circulation rate for the amine process more than halves. The associated energy demand decreases from 9 % to under 4 % of the incoming heating value.

As stated in section 2, contaminated gas mixtures with more than two components allow for operation of Condensed Rotational Separation in the vapor-liquid-solid region; as long as the solid fraction does not become too dominant. If the temperature at the first separation (RPS1) is lowered to -100°C , and the pressure is reduced to 22 bar, the contaminant level of $\text{CO}_2+\text{H}_2\text{S}$ in the gas after RPS1 is further reduced from 5% down to 1.5% while part of the CO_2 freezes out as a pure solid. At this separating condition only 0.5% of the fluid volume is occupied by solid CO_2 . As solid CO_2 is heavier than the liquid phase, both the solid CO_2 and the liquid are separated. The ratio of solid to liquid volume fraction is in the order of 50%. The separated solid and liquid are expected to form a diluted sludge of dry ice and liquid, which can be separated by the RPS as if it were just

table 1: Separation performance of CRS, applied as a bulk sweetening process for the Arabiyah and Hasbah gas fields and operated in both the vapour-liquid and vapour-liquid-solid region.

	VL-region		VLS-region	
Separation conditions:	rps1: -80°C, 40bar rps2: -80°C, 10bar		rps1: -100°C, 22bar rps2: -70°C, 4bar	
[%_{mole}]	vap. conc.	liq. conc.	vap. conc.	liq. conc.
N ₂	10.1	0	10.4	0
CO ₂	4.3	46.6	0.9	63
H ₂ S	1.1	40.8	0.6	34.4
CH ₄	83.7	6.6	86.9	0.8
C ₂ H ₆	1	6.0	1.2	1.8

a high viscous liquid. If the second separation temperature remains unchanged, and the pressure is decreased from 10 to 4 bar, solid CO₂ would also be formed in the second stage. With a ratio of solid to liquid volume fraction of 45%, a sludge of liquid and solid can again be expected. The methane content of the sludge leaving RPS2 drops till 1.3%. A sludge of solid and liquid is however not preferable as a waste product. More convenient is to heat up the second stage to -70°C, where no solid is formed anymore. The methane content in the liquid then even drops to <1%, and the ethane content decreases to 1.8%, which is significantly lower then in the case of CRS operated only in the vapor-liquid regime. Table 1 summarizes the separation performance for CRS operated in the vapor-liquid region (left) and in the vapor-liquid-solid region (right), and shows the benefits of operation in the VLS region.

3.2 Equipment size estimation

In [?] it is identified that performance for rotational separations can be expressed in terms of three variables which determine capital and operating costs. These were the flow rate, residence time and specific energy consumption. The results are a function of volume flow and not of mass flow. For the conditions of the first RPS operating in the VL-region (-80 °C, 40 bar), this leads to a volume of approximately 0.03 m³ for a treated gas flow of 1 kgs⁻¹. Although the massflow through the second RPS is smaller, also the pressure is lower, leading to a higher volume flow and consequently a larger volume (0.04 m³skg⁻¹). The design procedure as described in [23] is used to assess the dimensions of the RPS unit. Compared to the other parts of the process, the RPS requires a negligible amount of energy.

To estimate the size of the heat exchangers we use the transfer unit approach. The number of (heat) transfer units is defined as $NTU_H = UA/C_{min}$ with U being the overall heat transfer coefficient, A the heat transferring surface, and C_{min} the smallest capacity flow. The capacity flow is defined as $C = \dot{m}(h_{in} - h_{out})/(T_{in} - T_{out})$. For a single phase flow C is equal to $\dot{m}c_p$. With the definition of the capacity flow, the effectivity can be written as:

$$\epsilon = \frac{C_h(T_{h,in} - T_{h,out})}{C_{min}(T_{h,in} - T_{c,in})} = \frac{C_h(T_{c,out} - T_{c,in})}{C_{min}(T_{h,in} - T_{c,in})}, \quad (11)$$

where the denominator, or maximum possible rate of heat transfer, is based on the stream with the smallest capacity flow. The effectivity is a function of the ratio $C_r = C_{min}/C_{max}$ and the number of transfer units $NTU = UA_w/C_{min}$. For a counterflow heat exchanger we can obtain the relation [19]:

$$\epsilon = \frac{1 - \exp[(C_r - 1)NTU]}{1 - C_r \exp[(C_r - 1)NTU]}. \quad (12)$$

Introducing an entropy generation number as the entropy generation per unit heat transferred $N_s =$

$[S_{gen}(T_{h,in} - T_{c,in})]/\dot{Q}$, the entropy production due to the unbalance C_r can be derived as

$$N_{s,C_r} = \left\{ \frac{1}{C_r} \ln[1 - \epsilon C_r(1 - R)] - \ln[1 - \epsilon(1 - R)] \right\} / \epsilon, \quad (13)$$

where $R = T_{h,in}/T_{c,in}$. The derivative of relation (13), dimensionalized with the maximum entropy production to the number of transfer units is depicted in Fig.4. For all values of C_r the decrease in entropy production becomes small for NTU numbers larger than 2. We therefore fix the size of the heat exchangers to be used in the first stage at $NTU = 2$.

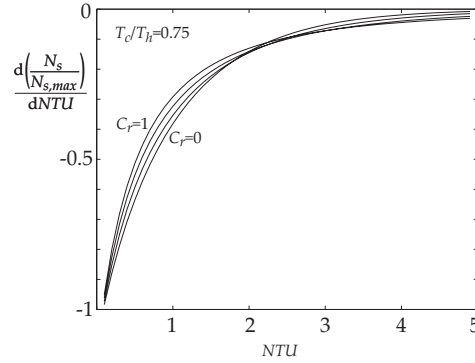


figure 4: Derivative of the dimensionless energy production to the number of transfer units.

Estimation of the dimensions of the heat exchangers is based on a multi-stream, spiral-wound-type [27] as commonly applied in LNG plants. Typical dimensions of the tubes are 10 mm with a spacing (centre to centre) of 15 mm. Reported Reynolds numbers vary between 2000 and 10000, with heating surfaces between 50 - 150 m^2/m^3 . The heat transfer coefficients were calculated as described by [28, 29], typically in the order of 500-1000 $Wm^{-2}K^{-1}$. For the conditions of HE2/4, a volume of 0.12 m^3skg^{-1} is required, for HE1/3/5 0.10 m^3skg^{-1} . The total estimated volume of the CRS pre-treatment unit is estimated at 0.31 m^3skg^{-1} . This compares to 2 m^3skg^{-1} for a typical stripper [30, 31]. Application of CRS as pretreatment halves the needed solvent circulation rate and also halves the necessary volume of the stripper. The overall saving in dimensions is 0.7 m^3skg^{-1} . An impression of the size of a CRS unit treating 200 $kg s^{-1}$ (665 MMscf/day), equivalent to one of the four lines foreseen for the combined Arabiyah and Hasbah gas streams is given in Fig.5.

In summary: even at contamination levels where absorption technology provides a sound economical gas treating option, the combination with CRS has the possibility to reduce the contamination level before the amine treatment unit by 50%, lowering both the energy costs and footprint by 50%. In the case of the Arabiyah and Hasbah gas fields, two of the four projected strippers become superfluous and 5% more methane is produced. Operation in the VLS region may even make amine treatment superfluous.

3.3 Severely contaminated gas field

The advantages of combining CRS with an absorption technology becomes even more profound for high contamination levels. As an example we look at a plant pre-treating 30 kg/s (100 MMscf/day) of severely contaminated gas with the composition 59% CH_4 , 27% H_2S and 14% CO_2 corresponding to the Huwaila field in Abu Dhabi.

Gas fields with contamination levels in this range are currently not explored as the costs for gas treating are considered to be un-economic. In section 2 it is already shown that applying CRS as a bulk separator makes it possible to enrich the methane concentration to 95% CH_4 . The corresponding process scheme is shown in Fig.6.

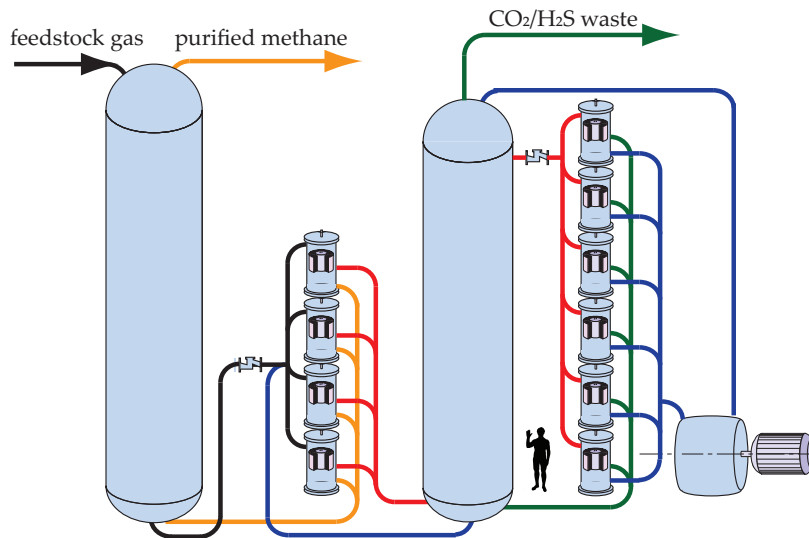


figure 5: Conceptual design of a 665 MMscf/day installation (on scale)

The compressors (COM1 and COM2 in Fig.6) both operate at a pressure ratio <4 , implying that standard single stage compressors, as commonly used in the LNG industry, can be utilized. It is noted that the addition of the regeneration loop hardly affects the size of the installation as equipment for an external cooling cycle would be of the same size.

Heat can be exchanged between HE2 and HE3 of Fig.6 in a single heat exchanger. This heat exchanger requires a volume of 10 m^3 . The cooling energy in the liquid product stream is sufficient to cool the incoming feed gas stream. This heat exchanger has a volume of 15 m^3 . This leaves the cooling energy in the methane stream available for other uses.

The energy costs of the cycle are again expressed as a percentage of the heating value of the incoming methane. The energy lost due to the methane in the liquid amounts to 1.9 %, the regeneration compressor consumes 0.4 % of the energy and the methane compressor 1.4 %. The conclusion is that approximately 4 % of the incoming heating value is used to bring the gas to a composition where conventional technology becomes economically feasible.

If we compare the energy costs with the moderately contaminated case, the compression costs increase from 0.2 to 2.8 %, amongst others due to the higher exit pressure. The costs associated with the loss of hydrocarbons in the liquid decrease however from 4.5 to 1.9 %.

4 Discussion

Condensed rotational separation is a process of bulk separation based on flash evaporation or pressure distillation. The rotational particle separator provides the means to separate the micron sized droplets formed after the flash from the gas. When applied to sour gas fields the concentration of contaminants can be lowered to a level that allows economic application of conventional physical and/or chemical absorption processes. The benefits of using condensed rotational separation for severely contaminated fields is obvious as there are no serious economic alternatives. But even for moderately contaminated fields where conventional methods are considered for gas sweetening, CRS can significantly lower the operational and capital costs as is shown for the Arabiyah and Hasbah gas fields in Saudi-Arabia.

Separation performance of CRS can be greatly improved by operation in the VLS-region, instead of the VL-region, as mentioned in section 3.1. During the expansion cooling of fluid towards

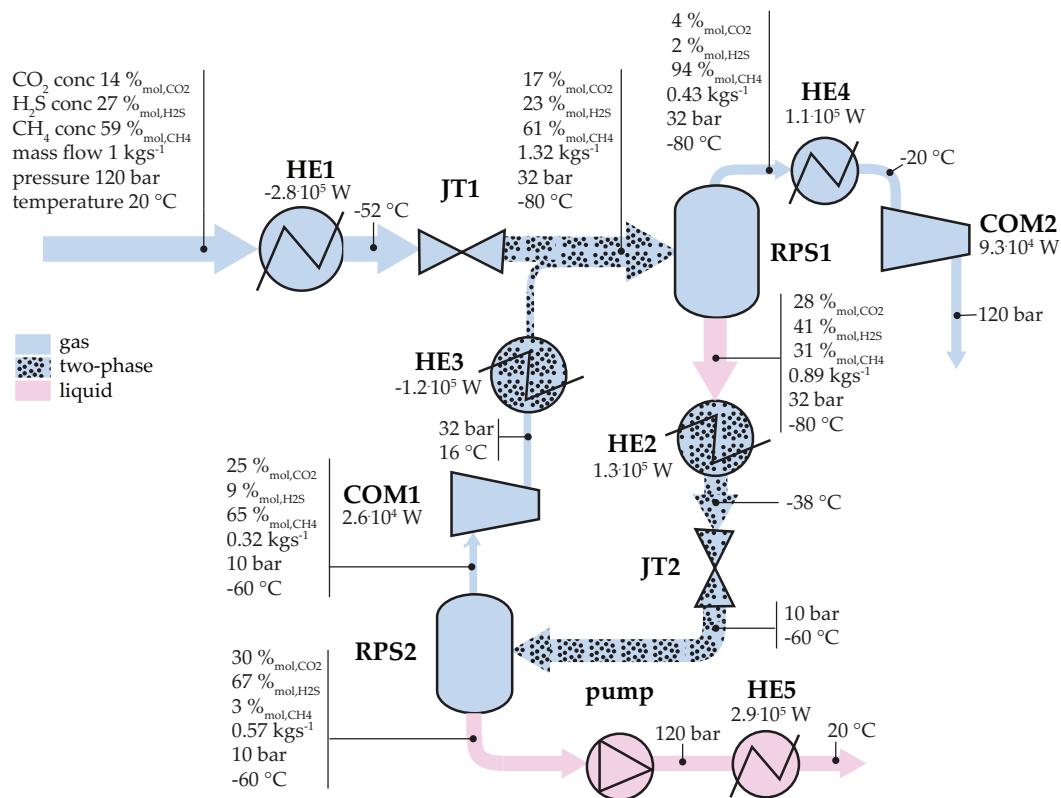


figure 6: Flow diagram of the two-stage separation process for a severely contaminated gas field

conditions in the VLS region, not only droplets are formed but also solid material or crystallized CO_2 is expected. Prediction of droplet and crystal growth are coupled by heat and mass transfer which make prediction of particle size difficult. As droplet and crystal growth occurs under non-equilibrium conditions and expansion is a fast mechanism of cooling, it is even a question whether solid crystals are formed or if the liquid remains at a subcooled state. So far there is no experience with expansive cooling into this area for the purpose of CRS. Prediction of particle growth and practical experience with expansion cooling into the VLS-region are topics of future interest. That this is practically feasible has been shown by the Cryocell process [3].

The notion that micron sized droplets are formed in any separation process involving a phase change, led to the investigation of the technology for other areas of application: removal of condensate from natural gas, removal of CO_2 from coal gasification producer gas and removal of CO_2 from coal combustion gas. These applications are shortly discussed.

Natural gas condensate is a low-density mixture of hydrocarbon liquids that are present as gaseous components in the raw natural gas produced from many natural gas fields. The incoming gas is cooled to below the dewpoint of the hydrocarbons of the feedstock pressure. In the separator a good part of the hydrocarbons are removed. Subsequently the gas condensate is throttled to a low pressure separator. The reduction in pressure across the throttle valve causes the condensate to undergo a partial vaporization where even more hydrocarbons can be removed. The RPS separation technology as described above can directly be applied to this process with the following advantages: higher separation efficiency, smaller installation and lower energy usage.

Application of CRS to conventional coal combustion power plants shows that large improvements can be made in separation efficiency when combined with a relatively small oxygen enrichment of the air. The energy costs of separation are below those of existing carbon capture technologies,

although low energy oxygen enrichment technologies need to be found. The key factor of the attractiveness of this technology is finding cheap methods for slight oxygen enrichment [32, 33].

5 Conclusions

The capability of collecting micron sized droplets from gas streams by the rotational particle separator enables the use of pressure distillation as a compact alternative to fractional distillation. This in turn allows new process designs for:

- 1 upgrading sour gas fields
- 2 collecting valuable condensate from wet gas
- 3 CO₂ removal in coal conversion

Condensed rotational separation in the vapor-liquid region can be used as a bulk sweetening process prior to conventional techniques. Combination of CRS with conventional separation techniques can decrease both footprint and energy cost.

Operation of CRS within the vapor-liquid-solid region shows great potential for improvement of yields and purities. In the gas sweetening of light contaminated gas fields, CRS has the potential to produce sweetened natural gas at (almost) pipeline specifications, with a CO₂ and H₂S content <1% in the sweetened gas, while methane content in the waste is small, < 1%.

The key elements of condensed rotational separation have been demonstrated at lab scale and semi-industrial scale. The next step is to start field tests of the RPS and perform a more detailed analysis of the CRS process and the other equipment.

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References

- [1] Olajire AA. CO₂ capture and separation technologies for end-of-pipe applications, A review, *Energy* 2010; 35(6):2610-2628
- [2] Mougin P, Renaud X, Elbaz G. Operational validation of the Spretl process for bulk H₂S and mercaptans removal, *The Gas Industry: Current & Future* 2008;(6):17-19 IFP, Rueil-Malmaison, France
- [3] Hart A, Gnanedran N. Cryogenic CO₂ capture in natural gas, *Energy Procedia* 2009; 1(1): 697-706
- [4] Mart CJ, Valencia JA, Northrop PS. Developing sour gas resources: controlled freeze zone technology with integrated acid gas management. In: *Sour Oil & Gas Advanced Technology*, Abu Dhabi; 2010
- [5] Clodic D, El Hitti R, Younes M, Bill A, Casier F. CO₂ capture by anti-sublimation, Thermo-economic process evaluation, In: *National Conference on Carbon Sequestration*, DOE, Washington; 2005
- [6] Schaber K, Körber J, Ofenloch O, Ehrig R, Deuffhard P. Aerosol formation in gas-liquid contact devices - nucleation, growth and particle dynamics, *Chem. Eng. Sc.* 2002;(57):4345-4356

- [7] de Best CJJM, van Kemenade HP, Brunner T, Obernberger I. Particulate Emission Reduction in Small-Scale Biomass Combustion Plants by a Condensing Heat Exchanger, *Energy and Fuels* 2008;22(1):587-597
- [8] Austrheim A. Experimental Characterization of High-Pressure Natural Gas Scrubbers, PhD thesis, University of Bergen, Norway, 2006
- [9] Willems GP, Golombok M, Tesselaar G, Brouwers JJH. Condensed rotational separation of CO₂ from natural gas, *AIChE Journal* 2010;56(1):156
- [10] Hinds W.C. *Aerosol Technology*. New York: John Wiley & Sons, 1999.
- [11] Strauss W. *Industrial gas cleaning*. 2nd ed. Oxford: Pergamon 1975.
- [12] Campbell JM, Norman OK. *Gas conditioning and processing. The equipment modules*, Campbell, 1994
- [13] Purchas DB. *Solid liquid separation technology*. Croydon: Uplands; 1981.
- [14] Clift R. *Inertial separators: basic principles in Gas cleaning in demanding applications*. ed. Seville JPK; London: Blackie; 1997.
- [15] Svarovsky L. *Hydrocyclones*. London: Holt; 1984.
- [16] Schinkelshoek P, Epsom HD. Supersonic gas conditioning - Commercialisation of twister technology. In 87th GPA annual convention, Grapevine, USA, March 2-5, 2008
- [17] van Wissen RJE. *Centrifugal Separation for Cleaning Well Gas Streams: From Concept to Prototype*, PhD thesis, Eindhoven University of Technology, 2006, the Netherlands
- [18] Haseli Y, Dincer I, Naterer GF, Exergy Analysis of Condensation of a Binary Mixture With One Noncondensable Component in a Shell and Tube Condenser, *J. Heat Transfer* 2008;130 084504
- [19] A. Bejan, *Advanced engineering thermodynamics - 2nd ed*. Wiley, New York, 1997
- [20] Bird RB, Stewart WE, Lightfoot EN. *Transport Phenomena - 2nd ed.*, Wiley, New York, 2002
- [21] Fuchs, N. A. *The Mechanics of Aerosols*; Pergamon Press Ltd.: New York, 1964
- [22] Bansal G, Golombok M, Brouwers JJH, Tesselaar G. CO₂ droplets condensed from natural gas. I&EC research [dx.doi.org/10.1021/ie1012689](https://doi.org/10.1021/ie1012689), published online January 31, 2011
- [23] Willems GP, *Condensed rotational cleaning of natural gas*, PhD-thesis, Eindhoven University of Technology, 2009
- [24] Munder B, Grob S, Fritz PM. Selection of wash systems for sour gas removal. 4th International Freiberg Conference on IGCC & XtL Technologies. Dresden, 3-5 May, 2010
- [25] Alami IA. Wasit gas plant: New sour gas developments in Saudi Arabia. In 6th Sour Oil and Gas Advanced Technology conference, Abu Dhabi, March 30 - April 1, 2010
- [26] Younger AH, *Gas Processing Principles and Technology - Part II*, University of Calgary, 2004
- [27] Catalogue entitled *Rohrbündel-Wärmeaustauscher*, Linde A.G., Werksgruppe, TVT, Munich, Germany.
- [28] Neeraas BA, Fredheim AO, Aunan, B. Experimental shell side heat transfer and pressure drop in gas flow for spiral-wound LNG heat exchanger, *Int. J. Heat and Mass Transfer* 2004;47; 353-361

- [29] Neeraas BA, Fredheim AO, Aunan, B. Experimental data and model for heat transfer, in liquid falling film flow on shell side, for spiral-wound LNG heat exchanger, *Int. J. Heat and Mass Transfer* 2004;47; 3565-3572
- [30] Gas Processors Suppliers Association. Engineering data book, 12th edition 2004
- [31] Kashani A, Fernandez Rodiles R, Giesen V, Katz T. The biggest challenge in sour gas: acid removal technologies. In 4th International Freiberg Conference on IGCC & XtL Technologies, Dresden, May 3-5 2010
- [32] Benthum R.J. van, Kemenade H.P. van, Brouwers JJH, Golombok M. CO₂ capture by Condensed Rotational Separation. In International Pittsburgh Coal Conference, Istanbul, October 11-14, 2010
- [33] Theunissen T, Golombok M, Brouwers JJH, Bansal G, Benthum, R.J. van, Liquid CO₂ droplet extraction from gases, *Energy* 2011;
- [34] Brouwers JJH, Kemenade HP van, Kroes JP. Rotational Particle Separator: an efficient method to separate micron-sized droplets and particles, 2011 AFS annual conference, May 10-12, Louisville, USA