Condensed rotational cleaning of natural gas

PROEFSCHRIFT

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Summary

Condensed rotational cleaning of natural gas

An increasing fraction of the world’s natural gas resources is contaminated with CO₂ and H₂S. This study describes the experimental verification of a novel gas separation process. The process speeds up centrifugal separation by preferentially condensing one of the components (CO₂) e.g. by Joule-Thomson expansion. The mixture splits into two phases: a liquid phase which is enriched in CO₂ and a gaseous phase that is enriched in methane. The liquid phase forms a mist of micron-sized particles. For normal separation, a costly high pressure low temperature section would then be required to enable droplet growth to a size where current separation technology can be applied. However the novel method, condensed contaminant centrifugal separation (C3sep) can rapidly remove the micron size particles using a rotating particle separator.

The process is an option for solving the problem of the vast amounts of natural gas fields contaminated with CO₂ and H₂S (21% of global gas reserves), where economic production with traditional methods is not feasible. The invented process also offers the possibility for cheap and energy-efficient removal of CO₂ from syngas of gasification power plants thus providing a breakthrough technology for reducing CO₂ emissions by capturing it in an easily storable form.

The process is demonstrated at laboratory scale for a broad range of binary CH₄/CO₂ gas mixtures (20-80 mole% CH₄). Results of experiments confirm theoretical predictions. Some preliminary droplet measurements that are relevant for the scaling of the separation equipment are included. To reliably test the performance of the separator a larger scale test is required. The film behavior within the vertically oriented RPS element is studied analytically under influence of gravitational and centrifugal acceleration in combination with large shear forces. From the film behavior the optimal element orientation is deduced. Using the experimental results, an industrial scale prototype separator is designed to operate at field scale throughput, pressure and temperature conditions. For fast preliminary separator testing purposes, a full size model separator (based on water/air) has been built to study liquid removal and separation efficiency.
# Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unit</th>
</tr>
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<tbody>
<tr>
<td>$A$</td>
<td>Area</td>
<td>$[m^2]$</td>
</tr>
<tr>
<td>$a$</td>
<td>Radius of induction section</td>
<td>$[m]$</td>
</tr>
<tr>
<td>$B$</td>
<td>Bypass flow rate</td>
<td>$[kg \ s^{-1}]$</td>
</tr>
<tr>
<td>$B_m$</td>
<td>Bypass % measured</td>
<td>$[%]$</td>
</tr>
<tr>
<td>$B_t$</td>
<td>Bypass % theoretical</td>
<td>$[%]$</td>
</tr>
<tr>
<td>$B_{tm}$</td>
<td>Bypass % theoretical/measured</td>
<td>$[%]$</td>
</tr>
<tr>
<td>$B_{0cr}$</td>
<td>Critical Bond number</td>
<td>[-]</td>
</tr>
<tr>
<td>$b$</td>
<td>Average value of $b$</td>
<td>$[m]$</td>
</tr>
<tr>
<td>$b_i$</td>
<td>Intersection point parabolic profile and $z$ axis</td>
<td>$[m]$</td>
</tr>
<tr>
<td>$b_i$</td>
<td>Bypass concentration for component $i$</td>
<td>[-]</td>
</tr>
<tr>
<td>$C$</td>
<td>Perimeter of droplet</td>
<td>$[m]$</td>
</tr>
<tr>
<td>$C$</td>
<td>Constant</td>
<td>[-]</td>
</tr>
<tr>
<td>$C_0$</td>
<td>Initial concentration</td>
<td>[-]</td>
</tr>
<tr>
<td>$C_c$</td>
<td>The Cunningham slip correction</td>
<td>[-]</td>
</tr>
<tr>
<td>$C_D$</td>
<td>Drag force coefficient</td>
<td>[-]</td>
</tr>
<tr>
<td>$c$</td>
<td>Concentration mole fraction</td>
<td>[-]</td>
</tr>
<tr>
<td>$D$</td>
<td>Diffusion coefficient</td>
<td>$[m^2 \ s^{-1}]$</td>
</tr>
<tr>
<td>$D_q$</td>
<td>Particle diffusion coefficient</td>
<td>$[m^2 \ s^{-1}]$</td>
</tr>
<tr>
<td>$D_{q}$</td>
<td>Quasi laminar turbulent diffusion coefficient</td>
<td>$[m^2 \ s^{-1}]$</td>
</tr>
<tr>
<td>$D$</td>
<td>Drag force</td>
<td>$[N]$</td>
</tr>
<tr>
<td>$D_h$</td>
<td>Hydraulic diameter</td>
<td>$[m]$</td>
</tr>
<tr>
<td>$D_i$</td>
<td>Tangential gas inlet</td>
<td>$[m]$</td>
</tr>
<tr>
<td>$d$</td>
<td>Droplet diameter</td>
<td>$[m]$</td>
</tr>
<tr>
<td>$d_c$</td>
<td>Channel height</td>
<td>$[m]$</td>
</tr>
<tr>
<td>$d_p$</td>
<td>Diameter of particle</td>
<td>$[m]$</td>
</tr>
<tr>
<td>$d_{pi}$</td>
<td>Droplet size interval</td>
<td>$[m]$</td>
</tr>
<tr>
<td>$d_{p,50%</td>
<td>_{pre}}$</td>
<td>$d_{p50%}$ of pre-separator</td>
</tr>
<tr>
<td>$d_{p50%}$</td>
<td>Diameter of particle collected with 50% probability</td>
<td>$[m]$</td>
</tr>
<tr>
<td>$d_{p100%}$</td>
<td>Diameter of particle collected with 100% probability</td>
<td>$[m]$</td>
</tr>
</tbody>
</table>
$E$  Youngs modulus  \[N \text{ m}^{-2}\]

$F$  Feed flow rate  \[kg \text{ s}^{-1}\]

$F$  Force  \[N\]

$F_d$  Resistance force  \[N\]

$f$  Friction factor  [-]

$f_i$  Volume fraction  [-]

$g$  Gravitational acceleration  \[m \text{ s}^{-2}\]

$\bar{g}$  Gravitational acceleration vector  \[m \text{ s}^{-2}\]

$ar{H}$  Averaged value of $H$  \[m\]

$H$  Parabolic profile height  \[m\]

$h$  Channel height  \[m\]

$h$  Volute height  \[m\]

$h$  Enthalpy  \[kJ \text{ kg}^{-1}\]

$\bar{h}$  Dimensionless channel height ($\phi$)  [-]

$\bar{h}$  Dimensionless channel height ($\delta_0$)  [-]

$I$  The moment of stiffness  \[m^4\]

$\dot{I}$  Rate of angular momentum  \[kg \text{ m}^2 \text{ s}^{-2}\]

$\dot{I}_d$  Total dissipation rate of angular momentum  \[kg \text{ m}^2 \text{ s}^{-2}\]

$\dot{I}_{db}$  Dissipation rate of angular momentum bottom  \[kg \text{ m}^2 \text{ s}^{-2}\]

$\dot{I}_{ds}$  Dissipation rate of angular momentum side wall  \[kg \text{ m}^2 \text{ s}^{-2}\]

$\dot{I}_f$  Rate of angular momentum of the feed  \[kg \text{ m}^2 \text{ s}^{-2}\]

$K_i$  Equilibrium ratio  [-]

$k$  Von Kármán’s constant  [-]

$L$  Length  \[m\]

$L$  Liquid flow rate  \[m^3 \text{ s}^{-1}\]

$L_{pre}$  Pre-separator length  \[m\]

$m$  Mass  \[kg\]

$m$  Mass flow rate  \[kg \text{ s}^{-1}\]

$m_f$  Mass flow rate of the feed  \[kg \text{ s}^{-1}\]

$M_{w_{tot}}$  Mole weight mixture  \[kg \text{ mole}^{-1}\]

$M_{w_{Vap}}$  Mole weight vapour  \[kg \text{ mole}^{-1}\]

$N$  Rotational velocity  \[rpm\]

$n$  Number of counts  [-]

$P_{frac}$  Product fraction  [-]

$P_k$  Turbulence production  \[m^2 \text{ s}^{-3}\]

$P$  Pressure  \[N \text{ m}^{-2}\]

$P$  Product flow rate  \[kg \text{ s}^{-1}\]

$p_{s}$  Saturation pressure  \[N \text{ m}^{-2}\]

$p$  Pressure  \[N \text{ m}^{-2}\]

$Q$  Volume flow rate  \[m^3 \text{ s}^{-1}\]

$Q_f$  Volume flow rate  \[m^3 \text{ s}^{-1}\]

$R$  Radius  \[m\]

$R$  Outer radius volute  \[m\]

$R_{co}$  Radius co-rotating wall  \[m\]

$R_{pre}$  Radius pre-separator  \[m\]

$R_{Shaft}$  Radius with half surface area  \[m\]
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>( R_{50%} )</td>
<td>Radius with half surface area ([\text{m}])</td>
</tr>
<tr>
<td>( r )</td>
<td>Radius ([\text{m}])</td>
</tr>
<tr>
<td>( r_b )</td>
<td>Intersection point parabolic profile and volute bottom ([\text{m}])</td>
</tr>
<tr>
<td>( \bar{r}_b )</td>
<td>Average value of ( r_b ) ([\text{m}])</td>
</tr>
<tr>
<td>( r_i )</td>
<td>Recovery for component ( i ) ([-])</td>
</tr>
<tr>
<td>( r_w )</td>
<td>Radius of wall ([\text{m}])</td>
</tr>
<tr>
<td>( r_{50%} )</td>
<td>Radius halfway ([\text{m}])</td>
</tr>
<tr>
<td>( \text{Re} )</td>
<td>Reynolds number ([-]</td>
</tr>
<tr>
<td>( \text{Re}_\delta )</td>
<td>Local Reynolds number (based on ( \delta )) ([-]</td>
</tr>
<tr>
<td>( \text{Re}_p )</td>
<td>Particle Reynolds number ([-]</td>
</tr>
<tr>
<td>( S )</td>
<td>Super saturation ratio ([-]</td>
</tr>
<tr>
<td>( s_{\text{gap}} )</td>
<td>Gap size ([\text{m}])</td>
</tr>
<tr>
<td>( T )</td>
<td>Temperature ([\text{K}/\degree\text{C}])</td>
</tr>
<tr>
<td>( T_{\text{solid}} )</td>
<td>Freeze out temperature ([\text{K}/\degree\text{C}])</td>
</tr>
<tr>
<td>( t )</td>
<td>Time ([\text{s}])</td>
</tr>
<tr>
<td>( U )</td>
<td>Volume flow rate per unit dept ([\text{m}^2 \text{s}^{-1}])</td>
</tr>
<tr>
<td>( U_b )</td>
<td>Bulk flow velocity ([\text{m} \text{s}^{-1}])</td>
</tr>
<tr>
<td>( u )</td>
<td>Velocity ((x \text{ direction})) ([\text{m} \text{s}^{-1}])</td>
</tr>
<tr>
<td>( \bar{u} )</td>
<td>Mean velocity ([\text{m} \text{s}^{-1}])</td>
</tr>
<tr>
<td>( u' )</td>
<td>Velocity fluctuation ([\text{m} \text{s}^{-1}])</td>
</tr>
<tr>
<td>( u_\ast )</td>
<td>The shear velocity ([\text{m} \text{s}^{-1}])</td>
</tr>
<tr>
<td>( V )</td>
<td>Volume droplet/ring ([\text{m}^3])</td>
</tr>
<tr>
<td>( V_{\text{T}} )</td>
<td>Vapour flow rate ([\text{m}^3\text{s}^{-1}])</td>
</tr>
<tr>
<td>( v )</td>
<td>Velocity ([\text{m} \text{s}^{-1}])</td>
</tr>
<tr>
<td>( v )</td>
<td>Velocity ((y \text{ direction})) ([\text{m} \text{s}^{-1}])</td>
</tr>
<tr>
<td>( \vec{v} )</td>
<td>Velocity vector ([\text{m} \text{s}^{-1}])</td>
</tr>
<tr>
<td>( v_{a,x,\text{pre}} )</td>
<td>Axial velocity pre-separator ([\text{m} \text{s}^{-1}])</td>
</tr>
<tr>
<td>( v_g )</td>
<td>Gas velocity ([\text{m} \text{s}^{-1}])</td>
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<tr>
<td>( v_{g,0} )</td>
<td>Initial gas velocity ([\text{m} \text{s}^{-1}])</td>
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<tr>
<td>( \bar{v}_g )</td>
<td>Tangential velocity ([\text{m} \text{s}^{-1}])</td>
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<tr>
<td>( v_{g,f} )</td>
<td>Tangential feed velocity ([\text{m} \text{s}^{-1}])</td>
</tr>
<tr>
<td>( v_0 )</td>
<td>Feed velocity ([\text{m} \text{s}^{-1}])</td>
</tr>
<tr>
<td>( \text{W} )</td>
<td>Waste flow rate ([\text{kg} \text{s}^{-1}])</td>
</tr>
<tr>
<td>( \text{W}_c )</td>
<td>Correction factor methane entrainment ([-]</td>
</tr>
<tr>
<td>( w )</td>
<td>Width of the plate ([\text{m}])</td>
</tr>
<tr>
<td>( x )</td>
<td>( x ) coordinate ([\text{m}])</td>
</tr>
<tr>
<td>( x_i )</td>
<td>Liquid/Waste concentration for component ( i ) ([-]</td>
</tr>
<tr>
<td>( x_0 )</td>
<td>Point of velocity inflection ([\text{m}])</td>
</tr>
<tr>
<td>( x_2 )</td>
<td>Distance to wall ([\text{m}])</td>
</tr>
<tr>
<td>( y )</td>
<td>( y ) coordinate ([\text{m}])</td>
</tr>
<tr>
<td>( y_i )</td>
<td>Vapour/Product concentration for component ( i ) ([-]</td>
</tr>
<tr>
<td>( z )</td>
<td>( z ) coordinate ([\text{m}])</td>
</tr>
<tr>
<td>( z )</td>
<td>Axial position ([\text{m}])</td>
</tr>
<tr>
<td>( z_i )</td>
<td>Feed concentration for component ( i ) ([-]</td>
</tr>
<tr>
<td>( z_i )</td>
<td>( z ) position of film interface ([\text{m}])</td>
</tr>
</tbody>
</table>
Greek symbols

\( \beta \)  
Empirical swirl factor 
[-]

\( \Delta \)  
Change 
[-]

\( \delta \)  
Film thickness 
[m]

\( \delta \)  
Inner to outer RPS filter diameter ratio 
[-]

\( \epsilon \)  
Turbulent dissipation 
[m² s⁻³]

\( \epsilon_{red} \)  
Reduction of the effective cross sectional area 
[-]

\( \eta \)  
Efficiency 
[-]

\( \theta_{sep} \)  
Separation angle 
[-]

\( \lambda \)  
Mean free path 
[m]

\( \mu \)  
Dynamic viscosity 
[Pa s]

\( \nu \)  
Kinematic viscosity 
[m² s⁻¹]

\( \xi \)  
Pressure loss factor 
[-]

\( \rho \)  
Density 
[kg m⁻³]

\( \sigma \)  
Surface tension 
[N m⁻¹]

\( \tau \)  
Residence time 
[s]

\( \tau \)  
Shear stress 
[N m⁻²]

\( \tau_t \)  
Turbulent stress 
[N m⁻²]

\( \tau_0 \)  
Shear stress 
[N m⁻²]

\( \phi \)  
Film thickness at \( x = x_0 \) 
[m]

\( \phi \)  
Total volume flow 
[m³ s⁻¹]

\( \psi \)  
Film feed term 
[m²]

\( \Omega \)  
Angular velocity 
[rad s⁻¹]

\( \omega \)  
Angular velocity of the film 
[rad s⁻¹]

Superscripts and subscripts

\( ax \)  
Axial direction

\( B \)  
Bypass stream

\( b \)  
Due to buoyancy

\( c \)  
Methane rich phase

\( c \)  
Due to centrifugal acceleration

\( c, ch \)  
Channel

\( co \)  
Co-rotating wall

\( d \)  
Due to drag

\( des \)  
Design

\( dyn \)  
Dynamic

\( e \)  
Element

\( F \)  
Feed stream

\( f \)  
Of the feed

\( fc \)  
RPS element

\( g \)  
Gas
$gap$ Corresponding to the gap
$gas$ Corresponding to the gas
$i$ Inner
$i$ Direction indicator
$i$ Particular species or component
$j$ Direction indicator
$l$ Liquid
$m$ Measured
$m$ Mean
$N$ In normal direction
$n$ Stage
$o$ Outer
$out$ Flowing out
$P$ Product stream
$p$ Particle
$post$ Post separator
$r, rad$ Radial direction
$sep$ Separation
$shear$ Due to shear
$su$ Due to surface tension
$t$ Tangential
$t$ Theoretical
$tot$ Total
$stat$ Stationary
$unbal$ Unbalance
$v$ Vapour
$W$ Waste stream
$w$ Liquid phase
$w$ Due to friction
$0$ Initial; maximum; laminar
$1$ Methane
$2$ Carbon dioxide
$3$ Hydrogen sulfide
Chapter 1

Introduction

1.1 Natural gas field contamination

Natural gas can be found in various places throughout the world. The quality of the natural gas fields varies substantially. Fields with a high purity of methane (CH$_4$) are commonly referred to as “sweet gas fields”. Fields that contain high levels of hydrogen sulfide (H$_2$S) are called “sour” gas fields, while, fields that are contaminated with significant amounts of acidic gases e.g. carbon dioxide (CO$_2$) or hydrogen sulfide (H$_2$S) are called “acid” gas fields.

The fraction of natural gas resources that is severely contaminated with CO$_2$ and H$_2$S is steadily growing. The reason is that easy-to-produce “sweet gas fields” are produced first leaving the more difficult fields for future generations. The more difficult fields are often not attractive to national oil companies (NOC’s) but may be of interest to International oil companies (IOC’s) [1].

Roughly 35% of the current worldwide amount of unproduced gas [2], is heavily contaminated with CO$_2$ or H$_2$S. i.e. levels exceeding 10 mole% CO$_2$ and/or 5 vol% H$_2$S. We only consider gas fields which fulfil the following practical limitations:

- the size of the field exceeds 0.5 Tscf (10$^6$ MMscf),
- associated fields have a gas/oil ratio exceeding 1000 scf/bbl,
- specific reports on CO$_2$ and H$_2$S concentrations are available.

This adds up to a total amount of contaminated gas, which is estimated at $2.2 \cdot 10^9$ MMscf (million standard cubic feet, the units conventionally applied in the gas business) or $4.5 \cdot 10^{13}$ kg of CH$_4$ [1]. This is equivalent to (2300 EJ) which is approximately four times the world annual energy demand. Note that many contaminated fields have never been properly logged due to the fact that they were considered to have no development potential.

Fields containing large fractions of contaminants, cannot be economically produced with currently available technology [3]. Thus there is a need to develop gas cleaning methods that can cope with high contamination levels up to 70 vol% H$_2$S and/or CO$_2$. 
1.2 Separation technologies

The processes for cleaning acid gas, can be divided into 5 categories [4]:

1. Chemical solvent/solution processes, e.g. amine treatment. These solvent processes employ chemical absorption of the contaminant. They thus involve a chemical reaction between the solvent and the contaminant. The absorption processes are mainly used for small amounts of contamination (contaminant levels typically below 15%). Regenerating the absorbent is expensive due to the high CO\textsubscript{2} desorption energy costs.

2. Physical solvent processes, e.g. Selexol which is a mixture of the dimethyl ethers of polyethylene glycol and Rectisol which uses refrigerated methanol. In these solvent processes use is made of physical absorption of the contaminant. These processes are mainly used for large degrees of contamination, they involve relatively low desorption energy costs. Installations, however, are very large, and solvent loads are considerable.

3. Direct conversion processes based on chemical conversion of the contaminant into other molecules, e.g. preferential sulfur oxidation from H\textsubscript{2}S. However this technology is only feasible for small levels of contamination [4]. Desorption energy costs are moderate.

4. Dry bed processes, e.g. zeolites and pressure swing adsorption, are based on chemical or physical adsorption of the contaminant onto solid matter. These processes are only applied for small fractions of contamination. In case of chemical adsorption the adsorbent is not regenerable. In case of physical adsorption the method has moderate desorption energy costs.

5. Miscellaneous processes, such as: water wash, low temperature condensation / freeze out, cryogenic distillation and membranes.

A general drawback of the above listed processes is that their implementation leads to either large installations requiring high investment costs or large amounts of energy consumption, or both (in most cases) [3]. These limitations have stopped heavily contaminated gas fields from coming into production. The new process of condensed rotational separation suffers much less from these limitations. The investment costs are low and the energy consumption is approximately 1% of the lower heating value of the produced natural gas [5]. This new process including the first experimental measurements is the subject of this thesis.
1.3 Principles of Condensed Rotational Separation

Condensed rotational separation, denoted as CRS, is a new method for separating mixtures of gases. The working principle is as follows [6]. The thermodynamic state of the mixture is changed such that one of the components preferentially condenses into a liquid. As a result a mist consisting of a large number of ultra-fine micron-sized aerosol particles is formed [7]. The small size of the droplets is due to slow diffusion within the binary gas mixture. These particles are subsequently separated by a newly developed centrifugal separator known as a rotating particle separator, in short RPS [6]. An illustration of the process is given in fig. 1.1. Bringing one of the gaseous components of the mixture into a liquid state generally implies cooling. This can be done by pressure reduction, either isentropic through an expansion turbine (shown in fig. 1.1) or isenthalpic by means of a Joule Thomson valve. An isenthalpic expansion path is shown in fig. 1.2 curve (A), as well as an isentropic expansion path (B) and direct isobaric cooling using a heat exchanger (C).

The separation of the liquid component (CO$_2$/H$_2$S) occurs by the RPS. The core of the separator is the RPS element, i.e. a rotating cylindrical body which consists of a multitude of axially oriented channels (see fig. 1.3). The diameter of the channels is typically 1-2 mm, its length 0.2-0.5 m. The element is about 0.4-1m in diameter. After entering the channels of the rotating body, liquid mist particles entrained in the gas are centrifugated towards the collecting walls (see fig. 1.3) [8–10]. They then form a film of liquid flowing downwards parallel with the gas (see fig. 1.4). At the exit of the channels, the liquid film breaks up into droplets of 50-100 $\mu$m in diameter (see fig. 1.4). These droplets are centrifugated to the casing wall and subsequently leave the device via a liquid drain.

The rotating element of the RPS can be set into rotation either by an external drive, or by the working fluid itself. In the latter case use is made of a static configuration designed such that the gas flow obtains a swirling motion. Examples are tangentially oriented blades or a tangentially mounted inlet pipe, like in a cyclone

![Figure 1.1](image.png)

Figure 1.1. Process overview comprising external cooling, expansion cooling, phase separation, liquid removal and recompression.
The process of condensed rotational separation was initially devised as a solution for the problem of heavily contaminated natural gas fields with CO\textsubscript{2} and/or H\textsubscript{2}S \cite{6}. Other ("downstream") applications which have arisen in the mean time are:

- CO\textsubscript{2} separation from H\textsubscript{2} in (coal)gasification processes,
- CO\textsubscript{2} separation from N\textsubscript{2} in (coal)combustion installations.

The incentive for these applications is the greenhouse gas issue and global warming: CRS is a possible breakthrough technology for CO\textsubscript{2} removal in electricity production from coal. The present thesis deals with CO\textsubscript{2} and/or H\textsubscript{2}S removal from methane only ("upstream"). The process is for that case referred to as: Condensed Contaminant Centrifugal Separation, abbreviated C3sep.

1.4 Economical aspects and benefits

The major incentive for pursuing C3sep are its economics. Both operational costs which mainly result from energy consumption of the process and capital cost which are related to the size of the installation appear to be favorable \cite{5}. Energy consumption is very low because of the high pressure of the gas coming from the pre-treatment. The gas coming from the well is expanded to the pressure of the surface facility manifold which is typically 100 bar \cite{3}. Expansion via a turbine expander already leads to (almost) sufficient cooling to arrive at the desired temperature for separation. The shaft power can be used to drive the compressor to bring the cleaned gas back to manifold pressure (see fig. 1.1). As the RPS requires hardly energy (ca. 40 kW for a 20 MMscf/d separator see 4.4.3), such a scheme of applying an expander-compressor combination would lead to almost no energy consumption at all.
This is in contrast with conventional methods based on absorption. These processes require large amounts of energy to an extent that in case of high contamination levels, they consume practically all energy of the methane produced.

It is because of the availability of RPS technology that the process of condensed rotational separation can be achieved in a compact installation. Applying rapid cooling, in about 0.1 seconds, a mist is created with particles as small as 1 micron in diameter. The RPS enables such particulate matter to be removed effectively with low energy consumption. Peripheral velocities of the RPS element are limited to 45
m/s implying modest mechanical and dynamical loading. Other ways of removing the liquid fraction are possible, e.g. by cyclones. Such methods however, require much larger droplet sizes [11] leading to residence times which are approximately $10^3$ times larger [12]. The size of the installation in which the liquid phase formation and subsequent separation would take place at temperatures of -50 °C would become correspondingly larger, this would increase capital expenditure. Capital investments for C3sep are mainly due to the expander-compressor combination. The components of the one step separation system can be found in fig. 1.1. The additional investments associated with the system are earned back in roughly one or two months considering a gas price which is equivalent to a conservative 40 dollar/barrel (700 k€ daily income).

The process serves as a pre-treatment for heavily contaminated gas. The gas is enriched up to approximately 86% CH$_4$ for pure CO$_2$/CH$_4$ mixtures and up to 96% CH$_4$ when a considerable amount (i.e. ca. 5%) of H$_2$S is present in the feed gas. The methane purification can be completed by conventional amine treaters downstream of the C3sep process to arrive at required pipeline specification (CO$_2$ <2,5 vol% and H$_2$S<5 ppm [3; 13]).

1.5 RPS technology

Many RPS’s have been designed and tested over the past 15 years [14–19], but for other areas of application: e.g. ash removal from flue gas of combustion installations, air cleaning in domestic appliances, product recovery in pharmaceutical and food industry and oil/water separation. The advantage for the food/pharmaceutical industry is that the separation takes place within a stainless steel environment which is easy to clean. Stainless steel can also operate at high temperatures which makes different processes viable. The oil water separation is mainly aimed at complying with increasing environmental legislation. The air cleaning device is designed for people with allergic/respiratory problem. Illustrations of designs applied in these areas are shown in fig. 1.5.

Most of the early designs were aimed at solid-gas separation and employ an air cleaning device. The current process deals with liquid-gas separation which has the advantage that the separated medium immediately flows out of the separation channels. Knowledge and design rules which have resulted from the activities related to these application areas have been incorporated in the development of RPS technology for C3sep.

1.6 Goal and outline

The first steps in the development of C3sep were reported in the PhD-thesis of van Wissen [5]. His work was focussed on thermodynamic analysis of condensing gas mixtures, principles of centrifugal separation [12] and the design of a lab-scale (50 kscf/d) version of C3sep to be built at Shell’s Global Solutions Laboratory in Amsterdam (SRTCA). The aim of the present research is to design, build, commission
and modify the test-loop in order to provide a basis for semi-industrial design and testing of C3sep. To achieve this goal a number of scientific and technical problems had to be tackled.

Having completed the lab-scale test rig in Amsterdam, a series of measurements were performed to validate thermodynamic behavior. A flow of 60 Nm$^3$/h which amounts to approximately $10^{-3}$ times the flow of a typical gas field, is studied consisting only of CH$_4$ and CO$_2$ with varying mixture ratios. Cooling was achieved by
a heat exchanger in combination with a Joule Thomson valve. Separation of CO$_2$
droplets occurred through a specifically designed RPS [5]. In chapter 2 the results of
laboratory scale experiments are presented. A broad range of binary CH$_4$/CO$_2$ gas
mixtures (20-80 mole%) was considered. The droplet size distribution was also mea-
sured and was used to predict typical droplet sizes present at large scale applications.
Droplet sizes varying between 1 to 10 micron were detected by a new measurement
technique provided by MTS (Meßtechnik Schwartz). Because of the inherently high
efficiency of rotating separation equipment at small scale, the separator could not be
thoroughly tested throughout the full range of operating conditions at lab scale. All
together the working principle of CRS/C3sep, i.e. the condensation and subsequent
centrifugal separation of a contaminating component from natural gas, was proven at
lab scale.

Another point of concern was the transport of liquid via a film in the channels of
the RPS. A theoretical analysis was performed to assess the thickness of the film and
its interaction with the gas stream for the cases of co-current flow and countercurrent
flow of gas and liquid through the channels. The effect of the gravitational force,
i.e. orientation of the RPS element was studied [20]. Chapter 3 is devoted to the
film behavior within a vertically oriented RPS element. An analytical model was
derived to predict film behavior under the influence of gravitational and centrifugal
acceleration in combination with high shear forces. From the model the optimal
element orientation and flow direction were deduced.

The validated theoretical assumptions were used to come to a scaled-up design
for the separator for testing at industrial scale i.e. >20 MMscf/d in chapter 4. The
separator was constructed in a way that it can handle a field scale (20 MMscf/d)
throughput, at the thermodynamically favorable pressures and temperatures. Special
attention was given to the mechanical construction in relation to semi-cryogenic and
corrosive conditions (due to the presence of the acidic gasses H$_2$S and CO$_2$). The
resulting conceptual design has to be evaluated in more detail prior to fabrication.

In addition to the testing in Amsterdam, a large scale RPS was built and sub-
sequently tested at Eindhoven University of Technology. The test unit operated at
atmospheric conditions using a mixture of air with dispersed water droplets. The
volume of air flow and the amount of water corresponded to the volumetric flow in
actual m$^3$/h and the amount of CO$_2$ droplets of a gas field of 80 MMscf/d. New for
the RPS in C3sep applications were the high liquid loads which resulted in drainage
problems in the small scale separator. To handle such liquid loads in the scaled up
separator, special attention was given to the dimensioning of the liquid drainage sys-
tem. In chapter 5, a full size model separator (based on water/air mixtures) was
tested to study liquid removal and separation efficiency. Efficient phase separation
for C3sep applications is possible with the current basic design.

In chapter 6 the conclusions with particular relevance for the entire process de-
velopment are restated and discussed. Special attention is given to the practical
applicability of the technology within future testing and application. Suggestions for
further research/work are included. Research and development on C3sep has now
come to a point where testing at real field conditions is imminent.
Chapter 2

Experimental validation of the C3sep process at lab scale

2.1 Introduction

After a thorough theoretical analysis of the process [5], an experimental test rig was required for validation. A 50 kscf/d test unit was designed and built at SRTCA (Shell Research and Technology Center Amsterdam). This chapter describes the setup, experiments, results and concluding remarks. The experiments have been performed on a wide range of binary compositions of CH₄/CO₂ varying from 20-80 mole% CH₄.

Because the droplets are in thermodynamic equilibrium with the gas, no complete separation of the gases can be achieved, even when the phases are separated completely. At all operating conditions the droplets will still contain traces of CH₄ and the gas will still contain CO₂. Therefore the cleaned gas needs treatment with other technology, e.g. amine treatment, to get up to the purity that is required to meet pipeline specifications [3; 13]. Depending on the concentration of the contaminated gas and operation conditions of the process a methane purity of 83-99 mole% can be achieved with C3sep. To investigate whether the C3sep technology performs up to thermodynamic predictions, the tests described in this chapter have been performed.

This chapter focuses on experiments performed with the first prototype RPS presented by van Wissen [5]. Section 2.2 describes the process background and the separation of the condensed phase. In section 2.3 the experimental setup is described and in section 2.4 the thermodynamic measurement results are discussed. In section 2.5 droplet formation is investigated in more detail and some preliminary droplet measurements are presented. In section 2.6 the optimal separation conditions and the dependency on waste recuperation is discussed.

2.2 Process background

The C3sep process incorporates expansion cooling, formation and growth of droplets rich in the contaminating substance and subsequent separation of these droplets, see fig. 2.1.

![Figure 2.1. Core separation process: expansion cooling, droplet formation and subsequent centrifugal separation of the condensed phase.](image)

The contaminated gas at high pressure is expanded e.g. with a Joule Thomson (JT)-valve. The gas then cools down due to the expansion and the contaminants preferentially condense into a mist of droplets. The droplets have time to grow while passing through the insulated induction section. The droplets of a few µm are subsequently separated with the rotational particle separator (RPS).

2.2.1 Thermodynamics

Due to the binary condensation a mist of small droplets is formed (see section 2.5) which quickly reaches thermodynamic equilibrium. When these droplets are in equilibrium with the gas phase, the droplets will only grow because of coagulation which is a process that relies on the mobility of the droplets. When droplets grow, the mobility rapidly decreases \[21\]. Overall this results in micron sized droplets when a growth time of approximately 1 second is available.

When a multi component mixture partly condenses, some molecules will be in the vapour phase and other molecules will be in the liquid phase. The liquid phase will mostly contain molecules of the species with the lowest partial vapour pressure \[11\]. The vapour phase will mainly contain molecules of the species with the highest partial vapour pressure. The two phases, liquid and vapour, finally reach equilibrium. The concentration depends on pressure, temperature and initial composition.

A phase separator as schematically depicted in fig. 2.2 is used to separate the vapour and liquid phase. The multi phase feed \((z)\) enters the phase separator, and is divided into an equilibrium gaseous \((y)\) and a liquid fraction \((x)\).

The (methane) enrichment is the change in mole fraction (methane) between feed \((z)\) and product \((y)\) stream \((y_1 - z_1)\). Where \(x_i, y_i\) and \(z_i\) are the mole fractions
of component $i$ in the liquid waste, product and feed, where $i=1$ refers to methane. The enrichment is therefore an important parameter to define the performance of the separation system for it determines the purity of the product. The ratio between the number of moles of methane in the product stream and the number of moles of methane in the feed stream is called recovery and can be defined as \[5\]:

\[
r_1 = \frac{y_1 Q_P}{z_1 Q_F} = \frac{y_1(z_1 - x_1)}{z_1(y_1 - x_1)}
\] (2.1)

where $Q$ is the mole flow rate, with the subscript $F$, $P$ and $W$ denoting feed, product gas and liquid waste respectively. With the described process we can choose between high recoveries and low enrichments or high enrichments at the expense of a lower recovery rate ($y_1 - z_1$ vs. $r_1$). Normally the higher the pressure the higher the enrichment but at the expense of a lower recovery rate because more methane will dissolve in the liquid CO$_2$. With pure CO$_2$ and CH$_4$ mixtures it is difficult to reach concentrations exceeding 85 mole% CH$_4$ in the gaseous product because of the thermodynamic properties [3]. When H$_2$S is present in the natural gas, the freeze out temperature of the CO$_2$ shifts to lower temperatures, thereby enabling possible enrichment up to gas concentrations over 95 mole% at high recovery rates [5].

In general, the lower the pressure at constant temperature, the higher the recovery of methane, but the enrichment diminishes due to the high amount of CO$_2$ that does not condense. The optimal pressures and temperatures can be found in the lower left corner of the "liquid and vapour" regime in the phase diagram (see fig. 2.3), close to the freeze out curve. The freeze out curve is the near vertical line at approximately -60 °C in fig. 2.3). The optimal process conditions for real applications have to be determined while incorporating the whole gas treatment facility in the analysis (see section 2.6).

For the experiments we use a binary mixture of methane (CH$_4$) and carbon dioxide (CO$_2$) with a mole fraction of methane ranging from $z_1=0.2$ to $z_1=0.8$. The phase diagram of the mixture varies considerably with the concentration. A typical phase diagram corresponding to a mixture fraction of $z_1=0.5$ can be found in fig. 2.3. On the lower right side of the phase diagram the vapour phase can be found. The two-phase region is located between the vapour and the liquid phase. The line on the lower right side of the phase diagram, separating the liquid and vapour from the vapour area is called the dew-point curve (see fig. 2.17). The line separating the
Figure 2.3. Phase diagram corresponding to $\text{CH}_4(1) + \text{CO}_2(2)$: $z_1 = 0.5$. The diagram is generated using an extended equation of state program based on a cubic equation of state of the Soave-Redlich-Kwong type with pure component parameters fitted to vapour pressures and liquid densities along with a composition dependent mixing rule. A freeze out model $T < T_{\text{solid}}$ $\text{CO}_2$ is incorporated. In the lower right corner the operation area of our setup is indicated.

liquid from the liquid and vapour area is called the bubble curve. On the left side of the diagram a vertical line is present, that indicates the formation of solid $\text{CO}_2$, the so called freeze-out curve. When the mixture is expanded from the gas phase to a pressure and temperature within the liquid and vapour regime, small liquid droplets are formed. The concentration of each phase as well as several other properties of both phases can be calculated using isothermal flash calculations.

2.2.2 Separation

One of the key features of the test setup is the novel phase separator, which has been described by van Wissen [5]. This phase separator is designed to separate large amounts of liquid $\text{CO}_2$ droplets larger than $1\mu$m from a semi-cryogenic (-60 °C) natural gas stream.

Because the separator is a novel apparatus the droplet separation process is studied in detail. The influence of the bends (in the induction section) on the separation of the small droplets is incorporated in the analysis. To estimate the effect of the bends, the separation performance of bends is estimated in appendix B. Because the bends can have a considerable effect on the droplet distribution, especially on droplets larger then $20 \mu$m, all bends located between the JT-valve and the separator have been removed.
Pre-separator performance

The rotating particle separator is constructed with a tangential inlet, as a pre-separator cyclone, to remove large droplets. To calculate the efficiency of the pre-separator, we first look at the efficiency of a gravitational separator. In case of a gravitational separator the terminal settling velocity \(v_T\) of the particle is used to predict the performance. The \(v_T\) is the velocity of a particle when the driving force is in equilibrium with the drag force on the particle. When viscous forces are dominating over the kinetic forces the particle is called a Stokes particle. The drag force of a particle or droplet with diameter \(d_p\) and relative velocity \(v\) can be described by [22]:

\[
F_d = \alpha v, \quad \alpha = \frac{3\pi \mu_g d_p}{C_c}, \quad C_c = 1 + \frac{2.52\lambda}{d_p} \tag{2.2}
\]

With: \(\mu_g\) the dynamic viscosity of the gas and \(\lambda\) the mean free path (air \(\approx 0.07 \mu\text{m}\)). The Cunningham slip correction \(C_c\) only becomes relevant when the particles size is smaller then 1 \(\mu\text{m}\) (especially at elevated pressures). The \(v_T\) for a small particle in the stokes regime \((C_c = 1)\) is described by:

\[
v_T = \frac{(\rho_p - \rho_g)d_p^2 g}{18\mu_g} \tag{2.3}
\]

with \(\rho_p\) the density of the particle/droplet, \(\rho_g\) the density of the gas and \(g\) the gravitational acceleration.

The particle diameter that a separator can collect with a 50% probability is called the \(d_{p50}\). This is analogous to the \(d_{p100}\) which is the droplet size that is collected with a 100% probability. The \(d_{p50}\) can be calculated with a relation based on the figure:

\[
d_{p50} = \sqrt{\frac{9\mu_g v_{ax}(r_w^2 - r_{50}^2)}{(\rho_p - \rho_g)v_t^2 L}} \tag{2.4}
\]

with \(v_{ax}\) the gas velocity in axial direction, \(v_t\) the gas velocity in tangential direction (which is constant with respect to \(r\)), \(r_w\) the radius of the wall, \(r_{50}\) the radius with
half the flow surface on the inside and half the flow surface on the outside of the radius, and \( L \) the length of the pre-separator. The centrifugal acceleration \( v_T^2/r \) was substituted for the gravitational acceleration in eq. (2.3). The tangential velocity is assumed constant within the whole pre-separator, due to the stabilizing influence of the rotating element where the velocity at the inside of the cyclone is equal to the element velocity.

Equation (2.4) yields a \( d_{p50\%} \) which varies between 1 and 3.3 \( \mu m \) depending on the tangential speed of the separator element and gas flow rate. The \( d_{p50\%} \) definition is used instead of the \( d_{p100\%} \) definition because the turbulent flow will interfere with the particle collection [9].

As a result of turbulence, it is impossible to define an efficiency of 100%, due to the fact that a small fraction of the droplets theoretically will never reach the wall. From Direct Numerical Simulation (DNS) calculations [8] it follows that to achieve 98% separation efficiency the mono disperse particle diameter \( d_p \) should be around \( 3d_{p50\%} \) or the \( d_{p50\%} \) should be chosen \( 1/3d_{p,100\%} \).

**RPS element performance**

For a single row of RPS channels at constant radius (see fig. 2.8 below) a relation for the \( d_{p50\%} \) of the channels can be derived with help of the \( v_T \) see fig. 2.5. In this figure, a single channel is depicted rotating around a central axis at a radius \( r \). A droplet that enters the channel on the left side is forced to the outer wall of the channel with a velocity equal to the \( v_T \). The distance the droplet has to travel to achieve 50% separation equals half the height of the channel for rectangular channels. In that case:

\[
v_T \frac{L_c}{v_{ax}} = \frac{1}{2} d_c \quad (2.5)
\]

with \( L_c \) the length of the channels and \( d_c \) the height of the channels. This can be rewritten as:

\[
d_{p50\%} = \sqrt{\frac{9d_c \mu_g v_{ax}}{(\rho_p - \rho_g)\Omega^2 r L_c}} \quad (2.6)
\]

where \( \Omega^2 r \) is the magnitude of the centrifugal acceleration with \( \Omega \) the rotational speed of the element (rad/s). The \( d_{p50\%} \) varies between 0.3 and 1.2 \( \mu m \) depending on the rotational speed of the separator.
2.3 Experimental setup

Summarizing, the pre-separator collects half of the droplets in the range of 1-3.3 \( \mu \text{m} \) and above while the RPS element collects half of the droplets from sizes as small as 0.3-1.2 \( \mu \text{m} \). Additional separation even at a rotational velocity \((N)\) of 0 rpm is caused by the impactor effect due to the small space between the element and housing at the entrance of the separator (see fig. 2.4). This impactor effect results in good separation efficiencies even at static conditions.

2.3 Experimental setup

The actual separation process in a gas field application is a once through process (see fig. 2.1). For testing, we have constructed a closed gas loop (60 Nm\(^3\)/hr \( \approx \) 50kscf/d) with a gas conditioning section and a separation section. The gas conditioning section simulates a gas well at variable composition (20-80 vol\% CH\(_4\) in CO\(_2\)) and pressures (above 80 bars). The conditioned gas is fed to the separation section. In the separation section the core process is located, which consists of:

- JT-Valve which expands and cools the gas
- Induction section where droplets get time to nucleate and grow until the droplets reach a size of a few microns
- Rotating phase separator which is a centrifugal separator with a bundle of axially orientated channels rotating around a common axis to separate the small waste droplets from the product gas.

In fig. 2.6 an overview of the test loop layout can be found. On the left side of the figure the gas conditioning section is displayed. This section simulates a gas well with the required concentration, pressure and temperature. When the gas is (re)conditioned the gas flows into the separation section. The gas separation section, comprising the core process, can be found on the right side of fig. 2.6.

2.3.1 Gas conditioning section

In the gas conditioning section the gas is (re)conditioned to represent a real contaminated gas field. The gas conditioning section is designed to cope with pressures up to 150 bars which is on the order of the surface manifold facility pressure of a gas treatment plant [4]. At startup the gas is supplied to the suction side of the diaphragm compressor from pre-mixed CH\(_4\)/CO\(_2\) pressurised gas bottles via a pressure regulator. The concentration can be fine-tuned using bottles of pure methane and carbon dioxide. The gas should be delivered to the separation section at a pressure exceeding 100 bars and a temperature around 20 °C. These conditions are necessary to get to the right separation conditions after the expansion. A diaphragm compressor (60 Nm\(^3\)/h) re-compresses the gas after it has been mixed to pressures up to 150 bars. After the gas is compressed it is cooled by a tap water chiller to around ambient temperature to remove the heat added by the compression. The conditioned gas is sent to the separation section.
2.3.2 Separation section

The separation section (fig. 2.6) of the test loop is constructed within an autoclave testing bunker and consists of an evaporator/cooler, a Joule Thomson (JT)-valve, an induction section, the RPS, a liquid CO$_2$ collection vessel, a couple of heaters and measuring equipment for pressures, temperatures and concentrations in the induction section.

The high pressure gas is first cooled in the evaporator with help of previously separated liquid carbon dioxide to approximately -5 °C. Then the pre-cooled gas is expanded by the JT-valve to around 30 bars. The expansion cooling is, due to the small flow rate, performed with help of a JT-valve instead of a turbine expander. It is not possible to buy an expansion turbine for these small flow rates and high pressures because this turbine would require very high rotational velocities and would still be very inefficient due to the high surface to volume ratio. During this isenthalpic expansion the gas is cooled to around -50 °C [5] and sent into the induction section (fig. 2.1) where droplets are formed.

Induction section

The induction section has variable volume in order to be able to investigate the growth rates of the droplets. Part of the induction section can be replaced, this gives the
opportunity to vary both the residence time and the surface to volume ratio. When condensable matter is cooled to a point where it can condense, the molecules can form stable clusters. The size of these stable clusters depends on the following parameters, the supersaturation ratio, surface tension, molecular volume and temperature. After these stable clusters have formed they can grow by condensation on, and coagulation of, the clusters. The diffusion and condensation of gas molecules on existing clusters, the wall, or other particles is called heterogeneous condensation. Coagulation will start to occur as soon as some stable clusters have formed. The mobility of the clusters rapidly decreases with increasing size because the drag forces rapidly increases with increasing diameter (see fig. 2.2).

For the measurements described in this chapter the residence time \( \tau \) that the gas is present in the induction section has been \( \tau \approx 0.3 \) seconds. This is the residence time corresponding to the smallest induction section. The smallest section is chosen to keep the droplets as small as possible to diminish the pre-separator influence. Unfortunately this section has a relatively large surface to volume ratio which can enhance wall condensation (see sections 2.5 and appendix C).

The required induction time is relevant because it determines the minimum size of the well insulated semi-cryogenic induction section on industrial scale that is needed to deliver sufficiently sized particles to the separator. A 200 MMscf/d gas treatment plant with a growth time of about 0.5 second already results in approximately 30 meters of 8.5 inch vacuum insulated and cooled piping. Because of the slow increase in droplet size a separator that can efficiently collect micron sized particles is indispensable, otherwise this would lead to very large and expensive equipment. On the small scale present in this experimental unit a cyclone separator would be possible. Actually the pre-separator cyclone in the set-up already collects most of the liquid. But on a field scale this will not be feasible, therefore we make use of the rotational particle separator for field applications.

**The separator**

The separator consists of a motor driven RPS element combined with a pre- and post-separator (liquid removal section see also 2.2.2). The element is driven by a DC motor that is connected via a magnetic coupling (type MAKX-40-2/12-EX of Burgmann Industries). The magnetic coupling consists out of a master and slave magnet separated by a PEEK (polyetheretherketone) cap that provides the gas tightness. The two phase gas stream is accelerated through a tangential inlet into the pre-separator as discussed in 2.2.2 (see fig. 2.7). The pre-separator is the zone of the separator where the coarse droplets are separated by the tangential velocity of the gas analogous to a cyclone separator. The coarse droplets collide with the wall and flow in a downward direction by gravity and are subsequently drained at the bottom side of the separator [20]. Because of the small scale of this particular separator, the pre-separator zone separates already micron sized droplets as discussed above. The small size is also the reason why the rotating element consists of only a single row of coagulation channels to prevent back flow through part of the RPS element.

Within the separator the droplets are spatially separated from the upward flowing carrier gas which is enriched in methane. As shown in fig. 2.8 the separator is
constructed with a single row of milled channels around the circumference of a solid Duplex steel cylinder. The outside of the channels is constructed by combining the, in an oven heated, shrink sleeve and the cooled element. When both temperatures settle at ambient conditions a rigid filter assembly is created. The gas flows in upward direction through the RPS element. Within the single row of small channels, the centrifugal force, forces the droplets to the outer walls of the RPS element. The droplets merge into a thin film on the inside of the shrink sleeve and depending on the gas flow velocity the film is expelled at the top or bottom side of the element. The liquid that is expelled at the bottom side is drained together with the liquid from the pre-separator and the liquid expelled from the top side is drained with help of a special flood groove.

Peripheral

Most of the equipment within the gas loop is only peripheral to ensure continuous recycling of the components. The incoming compressed gas flow is pre cooled within the evaporator, prior to the JT-valve expansion, with help of the separated liquid CO$_2$ that is evaporated.

After separation the clean product gas stream is heated (to prevent damage to the
2.3 Experimental setup

Figure 2.8. Cross section solid RPS element. The channels are milled in a solid duplex cylinder. The outside sleeve was shrinked around the cooled cylinder.

compressor) and sent back to the gas conditioning section. The liquid CO$_2$ streams (or waste streams) leaving the separator are both fed to a collection vessel which is also used for overnight CO$_2$ storage. To compensate for the pressure difference over the rotating element a dip-pipe is used within the collection vessel (see fig. 2.9).

In fig. 2.9 the feed stream coming from the gas condition section is cooled within the evaporator. The gas is then expanded over the JT-valve to temperature and pressures required for separation. The multi phase flow is then fed into the separator where both phases are separated. The two liquid lines connect to the same vessel, therefore a dip pipe is used to correct for the pressure drop over the rotating element.

There are two gas bypass lines, the gas bypass start-up and the gas bypass steady state. The gas bypass start-up is used to pre-cool the evaporator and thus the incoming gas during start-up, in order to diminish start-up time. The gas bypass steady state is used to enhance the liquid drainage from the separator during the measurements. The gas flow through this bypass line lowers the pressure within the collection vessel, and prevents liquid hold up in the separator.

2.3.3 Detection

There are several pressure gauges, both absolute (bara) and relative (barg), mounted in the setup. The temperatures are measured by several thermocouples. The flows (fig. 2.9) of feed (F), product (P), waste (W) and bypass (B) are measured with coriolis flow meters. Several other flows are controlled by mass flow controllers. The liquid levels in both the evaporator and liquid collection vessel are measured by radar level meters (Endress+Hauser, Levelflex M-FMP45). The gas concentrations (F,P,W,B) are monitored with a mass spectrometer or with a 4 channel gas chromatograph respectively.

To get a reliable thermodynamic measurement the system should be in steady state for about a hour. To get into this steady state the system has to run and cool down for a couple of hours to collect liquid CO$_2$. This cooling down is done with the gas bypass start-up line. This bypass line cools down the condenser and feed gas when there is not yet liquid CO$_2$ is available. When enough liquid is collected the
gas bypass start-up valve is closed and only liquid is sent to the evaporator. The heaters, gas supply and various other settings have to be adjusted, to let the system reach a steady state within two hours. During this steady state, the concentration, temperatures, pressures, flows and liquid levels will remain constant. All presented thermodynamic measurements are performed during stable steady states.

The setup first contained a quadrupole (Agilent) mass spectrometer (mass-spec) to measure the gas composition. Because we only have one mass spectrometer we can only sample one gas stream at the time. After a measurement it takes a few minutes to flush the measuring line. It is impossible to measure when the system is not in steady state for at least one hour. A shorter sample time is possible but the gas-consumption increases up to a point where the steady state of the system is compromised. The dynamic behaviour can be better observed with a continuous monitoring of all 4 streams, therefore a four channel gas chromatograph is installed.

**Concentration measurements by a Mass Spectrometer**

The setup first contained a quadrupole mass spectrometer (mass-spec/MS) to measure the gas composition. A mass spectrometer consists of three stages. In the first stage ions are formed. In the second stage the ions are accelerated and sent in a trajectory which is mass-to-charge ratio depended. In the last stage the actual detector is located. All three stages need to operate at very low pressure (<1e-4 Pa).

A typical concentration log measured via mass-spec can be found in fig. 2.10.
Because we have only one mass-spec we can only measure one channel at a time.

![Graph showing concentration measurement via mass-spectrometry](image)

**Figure 2.10.** Typical concentration measurement via mass-spectrometry

To get a measurement of the feed, product and waste streams, the MS is switched between 3 sampling lines. Because of the large dead volume in the tubing, the sample lines have to be flushed after switching. During the measurements performed with the mass-spec, the bypass line could not yet be sampled.

In fig. 2.10 it can be seen that during the first two hours the feed line is sampled. The normalized mole concentration of CH$_4$ is depicted with a continuous line, the normalized mole concentration of CO$_2$ is depicted as dots. The spike around 90 minutes is the transition which is an artefact of the loop construction and will be explained later. After 40 minutes, after the transition, the concentration has become stable, this can be seen because the concentration at 180 minutes is still the same. Besides a stable concentration also the flows, temperatures, motor current, and pressures have to remain constant. The product composition is measured at 150 minutes and the waste is sampled at 170 minutes. The reaction time of the MS, i.e. before the proper concentration is depicted, is about 10 minutes, this reaction time can only be achieved by venting the sample line to the incinerator. After 225 minutes the measurement is repeated. After 250 minutes, the steady state is lost.

There is a clear trade off between measuring accuracy of a specific sample line and system stability. A reliable measurement consists out of a lot of measuring points from a well flushed sample line. But because of this venting, the loop pressure will drop and the systems steady state will be lost.
Concentration measurements by a four channel Gas Chromatograph

The ASAP GC columns are packed with porus material, in our case PPQ (PoraPlot Q), to discriminate gas molecules with a low molecular weight. The packing has a different retention time for each of the components. The concentration is measured with help of a wheatstone bridge. A typical concentration log measured via GC can be found in fig. 2.11.

With the four channel gas chromatograph, all streams i.e. the feed, product, waste and bypass can be sampled simultaneously with a sample time of approximately 1.5 min. Note that, although the sampling lines are kept as small as possible, due to system delays (e.g. the liquid holdup in the liquid collection vessel) the various concentrations (F,P,W,B) can not be compared at a single time. So we need long duration steady state operation to get reliable measurements. Some insight into the dynamic behavior can be gained by considering and compensating for the fluid hold up, velocities of the fluids and mixing during storage in the collection vessel. The lag times are: F \approx 1 \text{ min}, P \approx 1 \text{ min}, W \approx 20 \text{ min} and B \approx 1 \text{ min}. Therefore, a reliable measurement can again only be performed during a steady state of about one hour. The time needed for a reliable measurement depends on the system stability, the liquid level in the collection vessel and mass flow rates.

2.3.4 Loop stability

The collection of all the liquids in the storage vessel gives rise to an inhomogeneous gas mixture during start up, i.e. the gas concentration in the gas conditioning section will become depleted in CO\textsubscript{2} whilst the the separation section will become enriched in CO\textsubscript{2}. This inhomogeneity has to be overcome by mixing, this mixing automatically takes place during operation. During the start-up phase a large part of the set-up has to be cooled down to -60 °C, this takes approximately three hours. While cooling the set-up, the gas composition has to be controlled by feeding pure CO\textsubscript{2} or CH\textsubscript{4} to reach the required concentration range. During this cooling, the separator gas bypass liquid line (see fig. 2.9) is opened to allow cold gas to cool-down the evaporator and to pre-cool the feed stream, the open valve at the same time also enhances the separator collection performance because of the higher pressure difference over the drainage pipe.

When enough liquid is collected and the set-up is cooled, the gas bypass steady-state (see fig. 2.6 and 2.9) is opened and the gas bypass start-up is closed, this is called the transition. After the transition it takes over an hour to get into steady state. Steady state is reached when gas concentrations, pressures, temperatures, liquid levels and mass flow rates are all stable.

Due to the loop design of the test rig, a stable steady state is necessary to give reliable measurements. The measurement errors therefore also relate to the loop stability. After the transition several settings have to be varied for the system to be able to reach a steady state. The system finds a steady state which can only be controlled slightly by varying some process settings. When the settings have to be varied too much, or the conditions before the transition are too far off, the system fails to become steady. It is because of this self found steady state, that the measuring
pressure and temperature can not be chosen arbitrarily. Because one can not intervene in the period where the system finds its steady state, it is difficult to measure at an specific pressure, temperature or concentration, and almost impossible to reach a stable steady state at a pre-defined combination of these variables.

2.3.5 Accuracy

There are three main sources of errors in these measurements: Loop stability, flow meters and concentration analyzers. The errors in the temperature and pressure measurements are very small. The error in the temperature measurements is in the order of ±0.2 °C. The error in the pressure is about ±0.01 bar. The stability of the loops steady state has been treated in section 2.3.4. The accuracy of the flow meters, mass spec analyzer and gas chromatograph will be explained in the next sections.

Bypass

In order to have reliable measurements, the bypass flow rate should be small compared to the feed-, product-, and waste-flow rates. The bypass concentration as well as the bypass flow rate could not be measured during some of the measurements, the measured Bypass/Feed mole flow rate percentage is defined as:

\[
B_m = \left( \frac{B(16b_1 + 44(1 - b_1))^{-1}}{F(16z_1 + 44(1 - z_1))^{-1}} \right) \times 100
\]

with \( B \) and \( F \) the mass flow rate of the Bypass and Feed stream, and \( b_i \) the concentration of the bypass stream where subscript 1 represents the concentration of methane and subscript \( m \) refers to measured.

From theory, the bypass/feed mole flow rate percentage \( B_t \) can be calculated according to theoretical concentrations (where the subscript \( t \) refers to theory):

\[
B_t = \left( \frac{F}{M_{wt}} - \frac{P}{M_{w,vap}} - \frac{(1 - P_{frac})}{M_{wt}} \right) \times 100
\]

With \( M_{wt}, M_{w,vap} \) the mole weight of the total flow and vapour stream and \( P_{frac} \) the product mole fraction. Equation (2.8) represents the mole flow rate of the feed minus the mole flow rate of the product minus the mole flow rate of the waste compared to the mole flow rate of the feed.

When calculated with the measured concentrations instead of the pure theoretical predicted concentration the bypass/feed mole flow rate percentage \( B_t \) changes into:

\[
B_{tm} = \left( \frac{F}{16z_{1m} + 44(1 - z_{1m})} - \frac{P}{16y_{1m} + 44(1 - y_{1m})} - \frac{FW_c(1-P_{frac})}{16z_{1m} + 44(1 - z_{1m})} \right) \times 100
\]

where \( tm \) refers to theoretical and measured input. \( W_c \) is a correction factor which corrects for the increased waste stream due to methane entrainment, which can be
described by:

\[ W_c = \frac{1 - x_{1f}}{1 - x_{1m}} \]  

(2.10)

These models have been validated using the measured values for the bypass flow rate and are in reasonable agreement with the measurements.

**Flow meters**

In the test rig four coriolis mass flow meters are present. The first a Promass 83m in the feed stream which has a typical mass flow rate of 50-100 kg/h. Coriolis mass flow meters give a well defined error when used with water, a commonly used pure substances. Because we have a binary mixture of varying compositions of methane and carbon dioxide (20-80 mole% CH\(_4\)) the error estimation becomes more difficult. Note that, the coriolis meters can not cope with two phase flows and that coriolis mass flow meters need a high pressure drop and gas speed to achieve a high accuracy. Because of the to high pressure drop of the previously advised meter, the Promass 83m has been installed which operates close to the lower limit of applicability and therefore has a greater error. For our tests we needed a pressure drop below 1 bar (corresponding to an internal velocity of 30 m/s) over the flow meter. This in combination with varying temperatures (0-+40 °C) and pressures (80-150 bars) makes error estimation difficult. Combining the above with information from the supplier an error of < ±5% is estimated.

The product flow (30-85 mole% CH\(_4\)) is metered with the an equal Promass 83m at lower pressure (25-35 bars), lower temperature (-30-+40 °C) and at smaller mass flow rates 30-70 kg/h. This also gives an estimated error of < ±5%.

The waste (0-+40 °C, 25-35 bars, 0-20 mole% CH\(_4\) and 0-30 kg/h) and bypass (-50-0 °C, 25-35 bars, 20-80 mole% CH\(_4\) and 0-30 kg/h) stream are smaller and the meters (RHM04-AGD-99-0 special CORI-FLOW) are operated closer to design conditions, the estimated errors are therefore < ±2%.

**Analyzers**

As mentioned before, two gas analyzers have been used, a quadrupole mass spectrometer and a four channel gas chromatograph. The errors of these devices depend on the manner of usage. Both devices are calibrated using pre-mixed bottles with a methane concentration of approximately: 0, 25, 50, 75, 100 mole%.

The mass-spec quadrupole is set to a specific setting which allows only ions with specific mass, e.g. 28 for nitrogen, to hit the detector. The mass-spec then counts the hits on the detector plate for this specific mass number. The pressure in the analyzer should be low enough (i.e. <1e-4 Pa) to make sure that the amount of hits does not saturate the detector. The mass-spec scans the range from 0-200 g/mole and stores al the counts. Because of the high voltage of the filament some molecules are partly destroyed, so besides CH\(_4\), CH\(_3\) (≈ 1%) and CH\(_2\) (≈ 0.01%) are also counted. Luckily the number of counts from destroyed molecules is low. Unfortunately some mass numbers can represent several species for example 28 which can represent nitrogen
(N₂) as well as carbon monoxide (CO). When most of the N₂ comes from air, one can correct (within the software) the number of CO by subtracting the N₂ with the help of the counts from oxygen (O₂).

\[ n_{CO} = n_{CO+N2} - n_{N2} \quad \text{with} \quad n_{N2} = \frac{78}{21}n_{O2} \quad (2.11) \]

where 78 and 21 are the molar percentages of N₂ and O₂ in air. When there is no CO present in the system these counts can be added to the counts of CO₂. Because there is a big difference in absolute number of counts for CO₂ and CH₄ calibration is necessary. On the number of counts measured during calibration, a linear regression analysis is performed to find a first order polynomial which converts number of counts into the corresponding concentration. By remeasuring the calibration gasses the average error can be estimated at approximately 2 mole%. The number of counts varies with time, pressure, temperature and moisture content within the analyzer. When the counts start to deviate to much, a new calibration should be performed.

The GC consists out of four thermally conditioned columns filled with a packing suited for methane carbon dioxide discrimination. The retention times for both gasses differ a few seconds. Helium is used as a carrier gas which is supplied at a constant flow rate and pressure. The sample is injected into the column by an injector. The gas flowing out of the column is passed along a Wheatstone bridge which measures the change in electrical resistance. This change is then depicted as a peak in a graph, the areas of the peaks are a measure for the concentrations. When the right packing is used a clear distinction can be made between the different gasses. The conversion into concentrations can again be calibrated with help of the pre-mixed bottles. The cumulative value i.e. the sum of the concentration of CH₄ and CO₂ should add up to 100% to be sure that the calibration is valid. Any deviation from 100% indicates a measurement error, e.g. a calibration float. The average error in the measurement is estimated at approximately 1-2 mole%.

### 2.3.6 Mass balance

To clarify the experimental results, one measurement day will be described here, and the mass balance will be resolved. In fig. 2.11 the concentrations plot of the day can be found. In table 2.1 the loop conditions during steady state can be found. The measured concentrations are in agreement with the theoretical predictions. The mass flow rates show an inconsistency of \( F - P - W - B \approx 1.9 \text{ kg/h} \). The deviation is less then 2.5% of the feed flow rate (77.5 kg/h).

When the theoretical bypass is calculated with equations (2.8) and (2.9) the bypass percentages equal: \( B_t \approx 15.8\% \) and \( B_{tm} \approx 15\% \). When the theoretical bypass is evaluated with the maximum errors in the mass flow meters, the bypass mole flow rate percentages range from: \( B_t \approx 9.1 - 21.8\% \) and \( B_{tm} \approx 8.3 - 21.1\% \).

This is in agreement with the actual "measured" value (eq.(2.7)) of bypass mole flow rate percentage \( B_m \approx 11.9\% \). As can be seen from the calculations, there is a large dependency within the calculated bypass percentage on the errors from the mass flow meters. The bypass percentage should be kept as small as possible, but at the same time large enough to ensure proper liquid drainage from the separator.
Experimental validation of the C3sep process at lab scale

Figure 2.11. Concentration measurement via Gas-chromatography

<table>
<thead>
<tr>
<th>variable</th>
<th>value</th>
<th>dimension</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pressure induction section</td>
<td>29.5</td>
<td>Bar (relative)</td>
</tr>
<tr>
<td>Temperature before separator</td>
<td>-30.1</td>
<td>°C</td>
</tr>
<tr>
<td>Temperature after separator</td>
<td>-27.7</td>
<td>°C</td>
</tr>
<tr>
<td>Separator rotational velocity</td>
<td>800</td>
<td>rpm</td>
</tr>
<tr>
<td>Feed flow rate</td>
<td>77.5</td>
<td>kg/h</td>
</tr>
<tr>
<td>Product flow rate</td>
<td>45.5</td>
<td>kg/h</td>
</tr>
<tr>
<td>Waste flow rate</td>
<td>21.5</td>
<td>kg/h</td>
</tr>
<tr>
<td>Bypass flow rate</td>
<td>8.6</td>
<td>kg/h</td>
</tr>
<tr>
<td>Feed concentration</td>
<td>38.8</td>
<td>mole% CH₄</td>
</tr>
<tr>
<td>Product concentration</td>
<td>48.3</td>
<td>mole% CH₄</td>
</tr>
<tr>
<td>Waste concentration</td>
<td>7.41</td>
<td>mole% CH₄</td>
</tr>
<tr>
<td>Bypass concentration</td>
<td>46.7</td>
<td>mole% CH₄</td>
</tr>
<tr>
<td>Predicted Product concentration</td>
<td>47.35</td>
<td>mole% CH₄</td>
</tr>
<tr>
<td>Predicted Waste concentration</td>
<td>6.55</td>
<td>mole% CH₄</td>
</tr>
</tbody>
</table>

Table 2.1. Loop conditions during steady state
2.4 Results and discussion

When performing a measurement, the unit should be in “steady state” to eliminate the fluctuations which result from the separation loop. The gas that is separated from the liquid droplets, is fed back to the compressor much faster than the liquid CO$_2$, which remains a relatively long time within the liquid collection vessel (see fig. 2.9). The long time delay to measurement arises from CO$_2$ liquid hold up. The density of the liquid CO$_2$ is much higher than the density of the gas and the volume of liquid path is much larger due to the collection vessel. The cycle time of the liquid CO$_2$ can be more than 20 min which results in a waiting time of more then 1 hour to achieve steady state. At start-up, liquid CO$_2$ is collected in the liquid collection vessel to be sure that there is a liquid lock present during steady state operation. This liquid lock is needed to prevent bypass gas from contaminating the liquid CO$_2$.

A typical concentration log measured via the GC can be found in fig. (2.12). The

![Figure 2.12](image)

**Figure 2.12.** Simultaneous measurement of feed, product, waste and bypass concentration via gas-chromatography. Operating pressure 31.8 (bara) and temperature -48 °C

figure shows a continuous concentration plot for all four streams simultaneously. The
four lines represent the methane concentration in mole percent of the feed, product, bypass and waste stream. The first 50 sample numbers are during start-up and cooling phase where liquid CO$_2$ is collected in the liquid collection vessel. When enough liquid is collected and the set-up is cooled, the gas bypass steady-state (see fig. 2.6 and 2.9) is opened and the gas bypass start-up is closed this is the transition. After the transition it takes over an hour to get into steady state. Steady state is reached when gas concentrations, pressures, temperatures, liquid levels and mass flow rates are all stable. In fig. 2.12 a downward spike in waste concentration is recorded that represents an aborted transition which has not led to a steady state. The second transition around sample number 80 has been performed successfully. After sample number 160 the system has reached steady state. The graph shows a stable feed concentration of 68% methane, a product concentration of 72% and a liquid waste stream which consists out of 11% methane. The bypass concentration is close to the feed concentration. Due to the relatively high temperature only a small increase in methane concentration is measured. Theoretical flash calculations at the operating pressure and temperature predict a product concentration of 72% which is in agreement with the measurements. At the measuring conditions, 99% of the methane present in the feed stream is recovered in the product stream.

Figure 2.13 shows the results taken from a number of runs such as that shown in fig. 2.12. The measured product concentration of methane $y_1$ is plotted as function of the measured feed concentration of methane $z_1$, these measurements have been done at several pressures and temperatures varied independently. The 45° line represents a product concentration $y_1$ equal to the feed concentration $z_1$ ($z_1 = y_1$) which is equal to no enrichment and 100% recovery. The plotted points represent the product concentration $y_1$ for several measured feed concentrations $z_1$ at various pressures and temperatures. The error bars reflect the stability of the steady state during measurement and the analyzer reproducibility.

Due to the operating limitations of the loop (see fig. 2.3) only a shallow penetration of the phase diagram can be accomplished, i.e. the conditions after expansion are still close to the dew-point line. Due to the shallow penetration of the phase diagram, all measurements have been performed at low enrichment and high recovery conditions. A table containing the most important measurement data can be found in appendix A. Because of the pressure and temperature limitations of the test set-up (mainly due to the compressor pressure ratio and small scale expansion) the operation window is limited. One of these limitations is the isenthalpic expansion over the JT-valve which results in less cooling for a certain pressure drop than the isentropic cooling via a turbine expander. The pressure in the separation section may be varied between 25-35 bar, this in combination with the isenthalpic expansion, limits the operating window (see fig. 2.3). The operation window is constrained in vertical direction between the minimal and maximal operating pressure of the loop. In the horizontal direction the operation is limited between the dew-point line and the isenthalpic expansion curve corresponding to the JT-valve, see fig. 2.3. The measurements that are presented in this chapter are therefore mainly done at pressures and temperatures that are non-optimal for highest enrichment at maximum recovery. Operation can only be performed in a small area of the phase diagram located in the lower right of the two
2.4 Results and discussion

![Graph showing measured product concentration versus measured feed concentration. The line x=y is plotted to guide the eye. The three plotted curves are the theoretical maximum achievable enrichments for a stated recovery rate.](image)

**Figure 2.13.** Measured product concentration versus measured feed concentration. The line x=y is plotted to guide the eye. The three plotted curves are the theoretical maximum achievable enrichments for a stated recovery rate.

Phase envelope. This limited operation window is an artefact of the loop construction. In practice this means that operation with this test rig is limited to high recovery and low enrichment conditions. Because these limitations are present, we can only validate model predictions at non-optimal conditions. Due to the shallow penetration of the phase diagram, all measurements have been performed at low enrichment and high recovery conditions.

The plotted lines corresponding to a recovery of $r_1 = 0.9$, $r_1 = 0.95$ and $r_1 = 0.98$ are the theoretical maximum achievable enrichments for these stated recovery rates. Incorporating a limited maximum initial pressure prior to expansion (van Wissen [5]). Note that the measurements do not, and should not relate to any of the plotted recovery lines directly because they are performed at different pressures and temperatures. These lines represent theoretical maximum achievable enrichments for
a stated recovery rate (eq.(2.1)) in combination with the prior stated restrictions. This predicted optimal performance can not be validated with the current test loop due to the stated pressure and temperature restrictions. The recoveries corresponding to the presented measurements vary between 93-100%.

The separation performance is determined by the gas analyzer. When in a stable steady state, the rotational velocity of the separator is changed, then the change in concentration, mass flow rates and pressures are monitored. Due to the small scale of the separator, rotational variations have a limited effect on the already high separation efficiency. This is exemplified by the high efficiency of the pre-separator even at static conditions. In the case of real high rotational velocities, the separation efficiency gets worse due to the additional power supplied to the element, and to the small gas stream and thereby increasing the temperature and the evaporation of liquid carbon dioxide. To achieve good separation, a bypass line was needed to enhance the liquid drainage from the separator. Therefore special care should be given to the design of the liquid removal from future separators.

Due to the small flow rates (50 kscf/d) in the test-loop, it is fairly easy to separate micron sized droplets. However, at full scale this will be one of the major challenges due to the decreasing performance at larger scales at constant energy consumption [12].

In fig. 2.14, a comparison is made between the, predicted and observed product concentrations \(y_1\) corresponding to a measured feed concentrations \(z_1\) shown in fig. 2.13. As can be seen, the predictions and measurements are in good agreement. Because the theory and measurements agree well, one can conclude that the two phases are separated almost completely. So preferential condensation and subsequent centrifugal separation of the condensed phase is possible. To measure at conditions leading to maximum enrichment, a different test rig is required. This rig should be build at a larger scale to incorporate the centrifugal separation testing. The enrichments and recoveries presented in this chapter are representative for the real, once through process, and are in good agreement with the thermodynamic predictions. Therefore one can conclude that when complete separation of the condensed phase is achieved on full scale, the predicted performance will be achieved.

2.5 Nucleation and coagulation experiments

Droplet growth due to condensation in general consists of three competing processes:
• homogeneous condensation
• heterogeneous condensation
• coagulation
of which the first two really withdraw molecules from the vapour phase whereas the latter only recombines previously formed droplets.

Due to e.g. expansion cooling the pressure will become higher than the saturation pressure of the mixture, this state is called supersaturation. The supersaturation ratio \(S\) is defined as:

\[
S = \frac{p}{p_s(T)}
\]  (2.12)
2.5 Nucleation and coagulation experiments

Figure 2.14. Comparison between the predicted and measured concentrations

with $p$ the pressure and $p_s(T)$ the saturation pressure of the gas mixture. When $S > 1$ condensation can occur. The condensation can take place on the walls or dispersed particles in which case the condensation is called heterogeneous condensation. When there are no particles or walls to condense on, clusters of the condensing component will form when the supersaturation ratio is high enough, this is called homogeneous nucleation. The subsequent condensation can take place on the formed clusters and walls as heterogeneous condensation or when the cooling again leads to high enough supersaturation, homogenous condensation will re-occur.

The formed droplets are dispersed within a gas flow and due to the random motion the droplets will collide and merge into bigger droplets. This process is called coagulation. It continues even if no additional condensation takes place. Due to the decreasing mobility for larger droplets combined with the diminishing particle concentration this process decelerates in time see fig. 2.15.

We have done some measurements on droplet formation and growth in the test...
Figure 2.15. Droplet diameter evolution calculated with monodisperse coagulation theory [5].

setup described in the beginning of this chapter. The measurements have been performed with the help of an optical reflectance measurement probe (ORM probe) provided by MTS (Meßtechnik Schwartz). The system measures the chord length of a droplet by measuring the time that the light reflection by the droplet exceeds a certain threshold. This time is in combination with the fluid speed a direct measure for the droplet chord length. When these chord lengths are corrected for biased sampling [23] towards larger droplets and converted via diameters to volume the diagram shown in fig. 2.16 can be constructed. The figure shows that for a mixture with 80 mole% CH\textsubscript{4} the droplet size typically varies between 2-15 µm. Approximately fifty percent of the liquid volume is contained by droplets smaller then 6 µm. If one would like to fully exploit the possible enrichments with C3sep, a phase separator which is capable of collecting almost all droplets down to a size as small as 2 µm is inevitable.

When more CO\textsubscript{2} is present in the gas, the droplets are bigger. A larger variety in droplet size is expected due to coagulation but this large increase could indicate wall influences. Wall condensation could explain the larger variations in droplet size as well as the very large droplet sizes that are measured. Re-entrainment of condensed liquid could be the reason for the much larger droplet sizes at elevated CO\textsubscript{2} concentrations. Although the droplet sizes are considerably larger, for efficient operation, droplets as small as 3 µm still need to be separated with a reasonably high efficiency.

Due to the large surface to volume ratio of the piping, which is an artefact of the small flow rates, wall condensation can rapidly decrease the amount of condensable matter. Especially in the small induction piping used in this test setup, the huge wall area that is present in combination with the small diffusion length make wall
condensation a factor that can be important. For wall condensation to occur the walls need to be colder than the condensation temperature of the contaminants. This could be the case when supersaturation driven condensation, is substantial, and few other particles are present.

To estimate whether wall condensation can have a significant influence, the laminar diffusion to the wall is estimated when the walls would have a theoretical equivalent concentration of \( c_{wall} = 0 \). The change of the concentration in time can then be estimated by:

\[
\frac{dc}{dt} = D \frac{d^2 c}{dr^2} \quad \frac{1}{t} \sim \frac{D}{a^2}
\]  

(2.13)

with \( D \) the gas diffusion coefficient, \( a \) the radius of the pipe and \( t \) the characteristic time. This results in a characteristic time for the diffusion process of approximately:

\[
t \geq \frac{a^2}{D}
\]  

(2.14)

with the smallest typical radius of the induction section \( a = 0.005 \) m, and a diffusivity of \( D \approx 10^{-6} \) m\(^2\)s\(^{-1}\) (note that \( \rho D \approx \) constant). This results in a characteristic wall
condensation time of 25 s, about 50 times the residence time in the induction section. From this it can be concluded that laminar diffusion alone is not able to bring the condensable vapour close enough to the wall to seriously affect the concentration.

Because the flow in the induction section is turbulent, the turbulent velocity fluctuations could enhance the vapour transport to, and thereby the vapour deposition, on the wall. Calculations indeed show the possibility that approximately 40% of the condensable species deposit on the walls of the induction section (see appendix C).

2.6 Thermodynamic conditions and waste recuperation

The optimal pressure and temperature conditions to perform the phase separation have been investigated. In general the following can be said, the phase equilibrium is most favorable at low temperatures, therefore the phase separation should be performed close to the freeze out curve of CO$_2$ which is a sloping near vertical line located around -60 °C. Depending on the required recovery the pressure can then be increased to gain a higher purity gas $y_1$ or decreased to increase the recovery of methane ($r_1$).

As previously described by van Wissen [5] H$_2$S (which is often present in a gas field) works as an antifreeze for CO$_2$ thereby releasing the opportunity to get up to higher purities of methane $y_1$ without sacrificing the achieved recovery $r_1$ by operating at temperatures as low as -80 °C (see fig. 2.17). Any other component with a similar effect on the freeze out curve will probably have the same advantages.

Figure 2.17. Phase diagram corresponding to CH$_4$(1) + CO$_2$(2) + H$_2$S(3): $z_1 = 0.8$, $z_2 = 0.14$ and $z_3 = 0.06$. An extended equation of state program based on a cubic equation of state of the Soave-Redlich-Kwong type with pure component parameters fitted to vapour pressures and liquid densities along with a composition dependent mixing rule is used. A freeze out model $T < T_{solid}$ of CO$_2$ is incorporated (green curve).
When operation within the vapour liquid solid area is possible, this leads to a more favorable equilibrium, which results in a higher purity gas (up to 95%) and liquid. So when the separation system can be designed to operate within this three phase area, a considerable gain in separation efficiency can be accomplished, without the presence of the health and safety issues related to $\text{H}_2\text{S}$.

The optimal separation conditions always involve a trade off between required recovery and possible enrichment. The higher the recovery, the less enrichment occurs and the higher enrichment is needed the more methane is lost with the liquid $\text{CO}_2$.

When a waste stream methane recuperation is applied, the optimal point shifts to higher separation pressures and therefore higher enrichments and a higher purity product gas (see fig. 2.18). The liquid that is separated from the carrier gas will then contain relatively high concentrations of methane (see table D.1). This methane can be recovered in the waste recuperation step. If the by the recuperation system produced methane purity is not high enough to mix with the product gas, the produced stream can be fed back to the inlet of the C3sep system. The mixing of the two stream prior to the C3sep process is called “asymmetric cascading”. Waste recuperation in combination with asymmetric cascading can provide optimal purity of the methane stream whilst reducing the methane losses via the liquid stream.

![Figure 2.18](image)

**Figure 2.18.** Maximum enrichment ($y_1$) as function of feed fraction ($z_1$). For reference the maximum enrichment ($y_1$) at fixed recovery rates ($r_1 = 0.9$, $r_1 = 0.95$ and $r_1 = 0.98$) are plotted.
2.7 Conclusion

In this chapter we have demonstrated for the first time, that condensed contaminant centrifugal separation (C3sep) can be used to clean contaminated natural gas containing large quantities of CO₂. The process combines expansive cooling along with a novel rotational particle separator (RPS). Use is made of the preferential condensation of CO₂ and subsequent mechanical separation of the condensed phase.

The separated waste stream is produced as a liquid - unlike in other processes where expensive recompression is required. This greatly reduces the costs as the waste stream liquid can simply be pumped to a sequestration and storage facility (such as return to the subsurface).

The unique advantage of this device is that it can remove much smaller droplets (c.a.1 µm) compared to other mechanical rotational separations such as cyclones (20 µm). In a process requiring cooled conditions, this means that separations are effected after much shorter residence times with accordingly much shorter lengths of cooled pressurised piping. This significantly cuts capital costs.

In this demonstration test, only a limited region of the phase diagram could be accessed because we were operating with a test loop rather than a once through system as would be the case under field conditions. The smaller scale (50 kscf/d) compared to field conditions (1000 MMscf/d) meant that a Joule-Thomson valve had to be used rather than a turbine expander. In the latter case, more effective cooling will be achieved and there will also be energy recovery for application in any subsequent compression.

Because the operating window of the setup is optimized for gas mixtures of 50 mole% methane, the flexibility of the setup decreases for other concentrations. With high CO₂ concentrations, the shift of the critical point to higher temperatures and the smaller two phase area within the phase diagram make it difficult to penetrate the phase diagram deep enough by expansion cooling performed with a JT-valve.

Once the CO₂ droplets have been collected as bulk liquid, a number of liquid drainage issues require special attention. Liquid hold up can suppress efficiency due to re-entrainment of liquid into the gas stream. This will also be a feature of a once through system.

The process is a good bulk separator of natural gas contaminants, however a tail gas treater such as an amine unit will always be required for the last few percent of contaminant to bring the gas up to pipeline specifications. For local use or electricity production the product can be used directly without any after treatment.

In this system, for safety reasons, we have only tested natural gas contaminated with CO₂. However the same separation principles apply for natural gas contaminated with H₂S. Indeed more efficient separations should be achievable using somewhat lower temperature conditions (e.g. -90 °C) which is possible due to the much lower solid line in the corresponding phase diagram. Operation in the solid region of the phase diagram result in a considerable gain in separation efficiency, without the presence of the health and safety issues related to H₂S.

The optimal separation conditions always involve a trade off between required recovery and possible enrichment. The higher the recovery, the less enrichment occurs and the higher enrichment is needed the more methane is lost with the liquid CO₂.
When a waste stream methane recuperation is applied, the optimal point shifts to higher separation pressures and therefore higher enrichments and a higher purity product gas. Waste recuperation in combination with asymmetric cascading can provide optimal purity of the methane stream whilst reducing the methane losses via the liquid stream.

Further development of this technology will, for the reasons identified above, be focussed on larger scale once through processes using slip streams from field gas reservoir facilities. For a robust separator design certain aspects have to be studied in more detail. To determine the optimal flow direction within the RPS element a theoretical analysis of the liquid CO$_2$ film is performed in the next chapter 3.
Experimental validation of the C3sep process at lab scale
Chapter 3

Creeping film model for the RPS element*

3.1 Introduction

In the C3sep process the RPS element is loaded with a considerable amount of liquid (ca. 10 vol%). In order to investigate whether the element will be blocked by the liquid films in the channels the film behavior is analyzed theoretically. The influence of the filter orientation on the liquid film, is simultaneously investigated.

The behavior and thickness of the liquid film in the RPS channels depend on several parameters, e.g. liquid loading, dimensions of the channels, fluid density, viscosity and the magnitudes of the different forces involved. For the design of the RPS, this behavior can be very important. Depending on the nature of the film in the channels, the fluid flows either in the upstream or downstream direction, breaks up into larger droplets and is collected.

Many researchers have worked on film behavior, mainly focusing on the heat transfer between a cold plate and a condensing saturated vapour. Nusselt [24] was the first to derive an analytical relation for the film thickness on a vertically orientated plate. Sparrow and Gregg [25] extended the theory with inertia effects. Chen [26] extended the model of Sparrow and Gregg by considering drag forces at the interface between the vapour and the liquid film. Leppert and Nimmo [27] used a Nusselt type analysis to investigate condensation on a surface perpendicular to inertial forces, analogous to centrifugal forces. Another study was conducted by Bejan [28], who derived a relation for the film thickness on an upward facing plate with free edges. According to Bejan [29; 30] the flow can be treated as a boundary layer type when the film thickness is relatively small. Sparrow and Gregg [31] looked at laminar film condensation on a rotating disk. Considerable research has been done on the stability of liquid films in rotating gas/liquid systems, like cyclones. Re-entrainment of fluid into the gas in

separators for the offshore oil and gas industry was studied by Swanborn [32]. However, most of the research described above deals with low gas velocities, and, as a consequence, small surface shear forces. In our case we deal with large flow rates and compact equipment which results in high flow velocities and high shear forces.

Mondt [10] was the first to present an investigation on film behavior in a RPS-like situation. In this work the channels are replaced by a concentric ring configuration. The work covers vertically as well as horizontally mounted rotating channels. Shear forces acting on a vertical film and pressure drop over the channel were not included.

This chapter presents a model for fluid film behavior in a vertically oriented RPS under the combined influence of centrifugal and gravitational forces. Because of the high gas velocities involved in the process, shear forces at the interface between the gas flow and the liquid film, and the resulting pressure drop over the channel, are considered as well. The gas velocity through the channels is corrected for the displacement effect of the liquid film.

This chapter focusses on the film behavior within the RPS separation channels. Section 3.2 describes the problem and introduces the geometry. In section 3.3 the film equations are derived that are solved in section 3.4. The model and derived conclusion are discussed in section 3.5.

3.2 Problem definition

A gas flowing through a centrifugal separator is being subjected to a centrifugal force. As a result, liquid droplets entrained in the gas flow are forced towards the outer walls, forming a liquid film, and are thus separated from the gas flow. A very efficient type of centrifugal separator is the rotating phase separator (RPS) [16] introduced in section 1.3. The core component of the RPS is a rotating RPS element consisting of a multitude of axially oriented channels, which rotate as a whole around a common axis (fig. 3.1). The rotating element of the RPS can be constructed in various ways. Normally, it is manufactured by winding up layers of corrugated and non-corrugated steel around a shaft and welding these layers together to form a rigid element.

For modelling purposes, a simplified filter geometry is considered, identical to the geometry used by Mondt [10]. A channel is modeled by an annulus formed by two concentric cylinders. In practical installations, radial walls in the annulus stabilize the flow in case of rotation. It is therefore assumed that the liquid film and the gas flow with entrained droplets co-rotate with the RPS element. In fig. 3.1, a schematic representation of an annulus with a liquid film is given. The channel height \( h \) is assumed to be small compared to the radius \( r \) of the annulus.

In fig. 3.2, a liquid film along the outer wall of a channel of length \( L \) is given. The film is subjected to gravity, centrifugal force and shear force induced by the gas flow. The flux of droplets to the liquid film is assumed to be homogeneous with a constant velocity \( v_0 \). This will generally not be true in practice, since the probability of the position of deposition for a droplet depends on its size. The resulting distribution of influx to the liquid film can however be calculated accurately for practical situations and used to adapt the results of this theory for liquid film build-up in a straightforward manner.
3.2 Problem definition

Figure 3.1. Annulus of a RPS element with a liquid film.

Figure 3.2. Liquid film in an annulus subject to gravity, centrifugal- and shear forces.
The liquid film is assumed to be slender, with film thickness $\delta$ being much smaller than the channel length. It can therefore be regarded as a boundary-layer type flow. The displacement effect of the liquid film on the gas flow is apparent from the gas velocity $v_g$, which depends on the local film thickness as

$$v_g = v_{g,0} \left( \frac{h}{h - \delta} \right),$$

where $v_{g,0}$ is the gas velocity in case there is no liquid film. This approximation is valid if $h \ll r$.

### 3.3 Film equations

#### 3.3.1 Boundary layer type flow

The steady incompressible flow in the liquid film is generally described by conservation of mass

$$\nabla \cdot \vec{v} = 0,$$

and conservation of momentum

$$(\vec{v} \cdot \nabla) \vec{v} = -\frac{1}{\rho_l} \nabla P + \nu \nabla^2 \vec{v} + \vec{g},$$

where $\vec{v}$ is the fluid velocity vector, $\rho_l$ the fluid density, $P$ the pressure in the liquid film, $\nu$ the kinematic viscosity, and $\vec{g}$ the gravitational acceleration. The thickness $\delta$ of the liquid film is considered very small compared to the radius $r$ of the cylinder wall, so that the effect of cylinder curvature can be neglected. The axisymmetric fluid flow problem can then be described in Cartesian coordinates. Conservation of mass can thus be written as

$$\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} = 0$$

For a slender liquid film ($\delta \ll L$), undergoing solid body rotation, the momentum equations reduce to

$$u \frac{\partial u}{\partial x} + v \frac{\partial u}{\partial y} = -\frac{1}{\rho_l} \frac{\partial P}{\partial x} + \nu \frac{\partial^2 u}{\partial y^2} + g,$$

in $x$-direction, and

$$0 = -\frac{1}{\rho_l} \frac{\partial P}{\partial y} - \Omega^2 r,$$

in $y$-direction, where $u$ and $v$ are velocity components in $x$- and $y$-direction respectively, and $\Omega^2 r$ the centrifugal acceleration, with $\Omega$ the angular velocity and $r$ the radius which can be considered a constant since it was assumed that $\delta \ll r$.

Mondt [10] shows that the Reynolds number in the film is usually small enough to allow for a creeping flow approximation, i.e. the inertia effects are small compared to the effects of viscosity. To illustrate this, consider the inertia and the friction terms in $x$-direction:

$$\text{inertia} = u \frac{\partial u}{\partial x} + v \frac{\partial u}{\partial y} \quad \text{and} \quad \text{friction} = \nu \frac{\partial^2 u}{\partial y^2}$$

(3.7)
From eq. (3.4) it follows that the velocity $u$ in $x$-direction scales according to

$$u \propto \frac{v_0 L}{\delta},$$  

(3.8)

with $v_0$ the velocity of the liquid influx to the film, $L$ the length of the channel, and $\delta$ the film thickness. This means that both inertia terms are of the same order

$$u \frac{\partial u}{\partial x} \propto v \frac{\partial u}{\partial y} \propto \frac{v_0^2 L}{\delta^2}$$  

(3.9)

and the friction term is of order

$$\nu \frac{\partial^2 u}{\partial y^2} \propto \frac{\nu v_0 L}{\delta^3}.$$  

(3.10)

Combining the above equations results in an expression for the ratio of the inertia and friction terms:

$$\frac{\text{inertia}}{\text{friction}} \propto \frac{v_0^2 L/\delta^2}{\nu v_0 L/\delta^3} = \frac{v_0 \delta}{\nu} \frac{\delta}{L} = \frac{Re_\delta \delta}{L},$$  

(3.11)

with $Re_\delta$ the local Reynolds number based on the thickness of the liquid film. Although $Re_\delta$ may grow to $\sim 10^2$ and still remain laminar, practical values of $\delta/L$ in the range of $10^{-4} - 10^{-3}$ are small enough to neglect the inertia terms from eq. (3.5), which then reduces to

$$0 = -\frac{1}{\rho_l} \frac{\partial P}{\partial x} + \nu \frac{\partial^2 u}{\partial y^2} + g.$$  

(3.12)

### 3.3.2 Fluid-gas coupling

The pressure gradient in $x$-direction in eq. (3.12) can be determined by equating the pressure $P$ in the liquid to the pressure $P_g$ of the gas flow at the interface between the two media. The liquid pressure $P$ follows from integrating eq. (3.6):

$$P(x, y) = -\rho_l \Omega^2 r y + K_1(x)$$  

(3.13)

with $K_1$ an integration constant possibly depending on position $x$. Using $P(x, \delta) = P_g(x)$ as a boundary condition, $K_1$ can be determined as

$$K_1(x) = \rho_l \Omega^2 r \delta + P_g(x).$$  

(3.14)

The relation for the pressure in the liquid film then becomes

$$P(x, y) = \rho \Omega^2 r (\delta - y) + P_g(x).$$  

(3.15)

The pressure distribution in the gas flow depends on local film thickness and gravity- and shear forces, and can be determined as follows. Consider a ring-shaped element of gas with a steady bulk flow velocity $v_g$ (fig. 3.3). Equilibrium of forces in positive
x-direction resulting from pressure difference, gravity and shear for this volume of gas, yields

\[ [P_g A_1 - (P_g + dP_g) A_1] + \rho_g V g - \tau A_2 = 0 \]  \hspace{1cm} (3.16)

with \( \rho_g \) the gas density, and

\[ A_1 = 2\pi r (h - \delta) \]  \hspace{1cm} (3.17)
\[ A_2 = 4\pi r dx \]  \hspace{1cm} (3.18)
\[ V = A_1 dx \]  \hspace{1cm} (3.19)

where use is made of \( h \ll r \) and \( d\delta/dx \ll 1 \). The shear stress \( \tau \) acts on the inside as well as on the outside wall of the annulus, and is defined as the stress exerted by the gas on the wall. Thus, the shear stress has a positive value for a gas flow in downward direction. The velocity of the liquid film is assumed to be small compared to the gas velocity and the friction factors of both the gas-solid interface, and the gas-liquid interface are assumed to be equal. When reducing \( dx \) to zero, it follows that

\[ \frac{dP_g}{dx} = -\frac{2\tau}{h - \delta} + \rho_g g. \]  \hspace{1cm} (3.20)

The gradient in \( x \)-direction of the pressure in the film can now be found from equations (3.20) and (3.15) as

\[ \frac{\partial P}{\partial x} = \rho_l \Omega^2 r \frac{d\delta}{dx} - \frac{2\tau}{h - \delta} + \rho_g g. \]  \hspace{1cm} (3.21)

By substituting eq. (3.21) into eq. (3.12), the \( x \)-component of the momentum equation is found

\[ \nu \frac{\partial^2 u}{\partial y^2} = \Omega^2 r \frac{d\delta}{dx} - (1 - \frac{\rho_l}{\rho_i}) g - \frac{2\tau}{\rho_i(h - \delta)}. \]  \hspace{1cm} (3.22)

### 3.3.3 Film thickness

The velocity distribution \( u(x, y) \) in the liquid film can be solved from eq. (3.22) using the following boundary conditions:

- no-slip condition at the solid wall

\[ u = 0 : \quad y = 0 \]  \hspace{1cm} (3.23)
- shear stress $\tau$ at the liquid-gas interface [33],

$$\frac{\partial u}{\partial y} = \frac{\tau}{\nu \rho_l} \quad : \quad y = \delta$$  \hspace{1cm} (3.24)

With these boundary conditions the velocity profile in the film is given by

$$u = \frac{1}{2 \nu} \left[ y^2 - 2y \delta \right] - \frac{\tau g}{\rho_l} \left[ 1 - \frac{\rho_g}{\rho_l} \right] g - \frac{2\tau}{\rho_l (h - \delta)} + \frac{\tau}{\nu \rho_l} y.$$  \hspace{1cm} (3.25)

When this velocity is integrated over the film thickness, a volume flow rate $U$ per unit depth is found:

$$U(x) = \int_0^\delta u \, dy = \frac{1}{3 \nu} \delta^3 \left[ \left( 1 - \frac{\rho_g}{\rho_l} \right) g - \frac{\Omega^2 r}{\nu L} \frac{d\delta}{dx} + \frac{2\tau}{\rho_l (h - \delta)} \right] + \frac{\tau}{2\nu \rho_l} \delta^2.$$  \hspace{1cm} (3.26)

In case the liquid film is only subjected to gravity, it is clear from this equation that the volume flow rate $U$ in the liquid film is positive, i.e. in downward direction, at each station $x$ along the channel. As a result, all the liquid will leave the channel at the bottom. When also centrifugal- and shear forces act on the film, the flow rate may be in upward direction along at least part of the channel, depending on the relative force magnitudes, the direction of the gas flow and the shape of the liquid film. Consider a general case, in which liquid leaves the channel both at the top and at the bottom. For a steady state condition this means that there is one unique position $x = x_0$ where the flow rate $U$ changes sign (fig. 3.4). As a result, the flow rate $U$ can be written as

$$U(x) = v_0 (x - x_0).$$  \hspace{1cm} (3.27)

with $v_0$ the feed velocity and $x_0$ the inflection position.

Substituting eq. (3.26) into eq. (3.27) results in a differential equation for the film thickness $\delta$ as a function of position $x$ along the channel

$$\frac{\Omega^2 r}{3 \nu} \delta^3 \frac{d\delta}{dx} + v_0 (x - x_0) - \frac{\tau}{2\nu \rho_l} \delta^2 - \frac{1}{3 \nu} \delta^3 \left[ \left( 1 - \frac{\rho_g}{\rho_l} \right) g + \frac{2\tau}{\rho_l (h - \delta)} \right] = 0.$$  \hspace{1cm} (3.28)

To assess the relative importance of gravity, shear- and centrifugal forces for the characteristics of the liquid film, eq. (3.28) is made dimensionless as follows

$$\delta = \frac{\delta}{\delta_0} \quad ; \quad \hat{h} = \frac{h}{\delta_0} \quad ; \quad \bar{x} = \frac{x - x_0}{L}.$$  \hspace{1cm} (3.29)

with $\delta_0$ the maximum film thickness, resulting in

$$A \hat{\delta}^3 \frac{d\hat{\delta}}{d\bar{x}} + \bar{x} + B \hat{\delta}^2 + C \hat{\delta}^3 + D \hat{\delta}^2 = 0.$$  \hspace{1cm} (3.30)

with

$$A = \frac{\Omega^2 r \delta_0^4}{3 \nu v_0 L^2} \quad \quad B = - \frac{\tau \delta_0^2}{2 \nu v_0 \rho_l L} \quad \quad C = - \frac{g \delta_0^3}{3 \nu v_0 L} \left( 1 - \frac{\rho_g}{\rho_l} \right) \quad \quad D = - \frac{2\tau \delta_0^2}{3 \nu v_0 \rho_l L (h - \delta)}.$$  \hspace{1cm} (3.31)
The relative importance of the centrifugal term can then be estimated by the following ratios

\[
\frac{\text{centrifugal}}{\text{gravity}} \sim \frac{|A|}{C} = \frac{\Omega^2 r \delta_0}{(1 - \frac{v_w}{v_l}) g} \cdot \frac{\delta_0}{L} \tag{3.32}
\]

\[
\frac{\text{centrifugal}}{\text{shear}} \sim \frac{|A|}{B + D} = \frac{2\Omega^2 r \rho_l \delta_0}{\tau} \cdot \frac{\delta_0}{L} \cdot \frac{\hat{h} - \hat{\delta}}{3\hat{h} + \hat{\delta}} \tag{3.33}
\]

In the majority of practical installations these two ratios are much smaller than unity, indicating that the centrifugal term, i.e. the first term in eq. (3.28), can be disregarded. The relative importance of the centrifugal force is further reduced by the fact that \(d\delta/dx\) is very small throughout the liquid film, apart from a relatively small section at the start of the film (so-called tip region), where the boundary layer approximation is not valid. This result shows that, although the centrifugal force is the driving force behind the precipitation of droplets in the RPS, it has negligible influence on the characteristics of the liquid film and the way in which the film eventually leaves the channels. Neglecting this term will lead to a small outward oriented flow at the beginning of the film unaccounted for.

The shear stress \(\tau\) acting on the liquid film can be written as

\[
\tau = f \frac{1}{2} \rho_g v_g^2
\]

Assuming a highly turbulent gas flow, the interfacial friction coefficient \(f\) is approximately constant so that \(\tau\) is proportional to the square of the local gas velocity \(v_g\). When using eq. (3.1), the shear stress can be written as

\[
\tau = \tau_0 \left( \frac{h}{h - \delta} \right)^2 \tag{3.35}
\]
where $\tau_0$ is the shear stress in the absence of a liquid film. Expression (3.35) is used to non-dimensionalize eq. (3.28) conveniently by

$$
\bar{\delta} = \frac{\delta}{\phi} ; \quad \bar{h} = \frac{h}{\phi} ; \quad \bar{x} = \frac{x - x_0}{L}
$$

(3.36)

where

$$
\phi = \left| \frac{3\tau_0}{2g\rho_l} \right| .
$$

(3.37)

Thus, disregarding centrifugal force, eq. (3.28) can be written in non-dimensional form as

$$
\bar{\delta}^3 \pm \left( \frac{\bar{h}}{\bar{h} - \bar{\delta}} \right)^2 \left( \frac{4\bar{\delta}}{3(\bar{h} - \bar{\delta})} + 1 \right) \bar{\delta}^2 = \frac{\psi L}{\phi^3} \bar{x},
$$

(3.38)

where the gas density $\rho_g$ is assumed negligible compared to the liquid density $\rho_l$, and $\psi = 3\nu \nu_0/g$ represents the feed term. In this expression the plus and minus sign represent a gas flow in downward and upward direction, respectively.

### 3.4 Solution

In this section eq. (3.38) is solved for the two cases of gas flowing in upward and downward directions through the annulus. The different flow regimes and film characteristics will be discussed.

#### 3.4.1 Downward flow

The simplest case is when the gas flow is in downward direction. Both gravity and shear forces act in the same direction on the liquid film. The flow inflection point $x_0$ has moved to $x = 0$, which means that all the fluid is expelled at the downstream side of the channel. The fluid velocity in $x$-direction is positive, i.e. in downward direction, as is apparent from eq. (3.25) when the centrifugal force term is neglected. Velocity profiles and film build-up will resemble the one depicted in fig. 3.2.

In the downward flow case the equation for the film thickness $\delta$ is given by

$$
\bar{\delta}^3 + \left( \frac{\bar{h}}{\bar{h} - \bar{\delta}} \right)^2 \left( \frac{4\bar{\delta}}{3(\bar{h} - \bar{\delta})} + 1 \right) \bar{\delta}^2 = \frac{\psi L}{\phi^3} \bar{x}, \quad 0 < \bar{x} < 1
$$

(3.39)

the solution of which is given in a graphical manner in fig. 3.5 for different values of $\bar{\delta}$.

When comparing different curves, one should note that parameters are scaled variables. The range in $\bar{\delta}$ and $\psi L \bar{x}/\phi^3$ depends on the value of $\phi$, as does $\bar{h}$. To illustrate this, a shaded area is given in fig. 3.5 which bounds the region where creeping film theory applies for a specific case, i.e. $Re_\delta \leq 100$ and $\delta/L \leq 10^{-3}$.

In case the film thickness is much smaller than the channel height, eq. (3.39) simplifies into

$$
\bar{\delta}^3 + \bar{\delta}^2 = \frac{\psi L}{\phi^3} \bar{x}, \quad 0 < \bar{x} < 1
$$

(3.40)
Figure 3.5. Normalized film thickness $\bar{\delta}$ as a function of position $\bar{x}$ along the channel, for a downward gas flow and different values of normalized channel height $\bar{h}$. Shaded area shows region were $Re_\delta \leq 100$ and $\delta/L \leq 10^{-3}$ (for $h = 0.001$ m, $L = 1$ m and $\nu = 10^{-5}$ m$^2$s$^{-1}$).

The solution to this equation can be found in fig. 3.6. It can be used for an estimation of the order of magnitude in an engineering process.

Figure 3.6. Normalized film thickness $\bar{\delta}$ as a function of position $\bar{x}$ along the channel, in case of a downward gas flow and $\bar{\delta} \ll \bar{h}$.
3.4 Solution

3.4.2 Upward flow

In case the gas flow through the channel is in upward direction, two different flow regimes for the liquid film are possible depending on the relative magnitude of the different terms involved. The equation for the film thickness is given by

\[
\bar{\delta}^3 - \left( \frac{\bar{h}}{h - \delta} \right)^2 \left( \frac{4\bar{\delta}}{3(h - \delta)} + 1 \right) \bar{\delta}^2 = \frac{\psi L}{\phi^3} \bar{x}, \tag{3.41}
\]

and the profile for the velocity \( u \) in \( x \)-direction follows from eq. (3.25), after disregarding the centrifugal force and writing in terms of non-dimensional quantities, as

\[
\frac{u\nu}{g\phi^2} = \left[ -\frac{1}{2} + \frac{2}{3(h - \delta)} \left( \frac{\bar{h}}{h - \delta} \right)^2 \right] y^2 + \left[ \delta - \frac{4\bar{\delta}}{3(h - \delta)} \left( \frac{\bar{h}}{h - \delta} \right)^2 - \frac{2}{3} \left( \frac{\bar{h}}{h - \delta} \right)^2 \right] \bar{y} \tag{3.42}
\]

where also the coordinate normal to the channel wall is normalized according to \( \bar{y} = y/\phi \). Note that the velocity profile is a quadratic function in \( \bar{y} \) at every station along the channel.

To help interpret equations (3.41) and (3.42), first a simplified case is considered. If the liquid film thickness \( \delta \) is very small compared to the channel height \( h \), then the displacement effect is negligible and eq. (3.41) simplifies into

\[
\bar{\delta}^3 - \bar{\delta}^2 = \frac{\psi L}{\phi^3} \bar{x}. \tag{3.43}
\]

Furthermore, if it is assumed that \( \bar{h} \gg 1 \), the velocity profile is given by

\[
\frac{u\nu}{g\phi^2} = -\frac{1}{2} \bar{y}^2 + \left( \bar{\delta} - \frac{2}{3} \right) \bar{y} \tag{3.44}
\]

or, alternatively, by

\[
\frac{u\nu}{(2\delta - \frac{4}{3})g\phi^2} = -\bar{y}^2 + \bar{y} \quad ; \quad \bar{y} = \frac{\bar{y}}{(2\delta - \frac{4}{3})} \tag{3.45}
\]

The solution to eq. (3.43) is given graphically in fig. 3.7, for positive values of \( \bar{\delta} \). It covers two different flow regimes denoted by I and II. Regime I is characterized by a growing film thickness in negative \( x \)-direction, whereas the thickness of a liquid film in regime II increases with \( x \).

For a liquid film in regime I, the coordinate \( \bar{x} = (x - x_0)/L \) has a maximum value of zero, indicating that \( x_0 = L \) where the film thickness \( \delta \) is equal to zero. From this position onwards the film thickness grows in negative \( x \)-direction to a maximum at \( \bar{x} = -1 \) at the end of the channel. A maximum film thickness \( \bar{\delta} = 2/3 \) at the end of the channel \( (x = 0) \) is reached in case \( \psi L/\phi^3 = 4/27 \). For larger values of \( \psi L/\phi^3 \),
Figure 3.7. Normalized film thickness $\bar{\delta}$ as a function of position $\bar{x}$ along the channel, in case of an upward gas flow and $\bar{\delta} \ll \bar{h}$, showing two possible flow regimes I and II.

Figure 3.8. Two flow regimes in case of a gas flow in upward direction.
the film behavior switches to regime II where now the thickness $\bar{\delta}$ grows in positive $x$-direction, starting from $\bar{\delta} = 2/3$ at $x = 0$. Film behavior for the two regimes is shown schematically in fig. 3.8.

In regime I, all the liquid contained in the film will eventually leave the channel at the top. For a film in regime II, however, liquid will leave the channel both at the top and at the bottom. The fraction of liquid that leaves at the top is given by $x_0/L$, the value of which is easily determined as follows. At $x = 0$, or $\bar{x} = -x_0/L$, the film thickness has value $\bar{\delta} = 2/3$. Substitution into eq. (3.43) gives

$$\frac{x_0}{L} = \frac{4}{27} \frac{\phi^3}{\psi L} \quad (\delta \ll h, \text{Regime II}) \quad (3.46)$$

The velocity profiles for both flow regimes as given by eq. (3.45), are presented in fig. 3.9. Note that the velocity $u$ changes sign at $\bar{y} = 2\bar{\delta} - 4/3$ for a liquid film in flow regime II.

![Velocity profile](image)

**Figure 3.9.** Velocity profile as a function of dimensionless $y$ coordinate.

Also in the general case of an arbitrary channel height $h$ and finite film thickness, both flow regimes can occur. In fig. 3.10 the solution to eq. (3.41) is given graphically for different values of $\bar{h}$. From this figure it becomes apparent that both regimes can occur for high values of $\bar{h}$, where regime II is restricted to flows with a relatively large feed velocity and small shear force. When the feed velocity is reduced, or the shear force (i.e. gas velocity) is increased, the liquid film is forced into regime I. For low values of $\bar{h}$, the liquid film is always in regime I.
For a liquid film in regime II, an expression for the fraction of liquid leaving at the top, similar to eq. (3.46), is easily derived. Since the minimum value of $\psi L\bar{x}/\phi^3$ is obtained at $\bar{x} = -x_0/L$, the fraction of liquid $x_0/L$ leaving at the top can be written as

$$\frac{x_0}{L} = -\min \left[ \bar{\delta}^3 - \left( \frac{\bar{h}}{h - \bar{\delta}} \right)^2 \left( \frac{4\bar{\delta}}{3(h - \bar{\delta})} + 1 \right) \bar{\delta}^2 \right] \frac{\phi^3}{\psi L} \quad \text{(Regime II)} \quad (3.47)$$

In fig. 3.11 the position $\bar{y}$ in the liquid film is given where the velocity $u$ in $x$-direction changes sign. It shows that, in general, a local region of recirculating flow can occur in a liquid film in regime I. As mentioned above, this phenomenon disappears when $\bar{h} \gg 1$.

![Figure 3.10. Normalized film thickness $\bar{\delta}$ as a function of position $x$ along the channel, in case of an upward gas flow, for different values of $\bar{h}$.](image)

3.5 Discussion and conclusions

In this chapter, a model for fluid film behavior in a specific type of centrifugal separator is presented. It describes the characteristics of the liquid film in channels of a vertically oriented rotating phase separator (RPS) in which droplets are collected from a gas flow. The film is subjected to gravity, centrifugal force, and shear force at the interface between the film and the gas flow. It is assumed that the liquid film is a slender, axisymmetric, and laminar boundary layer type flow, for which inertia effects can be neglected. This is true for many practical installations. Furthermore it is shown that, although the centrifugal force is the driving force behind the separation
of droplets, it is of minor importance to the characterization of the liquid film. When neglecting the centrifugal force, the error is restricted to a local region close to the beginning of the film where the boundary layer approximation is no longer valid.

A key issue in the design of a RPS is the amount of fluid which leaves the separator at either the top or the bottom of the rotating element. This study shows that it depends on the geometry of the channels, the magnitudes of the different forces and the gas flowing either in upward or downward direction. In case of a gas flow in downward direction the solution is straightforward; since both gravity and shear force on the film are acting downwards, all liquid leaves the channel at the bottom of the element. For a gas flow in upward direction, two flow regimes are possible. For small amounts of liquid and/or high shear velocity, all liquid is expelled at the top of the channel. Below a certain threshold, however, the state of the liquid film may be such that only part of the liquid leaves at the top while the remainder flows in downward direction and leaves the channel at the bottom. An expression for the amount of fluid leaving at both sides is derived in this chapter.

In this chapter, the influx of droplets to the liquid film is assumed to be homogeneous, i.e. with a constant velocity \( v_0 \). In an actual RPS, this will not be true in general. The position along the channel where a droplet reaches the film surface depends on its size. Once the droplet size distribution is known, the influx distribution \( v_0(x) \) can be determined accurately [16]. Adaption of the current model for film build-up is then straightforward. The central eq. (3.38) for film thickness distribution is simply replaced by

\[
\bar{\delta}^3 = \left( \frac{\bar{h}}{\bar{h} - \bar{\delta}} \right)^2 \left( \frac{4\bar{\delta}}{3(\bar{h} - \bar{\delta})} + 1 \right) \bar{\delta}^2 = \frac{3\nu}{g\phi^3} U(x),
\]  

Figure 3.11. Position \( \bar{y} \) of velocity inflection as a function of normalized film thickness \( \bar{\delta} \), in case of an upward gas flow, for different values of \( \bar{h} \).
where eq. (3.27) for the flow rate \( U(x) \) is replaced by

\[
U(x) = \int_{x_0}^{x} v_0(x')dx'.
\] 

(3.49)

In case of a gas flow in upward direction and a liquid film in regime II, \( x_0 \) is determined from

\[
\frac{3\nu}{g\delta^3} \int_{x_0}^{x} v_0(x')dx' = -\min \left[ \delta^3 - \left( \frac{\bar{h}}{h - \delta} \right)^2 \left( \frac{4\delta}{3(h - \delta)} + 1 \right) \delta^2 \right]
\] 

(3.50)

In view of the pressure drop over the rotating element, a liquid film as thin as possible is preferred. In fig. 3.12 the liquid film thickness is compared for gas flowing in upward and downward direction, for different values of normalized channel height. From this figure it is concluded that a gas flow in downward direction leads to the smallest possible film thickness and is thus favorable, although the difference decreases for smaller channel height.

**Figure 3.12.** Comparison of film thickness distribution for gas flowing in upward and downward direction, for different values of normalized channel height \( \bar{h} \).
Chapter 4

RPS upscale

4.1 Introduction

After a successful proof of principle at small scale (50 kscf/d), the new technology has to be scaled up to industrial scale. The flow rates of a typical gas field application are on the order of a few hundred MMscf/d, i.e. approximately $10^4$ times larger see, table 4.1.

<table>
<thead>
<tr>
<th>Field application</th>
<th>test-rig SRTCA</th>
</tr>
</thead>
<tbody>
<tr>
<td>Product gas flow rate</td>
<td>100 to 1000 MMscf/d</td>
</tr>
<tr>
<td>Pressure</td>
<td>8 to 50 bar</td>
</tr>
<tr>
<td>Operating temperature</td>
<td>-90 to -50 °C</td>
</tr>
</tbody>
</table>

Table 4.1. Operating conditions of the future field application compared to the operating conditions of the SRTCA test-rig.

In section 4.2 the requirements for such a separator are given. These requirements are partly derived from test cases representing typical gas fields. Then a conceptual design is deduced, including the working mechanisms and corresponding fundamental theory. Special attention is given to the construction of the separator and the relation to the highly demanding operating conditions. The design is then evaluated for the test cases, to predict the separator’s performance. In the last section the influence of non-ideal gas compositions on the thermodynamic performance is investigated.

4.2 Separator requirements

The first requirement is that the design is capable of separating liquid droplets from a methane gas stream at thermodynamically favorable conditions. In practice this means that the separator should be able to withstand temperatures between -100 and 100 °C and pressures varying between 1-50 bar. The design should also be able to handle all frequently found components (e.g. H$_2$S, CO$_2$, H$_2$O, Hg, and various
Within natural gas. Related issues like lubrication, bearings and seals should be addressed.

The flow rate for the design presented in this chapter is 20 MMscf/d because this size is big enough to reliably test the technology, and at the same time small enough for affordable and convenient testing. When e.g. a test-loop has to be constructed the flow rate is one of the cost determining factors. To be able to cope with some fluctuations in composition and flow rates, the calculations have been performed using 25 MMScf/d. Conservative safety factors are applied, resulting in a robust design that can theoretically handle flows up to 100 MMscf/d for several gas compositions.

Another variable with a major influence on the separator design is the droplet size of the dispersed contaminants. The size of the droplets is estimated to vary between 1-10 µm as described in section 2.5. The preliminary measurements confirm this prediction by showing a size typically ranging from 2-15 µm. In order to obtain a high overall separation efficiency the required $d_{50\%}$ has been set to 0.5 µm.

### 4.2.1 Composition

The compositions of contaminated fields vary considerably. The design presented in this chapter is able to handle the six test cases shown in table 4.2, which are based on typical gas field compositions.

<table>
<thead>
<tr>
<th>Component</th>
<th>Case 1</th>
<th>Case 2</th>
<th>Case 3</th>
<th>Case 4</th>
<th>Case 5</th>
<th>Case 6</th>
</tr>
</thead>
<tbody>
<tr>
<td>CO2</td>
<td>20</td>
<td>28</td>
<td>54</td>
<td>71</td>
<td>9.47</td>
<td>5.53</td>
</tr>
<tr>
<td>H₂S</td>
<td>0.001</td>
<td>0.001</td>
<td>0.005</td>
<td>0.5</td>
<td>33.2</td>
<td>25.58</td>
</tr>
<tr>
<td>N₂</td>
<td>0.54</td>
<td>0.49</td>
<td>2.4</td>
<td>0.5</td>
<td>0.38</td>
<td>0.46</td>
</tr>
<tr>
<td>C₁</td>
<td>75.78</td>
<td>68.2</td>
<td>42</td>
<td>28</td>
<td>55.85</td>
<td>44.76</td>
</tr>
<tr>
<td>C₂</td>
<td>2.07</td>
<td>1.87</td>
<td>1.1</td>
<td>1</td>
<td>0.58</td>
<td>11.84</td>
</tr>
<tr>
<td>C₃</td>
<td>0.54</td>
<td>0.49</td>
<td>0.2</td>
<td>0</td>
<td>0.19</td>
<td>7</td>
</tr>
<tr>
<td>iC₄</td>
<td>0.17</td>
<td>0.15</td>
<td>0.1</td>
<td>0</td>
<td>0.07</td>
<td>1.08</td>
</tr>
<tr>
<td>nC₄</td>
<td>0.16</td>
<td>0.14</td>
<td>0.1</td>
<td>0</td>
<td>0.08</td>
<td>2.15</td>
</tr>
<tr>
<td>iC₅</td>
<td>0.09</td>
<td>0.08</td>
<td>0.05</td>
<td>0</td>
<td>0.05</td>
<td>0.54</td>
</tr>
<tr>
<td>nC₅</td>
<td>0.04</td>
<td>0.04</td>
<td>0.045</td>
<td>0</td>
<td>0.04</td>
<td>0.53</td>
</tr>
<tr>
<td>C₆</td>
<td>0.07</td>
<td>0.06</td>
<td>0</td>
<td>0</td>
<td>0.05</td>
<td>0.36</td>
</tr>
<tr>
<td>C₇⁺</td>
<td>0.539</td>
<td>0.479</td>
<td>0</td>
<td>0</td>
<td>0.04</td>
<td>0.17</td>
</tr>
</tbody>
</table>

**Table 4.2.** Six typical gas field compositions (mole%) used for evaluation of the separator design.

The first four cases are fields containing mainly CO₂ contamination. Cases 3 and 4 represent typical heavily contaminated “acid” gas fields containing almost no H₂S (Case 4 represents an Indonesian gas field [34]). Cases 5 and 6 comprise severe H₂S contamination (mainly associated gas fields in the Middle East or Russian area) and are therefore referred to as “sour” gas fields. For each composition different separation conditions are required. The properties at the required conditions are calculated (appendix D) using a Shell in house equation-of-state model. The software
uses an extended equation of state based on a cubic equation of state of the Soave-
Redlich-Kwong type with pure component parameters fitted to vapour pressures and
liquid densities along with a composition dependent mixing rule.

The used flash temperatures and pressures are based on the optimized operating
conditions using pure CH$_4$/CO$_2$ mixtures as described in [5]. The maximum enrich-
ments were determined at fixed recovery rates ($r_1 = 0.9$, $r_1 = 0.95$ and $r_1 = 0.98$)
using a maximum pressure of 250 bars prior to the insentropic expansion from a tur-
bine inlet temperature of -5 °C. The properties of the complete mixtures described in
table 4.2 are calculated at conditions that correspond to the optimal separation con-
ditions for identical fractions of CH$_4$ in the pure CH$_4$/CO$_2$ mixtures. The separation
conditions can be found together with other relevant properties in appendix E.

4.2.2 Droplet size

Due to the expansion cooling of a binary mixture, classical nucleation theory predicts
small micron sized droplets to be formed [7]. The process of nucleation, condensation
and coagulation is described in chapter 2. The process of nucleation is succeeded
by large numbers of micron sized droplets slowly coagulating into large droplets [35].
To keep the high pressure, low temperature induction section small, a short growth
time is required. For a 100 MMscf/d gas treatment plant a maximum growth time
of about 1 second is required: this already results in approximately 30 meters of 8.5
inch vacuum insulated and cooled piping. Within this time the droplets can grow to a
mean droplet size of about 1-5 µm. When a residence time of 0.1 second is regarded,
the droplets will reach a size of approximately 1-2 µm [5].

4.3 Basic design

4.3.1 configuration

The separator is designed in a vertically mounted position with downward flow for
optimal film behavior (chapter 3) in terms of pressure drop and liquid throughput
capacity. In fig. 4.1 a schematic configuration design of the separator is depicted. The
gas is fed to the separator via a tangential inlet that is sized to match the gas velocity
with the tangential velocity of the rotating RPS element. The gas first encounters the
pre-separator where big droplets and possible dirt are removed from the gas stream.
The liquid is drained from the pre-separator with a tangentially connected drain pipe
(Tangential liquid exit 1). The gas, still containing small droplets, is fed to the RPS
element where these small droplets are removed from the gas flow by centrifugal force
(RPS element). The liquid is collected in the post-separator where it is again drained
via a tangential outlet (Tangential liquid exit 2). The cleaned gas is removed via a
big tangentially connected pipe to regain some of the energy contained within the
rotational momentum (Tangential gas outlet).
4.3.2 Pre-separator

The pre-separator is a cyclone separator stabilized by the rotating RPS element. The flow enters tangentially as can be seen in fig. 4.2. In a cyclone the swirling flow decays
in the axial direction due to friction. The tangential velocity \( v_t \) of such a decaying flow can be described by: \[^{5, 36}\]

\[
v_t = v_{t0} \exp \left( -\frac{z}{2R_{pre}} \beta \right)
\]

(4.1)

where \( z [m] \) is the axial distance, \( R_{pre} \) the radius of the pre-separator, \( \beta \) an empirical factor that will be chosen at \( \beta = 0.05 \) (typical for the conditions in cyclones \[^{5, 36}\]\) and \( v_{t0} \) is the initial tangential velocity. The residence time \( \tau \) i.e. the time a particle needs to travel from \( R_{50\%} \) to the outer wall \( R_{pre} \) (see fig. 4.2), depends on the length of the pre-separator \( L_{pre} \) and the axial velocity \( v_{ax,pre} \). The cut radius \( R_{50\%} \) is defined as the radius where 50\% of the particles is present at \( r > R_{50\%} \). Because the inner diameter of the cyclone is equal to the diameter of the central shaft, the cut radius \( R_{50\%} \) can be calculated as

\[
R_{50\%} = \sqrt{\left( R_{pre}^2 + R_{shaft}^2 \right) / 2}
\]

This is valid when a uniform flow and a monodisperse particle size distribution are considered. The particle diameter that is collected with a 50\% probability within a decaying cyclone is given by:

\[
d_{p,50\%|pre} = \sqrt{\frac{9 \mu_g (R_{pre}^2 - R_{shaft}^2)}{2(\rho_p - \rho_g)v_{t0}^2 R_{pre}} \frac{v_{ax,pre} \beta}{1 - \exp \left( -\frac{L_{pre} \beta}{R_{pre}} \right)}}
\]

(4.2)

Because of the short pre-separator, the decay of the swirl is small. For half a meter of pre-separator the axial decay in this tube is less than 5\%. Moreover, the inlet velocity of the pre-separator is matched to the circumferential speed of the rotating RPS element, resulting in no decay at all. The tangential velocity \( v_t \) can therefore be substituted by \( v_{t0} \). Therefore equation (4.2) simplifies into:

\[
d_{p,50\%|pre} = \sqrt{\frac{9 \mu_g v_{ax,pre} (R_{pre}^2 - R_{shaft}^2)}{2(\rho_p - \rho_g)v_{t0}^2 L}}
\]

(4.3)

The pre-separator separates droplets of approximately 10-20 \( \mu m \). The size of the separated droplets mainly depends on the gas flow rate and inlet velocity. When the big droplets and other particles are removed in the pre-separator, the mist consisting of small unremoved droplets enters the RPS element.

### 4.3.3 RPS element

Within the RPS element, the micron sized droplets are forced to the walls of the small channels. There they form a film which flows through the element. The performance of the RPS element, can be estimated using a relation derived for concentric rings \[^{14}\]:

\[
d_{p50\%} = \sqrt{\frac{13.5 \mu_g d_c}{(\rho_p - \rho_g) \pi L(1 - \epsilon)(R_o^3 - R_i^3)}} \phi \frac{\rho}{\Omega^2}
\]

(4.4)

with \( \mu_g \) the dynamic viscosity of the gas, \( d_c \) the distance between rings, \( (\rho_p - \rho_g) \) difference between particle and gas density, \( L \) the element length and \( R_i \) and \( R_o \) the
element inner and outer radius. The operational parameters $\phi$ and $\Omega$ are the total volume flow through the element and the angular velocity of the RPS element. The correction factor $(1 - \epsilon)$ is used to correct for the axial flow cross-sectional area that is occupied by the channel walls, thus resulting in a higher gas velocity. Equation (4.4) holds for a laminar flow with either a uniform flow distribution over the element, or a linear distribution which means that the axial velocity is proportional to the distance from the rotation axis.

With relation 4.4 and the required separation droplet size of $d_{p50\%} = 0.5$ $\mu$m the dimensions of the RPS element in the case of uniform flow and concentric channels can be determined (see table 4.3). In general the smaller the channels, the better the separation performance. However, when channels become too small the pressure drop over the channels increases considerably. A channel diameter of 1mm proves to be a good compromise between the latter arguments [12]. At the same time this size is easy to manufacture, and provides a reasonable fouling/wear resistance. The length of 0.2 m is chosen because of manufacturing limitations of the current supplier. The inner and outer radius follow from flow area requirements in combination with practical considerations.

**Gap leak flow**

The rotating RPS element rotates within a static housing. Therefore there will always be a gap between the static housing and the rotating RPS element through which a leak flow can occur. In figure 4.3 the size of the gap is exaggerated for clarity. The flow with entrained small droplets bypasses the RPS element and will not undergo the desired treatment. Therefore, complete separation of the small droplets can not be guaranteed. The bigger the gap size $s_{gap}$, the larger the flow rate that will leak past the gap. As a consequence, separation performances will be harder to predict. When the gap size is reduced the friction between the element and static housing increases, thereby increasing required drive power and heat production within the separator. A practical limitation to the gap size is the fact that some clearance is needed to cope with vibrations, thermal gradients, production inaccuracy and shaft deflections.

To determine the leak flow rate, it is assumed that there is a uniform pressure drop across the RPS. This results in the pressure drop over the gap being equal to the cumulative pressure drop over both the RPS element and the post separator (see

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Channel diameter $d_c$ [m]</td>
<td>0.001</td>
</tr>
<tr>
<td>Length $L_e$ [m]</td>
<td>0.2</td>
</tr>
<tr>
<td>Outer Radius $R_o$ [m]</td>
<td>0.2</td>
</tr>
<tr>
<td>Inner Radius $R_i$ [m]</td>
<td>0.1</td>
</tr>
</tbody>
</table>

Table 4.3. Chosen dimensions for the RPS element (with: $d_{p50\%} = 0.5$ $\mu$m and $Q = 25$ MMscf/d)
The pressure drop in the post separator can be neglected compared to the pressure drop in the RPS element. This is caused by the much larger hydraulic diameter of the post separator than the channel diameter $D_{h,post} >> d_c$. Therefore eq. (4.5) becomes:

$$\Delta p_{gap} = \Delta p_{ch}$$

which leads to:

$$\left( f_{gap} \frac{L_{gap}}{D_{h,gap}} + \xi_{gap} \right) \frac{1}{2} \rho_g \left( \frac{Q_{gap}}{A_{gap}} \right)^2 = \left( f \frac{L_{fe}}{d_c} + \xi_{fe} \right) \frac{1}{2} \rho_g \left( \frac{Q_{fe}}{A_{fe}} \right)^2$$

(4.7)

where $Q_{fe}$ is the volume flow rate through the RPS element, $A_{fe}$ is the flow area of the RPS element, $Q_{gap}$ indicates the volume flow rate through the gap and $A_{gap}$ is the flow area of the gap. By rewriting the flow velocity in the RPS element and the gap as $v_{ax,fe} = Q_{fe}/A_{fe}$ and $v_{ax,gap} = Q_{gap}/A_{gap}$ respectively, assuming that the total flow rate $Q_c$ is equal to $Q_c = Q_{fe} + Q_{gap}$ and the resistance coefficient of the gap and the RPS element are equal, $\xi_{gap} = \xi_{fe} = \xi$ eq. (4.7) becomes

$$\left( f_{gap} \frac{L_{gap}}{D_{h,gap}} + \xi \right) \frac{1}{2} \rho_g \left( \frac{Q_{gap}}{A_{gap}} \right)^2 = \left( f \frac{L_{fe}}{d_c} + \xi \right) \frac{1}{2} \rho_g \left( \frac{Q_c - Q_{gap}}{A_{fe}} \right)^2$$

(4.8)

Substituting the hydraulic diameter of the annular gap by $D_{h,gap} = 2s_{gap}$ the volume flow through the gap $Q_{gap}$ can now be written as a function of the gap size ($s_{gap}$):

$$Q_{gap}(s_{gap}) = \left( \frac{\sqrt{\frac{\xi_1}{\xi_2(s_{gap})}}}{1 + \sqrt{\frac{\xi_1}{\xi_2(s_{gap})}}} \right) Q_c$$

(4.9)
with $\zeta_1 = A_{fe}^{-2} \left( f \frac{L_{fe}}{x_e} + \xi \right)$ and $\zeta_2(s_{gap}) = A_{gap}^{-2} \left( f_{gap} \frac{L_{gap}}{x_{gap}} + \xi \right)$. The friction factor of the gap $f_{gap}(N)$ is a function of rotational velocity ($N$). The friction coefficients, $f$ and $f_{gap}$, are both dependent on the Reynolds number, which in turn is dependent on the flow area’s $A_{gap}$ and $A_{fe}$. An iterative method is used to solve equation (4.9). A maximum gap size of a few millimeters is required to prevent the leak flow from exceeding 10%.

### 4.3.4 Droplet break-off from the RPS element

To separate the small liquid CO$_2$ droplets from the natural gas flow, the rotating RPS element will cause the droplets to coagulate into a liquid film on the channel wall. When these films leave the RPS element at the end of the channels, they break-up into bigger droplets that are easier to collect at a co-rotating collection wall. The last step of the separation process thus takes place at the end of the rotating channels of the RPS element.

Because the low surface tension $\sigma$ of liquid CO$_2$ ($\sigma \approx 0.01$ N m$^{-1}$), the formed droplets will be quite small. A worst case estimation for the break-off droplet size is used to calculate and design the post-separator. There are three main forces acting on the droplet at the moment of break-off:

- The surface tension, keeping the droplet together.
- The centrifugal force in outward radial direction trying to break-off the droplet.
- The fluid shear force acting on the surface of the droplet, also trying to break-off the droplet.

In fig. 4.4 a droplet is shown prior to break-off.

![Figure 4.4. Droplet prior to break-off. The shear force ($F_{sh}$) and centrifugal body force ($F_c$) are trying to break-off the droplet. The break-off forces are in equilibrium with the surface tension ($F_{su}$) which pulls the droplet to the channel wall.](image)
4.3 Basic design

Droplet break-off due to shear force

First the droplet size as a result from the shear force \( F_{sh} \) will be derived. Prior to the moment of break-off the surface tension \( \sigma \) acting on the perimeter of the surface is in equilibrium with the shear force acting on the surface of the droplet \( F_{sh} = F_{su} \), therefore

\[
\frac{1}{2} C_D \rho_g v_g^2 A = \sigma C
\]  

(4.10)

with,

\[
A = \frac{1}{2} \pi d^2 \\
C = \frac{1}{2} \pi d
\]  

(4.11)

\( A \) is half of the cross sectional area of the spherical surface, where the shear force acts on the droplet and \( C \) the perimeter of the base of the droplet where the surface tension connects the droplet to the liquid film and RPS element. The drag coefficient \( C_D \) can be estimated by the drag coefficient corresponding to a sphere, because the drag coefficient is determined by the boundary layer thickness. To obtain a correct value for the drag coefficient, the Reynolds number of the flow around the particle has to be determined [22].

\[
Re_p = \frac{\rho_g v_{p,r} d_p}{\mu_g}
\]  

(4.12)

Here \( v_{p,r} \) is the relative velocity of the particle with respect to the flow. The particle diameter \( d_p \) is equal to \( d \) defined in fig. 4.4. The drag coefficient \( C_D \) can be described as [35]:

\[
Re_p < 1 : \rightarrow \quad C_D = \frac{24}{Re} \\
1 < Re_p < 10^3 : \rightarrow \quad C_D = \frac{24}{Re}(1 + \frac{1}{6}Re^{2/3}) \\
10^3 < Re_p : \rightarrow \quad C_D \approx 0.44
\]  

(4.13)  

(4.14)  

(4.15)

The estimations for \( A \) and \( C \) are conservative because in the real case the droplet forms in the wake of the liquid film and RPS element, therefore, the surface where the shear force acts is overestimated. This is also the reason why the precise direction of the surface tension force is neglected. From relation (4.10) a conservative droplet diameter \( d \) can be derived

\[
d = \frac{8\sigma}{C_D \rho_g v_g^2} = \frac{8 \cdot 0.01}{0.44 \cdot 30 \cdot 100} \approx 60 \mu m
\]  

(4.16)

with \( C_D = 0.44, \rho_g = 30 \text{ [kg/m}^3\text{]} \) and \( v_g = 10 \) [m/s] which are again conservative values. This prediction corresponds to the Weber number defined as:

\[
We = \frac{\rho_g v_g^2 d}{\sigma}
\]  

(4.17)

being equal to its critical value of \( 8/C_D \) [37].
Droplet break-off due to centrifugal force

It can also be the case that the droplet is torn off by the centrifugal force acting on the droplet. In the case that the centrifugal force is in equilibrium with the surface tension $F_c = F_{su}$ the force balance for the smallest droplet equals:

$$\rho_l \Omega^2 RV = \sigma C,$$  \hspace{1cm} (4.18)

where $V$ is the droplet volume, and $R$ is the outer radius of the RPS element. Again the direction of the forces are not taken into account. When the droplet is assumed to be more or less spherical the following volume can be substituted:

$$V = \frac{1}{6} \pi d^3 \hspace{1cm} (4.19)$$

At the moment of equilibrium this results in:

$$\frac{\Omega^2 R \rho_l d^2}{\sigma} = 3$$ \hspace{1cm} (4.20)

The droplet diameter $d$ at the moment of break off can then be rewritten as:

$$d = \sqrt{\frac{3 \sigma}{\rho_l \Omega^2 R}} = \sqrt{\frac{3 \cdot 0.01}{1000 \cdot 7000}} \approx 60 \mu m \hspace{1cm} (4.21)$$

with the centrifugal acceleration estimated at $\Omega^2 R = 7000$. This prediction corresponds to the Bond number defined as:

$$Bo = \frac{\Omega^2 R \rho_l d^2}{\sigma}$$ \hspace{1cm} (4.22)

being equal to its critical value of 3 [37]. Both forces thus yield the same droplet size of approximately 60 $\mu$m.

Droplet break-off due to both forces

For determination of the length of the post-separator it is assumed that both the shear and the centrifugal force coincide and work in the same direction, thus both forces counteract the surface tension $F_{sh} + F_c = F_{su}$. The force equilibrium can then be described by:

$$\frac{1}{2} C_D \rho_g v_g^2 A + \rho_l \Omega^2 RV = \sigma C.$$ \hspace{1cm} (4.23)

This gives a minimum break-off droplet size of $d \approx 40 \mu m$ to be considered as a worst case estimate. It is very small compared to the film thickness at the element exit of about 300 $\mu m$ in a highly loaded RPS (chapter 3). Therefore the droplets will probably be a lot larger than the estimated 40 $\mu m$. 
Droplet break up by turbulent stress

The droplets that are broken up behind the RPS channels could be destroyed by the turbulent stresses \( \sigma_t \) during the collection process. The turbulent stresses affecting the droplet can be determined by [38–40]:

\[
\tau_t = \rho \left( u'(x, t) - u'(x + d, t) \right)^2 \approx \rho C (\epsilon d)^{\frac{2}{3}}
\] (4.24)

where \( u' = u - \bar{u} \) is the fluctuation of the velocity over the diameter \( d \) of the droplet. The constant \( C \) is approximately equal to 2 for homogeneous turbulence and the turbulent dissipation \( \epsilon \) is approximately equal to the turbulence production \( P_k \). If small redistribution terms are neglected, the turbulence production can be calculated using [38]:

\[
\epsilon = P_k = -u'_i u'_j \frac{\partial u_i}{\partial x_j} = \frac{2k^{-1} u^3}{d_c}
\] (4.25)

with the Von Kármán constant \( k \approx 0.4 \), the shear velocity \( u_* = \sqrt{\tau_0/\rho} \), with \( \tau_0 \) the mean shear stress and \( d_c \) the RPS channel diameter. The turbulent stresses act as a force onto the droplet surface. When this force exceeds the force of the surface tension (acting on the perimeter of the droplet) the droplet will break-up. With this force balance a maximum stable droplet size within the turbulent flow can be estimated:

\[
\tau_t d^2 = \sigma d
\] (4.26)

Rewriting with the help of eq. (4.24) gives a maximum droplet size of:

\[
d \leq \left( \frac{\sigma}{C \rho \epsilon^{\frac{2}{3}}} \right)^{\frac{3}{5}}
\] (4.27)

for typical circumstances where the shear velocity is about 5% of the bulk velocity this results in a maximum droplet size in the order of 1 mm.

4.3.5 Collection process in the post-separator

The last part of the separation process is called the liquid collection. The liquid collection is designed in the following way:

- From the size of the droplets that break off from the RPS element, the length of the post-separator and co-rotating collection wall is calculated.
- The droplets are then collected on the co-rotating wall of the post-separator, where they will form a new film.
- This film will again break off into droplets/streams that flow into the liquid drainage volutes, where the liquid CO\(_2\) is drained from the RPS.

As shown in the previous section the film of liquid CO\(_2\) will break up in small droplets when leaving the RPS channels. Within the post-separator collector the gas still rotates as a solid body with the speed of the RPS element. The centrifugal force causes the droplets to migrate outwards (see fig. 4.5). Depending on the size of the droplets after break-off the necessary length \( L_{\text{post}} \) of the post-separator can be calculated. To determine the path of the droplet a force balance can be formulated.
Figure 4.5. Droplet migration due to centrifugal force ($F_c$). The centrifugal force is in equilibrium with the drag force ($F_d$) and buoyancy force ($F_b$).

On a CO$_2$ droplet the centrifugal force and two counter acting forces, the buoyancy force $F_b$ and the drag force $F_d$, are working. Assuming that the acceleration time for the moving particle is very short the force balance equals:

$$F_c = F_d + F_b \quad (4.28)$$

The drag force is dependent on a drag force coefficient $C_D$ and the frontal area of a particle. The velocity can be treated to be only a function of radial position $r$. The radial velocity of the particle $v_{p,r}$ can be described as

$$v_{p,r}(r) = \sqrt{\frac{2(\rho_p - \rho_g)m_p\Omega^2r}{C_D\rho_gA_p\rho_p}} \quad (4.29)$$

When the particle is assumed to be spherical, the mass $m_p$ and frontal area $A_p$ are $m_p = \frac{1}{6} \pi d_p^3 \rho_p$ and $A_p = \frac{1}{4} \pi d_p^2$ respectively, the radial settling velocity $v_{p,r}$ becomes

$$\frac{dr}{dt} = v_{p,r}(r) = \sqrt{\frac{4(\rho_p - \rho_g)d_p\Omega^2r}{3C_D\rho_g}} \quad (4.30)$$

With the radial velocity of the CO$_2$ droplet known, the required residence time $\tau_{post}$ within the post-separator collector can be determined. This is the time it takes for a particle to travel from the inner radius ($R_{shaft}$) to the outer radius ($R_{co}$) of the post-separator. Because this is the largest distance to travel this will determine the length of the post-separator.

Integrating eq. (4.30) and writing $R_{shaft} = \delta R_{co}$ the residence time of the particle in the post-separator can be calculated as.

$$\tau_{post} = \sqrt{\frac{3C_D\rho_gR_{co}(1-\sqrt{\delta})^2}{(\rho_p - \rho_g)d_p\Omega^2}} \quad (4.31)$$
To obtain the correct value for $C_D$ equations (4.13)-(4.15) can be used. The length of the post-separator can now be described with eq. (4.32).

$$L_{post} = \tau_{post} \cdot v_{ax,post} \quad (4.32)$$

The separator has been designed with a post-separator length of 0.1m based on the calculations described above. Due to the conservative break-off droplet model, no additional safety factor has been used for the length of the post-separator. Because the required length is sensitive to the droplet break-off size, experimental validation (with liquid CO$_2$ at realistic pressures), of the calculated droplet break-off size is required.

### 4.3.6 Liquid drain volutes

The liquid has now formed a thin film on the inside of the rotating post separator wall. This film will flow downward and again break up into droplets that move outwards and have to be collected. The collection is performed with the help of a volute with a tangential outlet. The volute needs to be big and smooth enough to ensure proper removal of the turbulent liquid flow. The flow resistance is kept as small as possible to smoothly drain the liquid. To estimate the required size of the volute, the dissipation of momentum within the volute is regarded, considering a u-shaped volute (see fig. 4.1). In fig. 4.6 the top view of the u-shaped volute is depicted.

![Figure 4.6](image.png)

**Figure 4.6.** Top view of the u-shaped volute. The liquid is fed into the volute via the outer wall, with a constant velocity ($v_{t,f}$).

The fluid is fed by the film flowing out of the post-separator collector (see fig. 4.5) into the u-shaped volute at $R_f$ with a tangential velocity of $v_{t,f}$. The contained angular momentum $I_f$ is given by:

$$I_f = \dot{m}v_{t,f}R_f \quad (4.33)$$
with \( \dot{m} \) the mass flow rate, \( v_{t,in} \) the inlet velocity and \( R_f \) the radius at which the liquid is fed into the volute. To first order this can be estimated by the momentum contained within the liquid at the moment it leaves the RPS element. The difference in rotational momentum between inlet and outlet is equal to the dissipation within the volute:

\[
\dot{m}(v_{t,f}R_f - v_{t,out}R_m) = \int_{\Delta A} \tau r dA
\]

\[
\tau = \frac{1}{2} f \rho v^2
\]

where \( v \) and \( dA \) are the local liquid velocity and surface area at radius \( r \), and \( \tau \) the shear stress. Assuming that both the bottom and the outside wall of the u-shaped volute are wetted this can be estimated using:

\[
\dot{m}(v_{t,f}R_f - v_{t,out}R_m) \approx \frac{1}{2} f \rho \overline{v^2} \left(2\pi h R_{out}^2 + \pi (R_{out}^2 - R_{in}^2) R_m\right)
\]

When a certain geometry size is chosen the momentum dissipation can be calculated, which leads to an outflow velocity with help of eq. (4.36). The velocity should not change too much between inlet and outlet in order to get an accurate prediction with eq. (4.36). With the mass flow rate present in the volute the outlet velocity should be able to fit the chosen volute geometry with a realistic flow profile. A more detailed model for the post-separator volute film and an experimental evaluation of this detailed model can be found in the next chapter.

### 4.4 Construction

A decisive part of the design of the separator is the mechanical construction. Within the oil and gas business reliability is often more important than cost reduction. The design of the separator has to be robust and able to withstand the harsh operating environment. The low temperatures and corrosive gases lead to special demands for materials and construction. The definite choice of gaskets, flanges and materials has to be done while incorporating the required design codes (e.g. DEP, ASME, Stoomwezen, BS5500, NACE, ANSI, NEN, ISO, ATEX, IEC) which are required at the operating location. Of course care should be given to the manner of operation, because a demonstration unit will differ from an industrial production unit.

#### 4.4.1 Construction material

The natural gas processing will take place at temperatures as low as -90 °C (determined by the thermodynamical optimal point for \( \text{H}_2\text{S} \)) which limits the use of several metal alloys. Within the gas some highly corrosive substances e.g. chlorides and sulphides can be present that can lead to several forms of corrosion e.g.: pitting corrosion, sulphide stress cracking and crevice corrosion [41]. Although the unit is operated in the “severe sour regime”, due to the low temperatures the corrosion risk is less severe [41].
Stainless steels are known for their corrosive resistant properties. There are seven basic families of stainless steels: ferritic, austenitic, precipitation hardenable, super-ferritic, martensitic, duplex (ferritic/austenitic) and super-austenitic. For the operating regime described above, austenitic, super-austenitic and duplex steels are the most useful. Duplex steels often get brittle at temperatures below -70 °C. From the austenitic family, AISI 316(L,LT) is useful at low temperatures and also fairly corrosion resistant (see appendix G). An additional advantage of this grade of steel is the good availability due to the fact that it is commonly used. Super-austenitic steel has more favorable corrosion resistance, but is more difficult to machine, purchase and is relatively expensive.

The separator at SRTCA (described in chapter 2) was constructed out of duplex. Because testing is done with pure CH$_4$/CO$_2$ mixtures the operating temperature never reaches values below -60 °C. Duplex steel was chosen because of the low thermal expansion coefficient, which was useful because of the small tolerances[5]. These small tolerances were needed because of the required small leak flow rate due to the single row of channels. For the upscale design AISI 316(L,LT) is anticipated because of the good availability of several pre-manufactured parts, the relatively short operating times of the test-separator and the regular checks of the experimental equipment. These regular checks will reveal corrosion damage before the structural integrity of the separator is compromised.

4.4.2 Pressure housing

The RPS is placed inside a standard 24 inch certified pressure pipe. This pipe can then be adapted by welding on all the supply and outlet pipes. The top and bottom side of the RPS use standard blind flanges, which are adapted to fit the bearing housing (see figure 4.9). Because of the adaptations extensive pressure testing and X-ray testing for the welds has to be done in order to complete the required paperwork. The calculations have been performed using a commercial software package called Ansys. The clean gas outlet is one of the weakest points of the construction due to the large cutout. The deflection of the blind flanges and its influence on the bearings has been investigated using finite element package (Ansys). The deflection is limited to 0.1 mm to prevent problems with the tolerances.

The RPS can operate between 1 and 50 bar. Therefore, the design pressure $p_{des}$ is chosen at 10% above the high value, which is 5 percent above the maximum operation pressure ($p_{des} = 58$ bar). When the design pressure is 58 bars the pressure testing has to be performed at 1.43 to 1.5 times design pressure $1.5p_{des} = 87$ bar. For safety reasons the design calculations will be performed with a 100 bar pressure loading to prevent damage during pressure testing. The required pressure class will most likely be the 600 or 900 pounds class but will be determined by the test-facility design pressure.

4.4.3 Drive system

The RPS element is placed within the static housing. It should be able to rotate around the centreline while clearing the gap. The element’s rotation is controlled by
an externally powered drive system.

**Figure 4.7.** Preliminary design of the RPS shaft assembly (rotated 90°).

### Element

The key component of the RPS is the RPS element. The dimensions have been determined using the performance calculations described in section 4.3.3 in combination with a required $d_{p50\%}$ of 0.5 µm. This is a conservative value since the size of the droplets is expected to vary between 1 and 15 µm. The outer radius is chosen at 200 mm (see table 4.3), the inner radius at 100 mm and the length of the RPS element is 200mm. The inner radius is sized to allow for a stiff shaft and at the same time preventing small particles from leaking through the RPS element at small radii.

The element can be constructed by winding up two sheets of metal, one plain sheet and one corrugated sheet (see I and IV in fig. 1.3). These sheets have to be (laser) welded together to construct a rigid RPS element. Another option is to construct the element from a bundle of small tubes or needles (see II in fig. 1.3).

The outer sleeve of the element is extended (Collector in fig. 4.7) to form the co-rotating wall of the post-separator where the big droplets (that broke off from the RPS element) are collected (see fig. 4.7). Optionally plates can be placed within the post-separator collector to ensure the solid body rotation.

### Shaft

The RPS element is mounted on a stiff shaft. The shaft consists of 2 parts, the inner shaft and the outer shaft (see fig. 4.7). The outer shaft is used to fill up the gap between the inner shaft and the RPS element. The inner shaft needs to provide the required stiffness and strength.

The whole shaft assembly weighs around 200 kg. The stiffness and strength have been investigated using a finite element package called MSC.Marc Mentat. A small radial deflection of the shaft is important because the shaft has to clear the small gap (see fig. 4.3).

The shaft is supported by two bearings at the top and bottom flange (see fig. 4.9). One bearing will accommodate an axial and radial support whilst the other only accommodates a radial support to allow for thermal expansion. The shaft is placed vertically therefore the radial force will be mainly due to unbalance. The
vertical force (ca. 8.5 kN) is due to the weight of the shaft assembly in combination with the pressure drop over the RPS element.

The radial force $F_r$ is limited to 3.5 kN which limits the unbalance to approximately 100 gram ($F_r = m\Omega^2 r$ at maximum rotational velocity and maximum radius). The shaft deflection will then be less than 0.036 mm (calculated with MSC.Marc Mentat). To check whether eigen vibrations could cause trouble a simple dynamic analysis was performed on the inner shaft. The natural eigenfrequency of a static beam equals [42].

$$\omega = \frac{n^2 \pi^2}{L^2} \sqrt{\frac{EI}{m}} \quad (4.37)$$

where $n$ indicates the $n^{th}$ mode of the natural frequency, $L$ and $m$ the length and mass of the shaft, $E$ the E-modulus and $I$ the moment of stiffness. The first eigenmode has a frequency corresponding to a rotational velocity of 15000 rpm, which is high enough to be disregarded. When the outer shaft would be incorporated in the analysis the influence of the stiffness would increase faster than the influence of the extra weight leading to an even higher eigenfrequency. Torsional vibrations have not been included in the analysis because they are not expected to be a problem.

**Bearings**

As is mentioned before, the shaft is mounted with the help of two bearings, one placed in the upper and one in the lower blind flange. The maximum radial force is 3.5 kN which is equally distributed over both bearings. The axial loading consists of a static and a dynamic part. The static radial loading is mainly due to the weight of the shaft assembly which leads to a loading of 2.1 kN eq.(F.2). This force can also be used to apply pre-tension to one of both bearing assemblies. The dynamic axial force, mainly due to the pressure drop over the RPS element, is as large as 6.3 kN eq. (F.3). The pure radial bearing is positioned in the lower flange and the radial/axial bearing is positioned in the upper bearing assembly. This is done in order to be able to hoist the complete shaft assembly out of the housing in one piece (see fig. 4.9).

Different types of bearings were considered; active magnetic bearings (AMB’s), gas bearings, ceramic plain bearings, hybrid bearings and normal roller bearings. There are several advantages and disadvantages for each type of bearing (see table F.1).

Roller bearings have difficulty reaching the required speeds up to 4000 rpm for the large diameters that are needed in this application. The low operation temperature below -60 °C makes lubrication problematic.

Gas bearings need a very small clearance between the stator and rotor (0.01 mm). These small tolerances are difficult to accomplish due to the thermal expansion. The stiffness of the bearing could be insufficient and the required auxiliary gas compressor is not ideal.

Three options then remain: hybrid roller bearings, ceramic plain bearings and AMB’s (active magnetic bearings). AMB’s seem to be most suited for this application. AMB’s provide low friction contactless levitation of the shaft with the help of magnetic forces. The attainable rotational speeds are high enough. When the bearing
is canned (integrated) into a stainless steel casing corrosion can be prevented. The high capital expenditure is the biggest disadvantage of this type of bearing especially for a prototype with a relatively small number of operation hours.

Hybrid roller bearings and ceramic plain bearings can be lubricated with the help of the process medium, in this case liquid CO$_2$. Both can handle the required loadings and rotational velocities. Hybrid bearings are cheaper but less reliable.

Ceramic plain bearings can be constructed from silicon carbide, which is a very hard material with almost no thermal expansion. The bearings can run without lubrication for a short period of time e.g. during start-up. This will reduce their life time expectancy. When liquid CO$_2$ is supplied for lubrication the wear and heat production will diminish.

For this application hybrid roller bearings, ceramic plain bearings and AMB’s can all be used. The final selection will depend on the specific operating conditions and operation times.

**Electrical drive**

The RPS can be driven in two different ways, naturally driven and externally driven. Naturally driven means that the incoming momentum of the process medium provides enough torque to drive the RPS element up to the required speed. This is effected with the help of a swirl generator or tangential inlet configuration. A disadvantage of this drive is the lack of controllability over the rotational velocity of the RPS element. Especially when the RPS has to cope with different gas compositions the natural drive will not be efficiently used. For this demonstration prototype an external drive is therefore incorporated. This has the advantage that it can be used to precisely control the rotational velocity of the shaft. It can be used to speed up the assembly but also to slow it down when the velocity resulting from the incoming momentum exceeds the required value (dynamic breaking).

The most simple external drive is a standard electric motor. The motor can be integrated into the housing, this is called a canned motor design. The canned motor design could have some advantages. The main advantage is that the rotating shaft no longer has to be dynamically sealed for it is no longer fed-through the flanges. A disadvantage of the canned design is that the heat produced by the canned motor will transfer to the gas flow. The produced heat needs to be considered to conserve the favorable thermodynamic state. A standard electrical motor can also be applied at the cost of a more complex seal or coupling which will be described in the next section.

The size of the motor to drive the assembly up to the required speeds will be around 10 kW (40 Nm torque). When looking at the dynamic braking the maximum required power will be around 20 kW (148 Nm) to be able to dissipate enough energy. For the prototype an electric motor of around 30 kW (200 Nm) is advised for optimal controllability.
4.4 Construction

Coupling

As stated above the electric motor has to drive the shaft via a coupling. This can be done by a dynamic seal, i.e. a seal that is connected to the stationary housing and to the rotating shaft at the same time. This seal will always be subject to wear and will be a point of concern in the case of very toxic gases.

Another option is a magnetic coupling. Then a stationary seal is applied and the shaft is driven by a slave magnet that follows the magnetic field produced by a master magnet that is connected to the motor. In between the two magnets, a can provides the physical boundary for the gas.

The magnetic couplings can function at low temperatures and the required pressures. The main disadvantage are the produced eddy currents in the coupling can. These generate a considerable amount of heat, especially when a metallic coupling can is used. The eddy currents can be reduced by using a non metallic coupling can, e.g. made from ceramics or plastics (PEEK). Both materials can cope with the required pressures and temperatures.

The size of the coupling is mainly determined by the static break-away torque. From calculations it follows that the required torque of 200 Nm can be transferred at 3000 rpm with a heat production of less then 1 kW which is acceptable.

Seals

The housing has to be gastight to prevent the gas from escaping from the separator. When a considerable amount of the very toxic H$_2$S is present in the gas (see cases 5 and 6 table 4.2) the sealing becomes an important issue. H$_2$S can already be lethal at a concentration of a few ppm.

Within the current design seals are needed in three places: the upper blind flange, the bottom blind flange and for the magnetic coupling (see fig. 4.9). The upper and bottom blind seals are pressed between the pipe flanges and blind flanges. The coupling seal is pressed between the coupling can and the blind flange. All these seals are static seals which significantly reduce the risk of leakage. The preferred type of gaskets for flange sizes up to 24 inch are spiral wound gaskets. For larger sizes cam-profile gaskets are often used.

4.4.4 Design

Considering all the requirements and broad operating range, a concept design has been created. With this concept design in combination with the required design codes a detailed design can be made, from which detailed production drawings can be constructed. In fig. 4.8 a rendered image of the separator can be found. The housing is constructed from a standardized 24 inch pipe in combination with two weld-on-flanges. This pipe is bolted to two blind flanges with a gasket in between to ensure gas-tightness.

In fig. 4.9 a cross-sectional view of the separator design is shown. On the lower blind flange the lower bearing compartment is constructed. Depending on the type of bearing that will be used the detailed construction of this compartment will vary.
Another bearing compartment is needed at the upper blind flange to support the upper bearing. The shaft extends through a hole in the upper blind flange into the magnetic coupling can. Within this coupling can a slave magnet is placed on the shaft, which is driven by a master magnet rotating on the outside of the coupling can. The coupling can is present to provide the leak-tightness of the separator. The coupling can is mounted with a static seal onto the upper flange. The static seal, gaskets and electric driving motor are not depicted in fig. 4.9. The mixture inlet can not be seen in the cross-section, it is located at the upper left corner of the pre-separator. The clean gas outlet can be seen at the lower left corner. For the liquid removal there are two liquid outlets, the pre- and the post separator outlet. Note that, in the drawings in this chapter, the liquid outlets have been constructed perpendicular instead of tangentially, this makes it easier to present a clear cross sectional view.

When the gas enters the pre-separator through the gas inlet, the size of the inlet is such that the gas enters the pre-separator with a velocity just below the rotational velocity of the RPS element (ca. 10 m/s). The pre-separator can optionally be constructed with guide vanes as shown in fig. 4.9. The coarse droplets and debris move outwards and enter the pre-separator volute. The mist of finer droplets enters the small channels of the RPS element and post separator. The construction of the post-separator including vanes can be seen in the rendered image in fig. 4.10. From the co-rotating wall of the post-separator the liquid is fed into the post-separator volute where it is drained.
Figure 4.9. Preliminary design of the rotating phase separator
4.5 Predicted performance

When the design is completed the theoretical performance can be calculated. Due to the broad range of gas compositions within the 6 cases, it is difficult to optimize the design. Since no gas location has such broad variations as the six cases a more optimal design can be made for a real field application. Each case can be operated at different conditions (mainly pressure and temperature) again resulting in variation in flow rate, density and other parameters. When a unit is built for a fixed concentration and mass flow rate a further optimization can be performed.

In the table 4.4, the required rotational velocity to achieve a $d_{p50\%}$ of 0.5 $\mu$m, for each case are depicted. The large variation in gas flow rate is due to the different pressure and temperature of separation. The higher flow rates mainly correspond to low separation pressures (due to the fixed size of 20 MMscf/d at atmospheric conditions). The separation calculations have been performed at the theoretical optimized conditions for pure CH$_4$/CO$_2$ mixtures with a recovery of methane of $r_1 = 0.9$, 0.95 and 0.98. In this table also the $d_{p50\%}$ of the pre-separator is calculated with the help

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Case 1</th>
<th>Case 2</th>
<th>Case 3</th>
<th>Case 4</th>
<th>Case 5</th>
<th>Case 6</th>
</tr>
</thead>
<tbody>
<tr>
<td>Min airflow $[m^3s^{-1}]$</td>
<td>0.06</td>
<td>0.07</td>
<td>0.15</td>
<td>0.18</td>
<td>0.10</td>
<td>0.09</td>
</tr>
<tr>
<td>Max airflow $[m^3s^{-1}]$</td>
<td>0.14</td>
<td>0.19</td>
<td>0.37</td>
<td>0.50</td>
<td>0.21</td>
<td>0.20</td>
</tr>
<tr>
<td>Minimum [rpm]</td>
<td>1227</td>
<td>1176</td>
<td>1351</td>
<td>1468</td>
<td>1169</td>
<td>1266</td>
</tr>
<tr>
<td>Maximum [rpm]</td>
<td>1563</td>
<td>1680</td>
<td>2153</td>
<td>2446</td>
<td>1638</td>
<td>1836</td>
</tr>
<tr>
<td>$d_{p50%}$ pre-sep [$\mu$m]</td>
<td>12-22</td>
<td>10-18</td>
<td>6-10</td>
<td>6-9</td>
<td>9-13</td>
<td>10-16</td>
</tr>
</tbody>
</table>

Table 4.4. Case specific design parameters for a $d_{p50\%} = 0.5$ $\mu$m (feed flow rate of 25 MMscf/d), the RPS element dimension as stated in table 4.3 and a tangential gas inlet diameter of $D_i = 0.15$ m.
of eq. (4.3). For the tangential velocity the inlet velocity corresponding to an inlet diameter of 0.15 m is used. In case of large variation in gas flow rates the inlet velocity can best be chosen close to (but less than) the rotational velocity of the RPS element.

Although most of the RPS applications up till now were at atmospheric conditions and therefore laminar flow within the channels, the axial Reynolds number in this application varies between $Re_{ax} = 5000$-$11000$. This means that the flow through the channels is turbulent. According to DNS calculations performed by [8; 9] this will only slightly affect the separation efficiency. The efficiency mainly diminishes for large particles. The efficiency for small particles that are only partially collected will not be affected because the random turbulent fluctuations will sometimes enhance and sometimes worsen the separation (see fig. 2.5). The efficiency of large particles however, that in the laminar case would have been collected with a 100% probability, can only be affected in a negative way. The separation efficiency diminishes mainly for large particles and the size of particles that are separated with 100% probability is increased by the turbulence.

In order to get an efficient process approximately 98% of the liquid has to be collected. In case of turbulent flow, collection of 98% of the particles requires a mean particle diameter of approximately 3 times $d_{p50\%}$ (for more details see chapter 5).

Due to the high centrifugal force, particles that get stuck in the channels could block the RPS element. With increasing size of a fouling particle the volume to surface ratio increases. The centrifugal force $F_c$ is in equilibrium with the buoyancy force $F_b$ and the resulting normal force $F_N$. The normal force gives rise to a friction force $F_w$ which keeps the particle in the channel. The drag force $F_d$ exerted by the liquid CO$_2$ is trying to force the particles out of the channel. Because the centrifugal force acts on the mass of the particle and the drag force on the surface of the particle, the dominating force changes with particle size. This means that the bigger particles are more likely to block the channels because the friction force becomes dominant. From a short theoretical analysis it follows that the size of particles that is likely to block the channels will already be removed in the pre-separator. An overview of several fouling mechanisms is shown in appendix I.

![Figure 4.11. Fouling particle under influence of several forces.](image_url)
4.6 Thermodynamic performance of complete mixtures up to \( C_7 \)

The six fields (table 4.2) are simulated now including all the higher hydro carbons up to \( C_7 \) using an in house equation of state (CPA). For each of the six fields recovery rates and enrichments have been calculated. Enrichment (2.2.1) is defined as the increase of \( CH_4 \) concentration and the recovery rate \( r_1 \) is defined as the total amount of methane recovered divided by the total amount of methane supplied within the feed stream. So \( 1 - r_1 \) is the relative amount of methane lost in the waste. Enrichments and recovery rates have been calculated for pressures \( P \) and temperatures \( T \) which were previously identified as optimal by Ralph van Wissen, i.e. the values at which for a given recovery rate the enrichment reaches a maximum. This optimal is determined using the same constraints as explained in chapter 2, i.e. without any consideration concerning the higher hydrocarbons. It is difficult to optimize for these complete mixtures because the plant design as well as the economical values for each of the components should then be incorporated into the analysis. Results obtained for an original recovery rate of \( r_1 = 0.95 \) are summarized in the table 4.5 supplemented with additional values corresponding to slightly changed pressure and temperature conditions.

Clearly, the approach in which optimal values of \( P \) and \( T \) are based on analysis of \( CH_4/CO_2 \) only, will not lead to the best separator performance for the case where the \( CH_4/CO_2 \) mixture includes the high HC components. So, when higher components are present, the optimization procedure has to be repeated, i.e. for each chosen recovery rate the values of \( P \) and \( T \) have to be optimized for maximum performance. To obtain a first insight in what will happen for different values of \( P \) and \( T \), some additional calculations have been performed using the model with higher HC’s included at slightly different pressures and temperatures. The results have been presented in the table as well. The first row represents the pure \( CH_4/CO_2 \) mixtures. The second row represent the complete mixture at the same temperature and pressure. The third and fourth line are the results obtained at slightly different pressure and temperature.

Considering the results presented in the table it can be concluded that separator performance using higher HC components is comparable to previous results of Ralph van Wissen in which higher HC’s have been disregarded. Re-optimization of pressures and temperatures will probably lead to some improvement in most cases. Improvement without any change in operating conditions can be seen in the simulation of Case 5. Here at equal recovery rate (0.95) the enriched gas contains 85 mole% \( CH_4 \) instead of the previously predicted 75 mole% \( CH_4 \). Note that the purity and recovery of the methane is investigated and not the amount of condensed \( CO_2 \).

The recovery \( r_1 \) only concerns the \( CH_4 \) content. It is inconclusive for the recovery of the higher hydrocarbons. Most of the heavier HC are completely lost with the liquid \( CO_2 \) and even for ethane (\( C_2 \)) a substantial amount is contained within the waste stream. Therefore a dedicated waste treatment should be considered when the higher HC’s represent substantial value.

The predicted performance with pure \( CH_4/CO_2 \) mixtures will be achievable when
<table>
<thead>
<tr>
<th>Field</th>
<th>$P$ [bar]</th>
<th>$T$ [°C]</th>
<th>$z_1$</th>
<th>$y_1$</th>
<th>Enrichment</th>
<th>$r_1$</th>
<th>Feed</th>
</tr>
</thead>
<tbody>
<tr>
<td>Case 1</td>
<td>38</td>
<td>-62</td>
<td>76</td>
<td>85</td>
<td>9</td>
<td>0.95</td>
<td>CH4/CO2 only</td>
</tr>
<tr>
<td></td>
<td>38</td>
<td>-62</td>
<td>76</td>
<td>85</td>
<td>9</td>
<td>0.90</td>
<td>High HC included</td>
</tr>
<tr>
<td></td>
<td>43</td>
<td>-64</td>
<td>76</td>
<td>86</td>
<td>10</td>
<td>0.82</td>
<td>High HC included</td>
</tr>
<tr>
<td></td>
<td>30</td>
<td>-60</td>
<td>76</td>
<td>81</td>
<td>5</td>
<td>0.96</td>
<td>High HC included</td>
</tr>
<tr>
<td>Case 2</td>
<td>30</td>
<td>-61</td>
<td>68</td>
<td>80</td>
<td>12</td>
<td>0.95</td>
<td>CH4/CO2 only</td>
</tr>
<tr>
<td></td>
<td>30</td>
<td>-61</td>
<td>68</td>
<td>81</td>
<td>12</td>
<td>0.92</td>
<td>High HC included</td>
</tr>
<tr>
<td></td>
<td>38</td>
<td>-62</td>
<td>68</td>
<td>84</td>
<td>16</td>
<td>0.84</td>
<td>High HC included</td>
</tr>
<tr>
<td></td>
<td>23</td>
<td>-59</td>
<td>68</td>
<td>75</td>
<td>7</td>
<td>0.97</td>
<td>High HC included</td>
</tr>
<tr>
<td>Case 3</td>
<td>14</td>
<td>-57</td>
<td>42</td>
<td>65</td>
<td>23</td>
<td>0.95</td>
<td>CH4/CO2 only</td>
</tr>
<tr>
<td></td>
<td>14</td>
<td>-57</td>
<td>42</td>
<td>58</td>
<td>16</td>
<td>0.97</td>
<td>High HC included</td>
</tr>
<tr>
<td></td>
<td>20</td>
<td>-58</td>
<td>42</td>
<td>67</td>
<td>25</td>
<td>0.92</td>
<td>High HC included</td>
</tr>
<tr>
<td></td>
<td>12</td>
<td>-56</td>
<td>42</td>
<td>50</td>
<td>8</td>
<td>0.99</td>
<td>High HC included</td>
</tr>
<tr>
<td>Case 4</td>
<td>11</td>
<td>-56</td>
<td>27</td>
<td>50</td>
<td>23</td>
<td>0.95</td>
<td>CH4/CO2 only</td>
</tr>
<tr>
<td></td>
<td>11</td>
<td>-56</td>
<td>27</td>
<td>47</td>
<td>20</td>
<td>0.95</td>
<td>High HC included</td>
</tr>
<tr>
<td></td>
<td>13</td>
<td>-57</td>
<td>27</td>
<td>56</td>
<td>29</td>
<td>0.92</td>
<td>High HC included</td>
</tr>
<tr>
<td></td>
<td>09</td>
<td>-54</td>
<td>27</td>
<td>33</td>
<td>6</td>
<td>0.99</td>
<td>High HC included</td>
</tr>
<tr>
<td>Case 5</td>
<td>22</td>
<td>-59</td>
<td>56</td>
<td>75</td>
<td>19</td>
<td>0.95</td>
<td>CH4/CO2 only</td>
</tr>
<tr>
<td></td>
<td>22</td>
<td>-59</td>
<td>56</td>
<td>85</td>
<td>29</td>
<td>0.95</td>
<td>High HC included</td>
</tr>
<tr>
<td></td>
<td>28</td>
<td>-61</td>
<td>56</td>
<td>88</td>
<td>32</td>
<td>0.93</td>
<td>High HC included</td>
</tr>
<tr>
<td></td>
<td>17</td>
<td>-57</td>
<td>56</td>
<td>82</td>
<td>26</td>
<td>0.97</td>
<td>High HC included</td>
</tr>
<tr>
<td>Case 6</td>
<td>18</td>
<td>-57</td>
<td>45</td>
<td>68</td>
<td>23</td>
<td>0.95</td>
<td>CH4/CO2 only</td>
</tr>
<tr>
<td></td>
<td>18</td>
<td>-57</td>
<td>45</td>
<td>81</td>
<td>36</td>
<td>0.82</td>
<td>High HC included</td>
</tr>
<tr>
<td></td>
<td>22</td>
<td>-59</td>
<td>45</td>
<td>85</td>
<td>40</td>
<td>0.74</td>
<td>High HC included</td>
</tr>
<tr>
<td></td>
<td>12</td>
<td>-56</td>
<td>45</td>
<td>74</td>
<td>29</td>
<td>0.91</td>
<td>High HC included</td>
</tr>
</tbody>
</table>

Table 4.5. Thermodynamic performance including higher hydro carbons, at varies non-optimized conditions.

higher HC’s are present and can probably be surpassed.
Chapter 5

Performance of atmospheric upscale prototype

5.1 Introduction

Measurements on the small scale RPS described in chapter 2, showed a loss of 10\% in separation efficiency at certain process conditions [43]. The liquid removal lines became quickly clogged with liquid leading to backed-up fluid which interfered with the separation process. This was due to a lack of pressure drop over the liquid removal tube leading to the liquid collection vessel. As a result the RPS unit was filled with liquid, free rotation of the element was hindered, and waste CO\textsubscript{2} was re-entrained in the product CH\textsubscript{4} flow leading to non optimal system performance.

In chapter 4 an upscale RPS design for a feed rate of 20 MMscf/day (∼ 24.000 Nm\textsuperscript{3}/h) is described. The separator has a high liquid loading (comparable to a typical gas well). The large diameter partially filled drainage pipes have to ensure adequate removal of condensed liquid. To make use of the angular momentum of the fluid in the separator, the removal pipes are mounted tangentially. In a field installation the separated liquid will be collected in two vessels to ensure a smooth connection to the rest of the gas conditioning plant [3]. The liquid levels in the vessels will be controlled by a liquid pump and control system. Alternatively, completely filled liquid discharge pipes can be used, in combination with a degassing line from the vessel back to the separator.

In order to obtain a better understanding of the dominating physical phenomena during liquid drainage, a visually accessible industrial-scale prototype has been built, based on the upscale design presented in chapter 4. In this way potential flaws in the design can be tackled before final construction. The prototype was connected to an atmospheric test rig with water and air as working fluids. The test setup models a 80 MMscf/d installation on a natural gas well with 30 vol\% contamination. This chapter

reports two different experiments concentrating on removal of separated liquid and on separation efficiency of droplets.

The goal of the liquid removal measurement is to determine whether the designed separator can cope with the large amounts of liquid that will be encountered under real process conditions (up to 7.5 kg/s). A model is developed to describe hydrodynamic behavior of the liquid in the collecting volute.

The separation efficiency measurements are used to validate whether design calculations concerning the capture fraction of small droplets are correct. The advantage of an RPS compared to cyclones in terms of separation efficiency only becomes significant in large scale units [12]. This prototype is the first industrial-scale unit for this application. To measure the separation efficiency curve, a mist of droplets is injected at the inlet and particle concentrations and size distributions are measured in the outlet gas stream using laser diffraction. A significant amount of droplets in the range 0–10 \( \mu \text{m} \) was created, and the size distributions of these droplets could be measured accurately.

In section 5.2 the working principle and theoretical drainage model are presented. Section 5.3 describes the experimental setup and measurement methods. In section 5.4 results of the overall liquid removal measurements are presented and compared to the theoretical drainage model. Also the measured efficiencies are presented and analyzed in detail.

5.2 Background

5.2.1 Droplet removal

A schematic of the separator designed in chapter 4 is shown in fig. 5.1. The unit can be divided into three main sections, which are described below.

Pre-separator

Gas containing a mist of droplets enters the unit via a tangential inlet. This so-called pre-separator acts as a cyclone. The pre-separator first separates coarse droplets (larger than 10 \( \mu \text{m} \)) and then collects them in a stationary volute. The liquid leaves via a tangentially connected exit.

Rotating element

The gas stream, containing the remaining mist of mainly micron-sized droplets, enters a rotating element (fig. 5.2). The flow direction through the element is downward for optimal film behavior and minimum pressure drop [20]. The element is a bundle of channels with diameters of approximately 1 mm, rotating around an axis. While traveling in axial direction through the rotating channels, the droplets are driven to the channel walls by centrifugal force. On the walls the mist droplets coagulate into a thin film. Due to gravitational and shear forces, this film is forced out of the channels. At the end of the channels the film breaks up into droplets of typically 50 \( \mu \text{m} \). The rotating element thus acts as a droplet coalescer.
Post-separator

Downstream of the element the post-separator section is entered, where the liquid is actually separated from the gas stream and collected in a stationary volute (D in fig. 5.3). The post-separator can be divided into two sections, a separation section and a liquid drainage section:
1. As shown in fig. 5.3, the outer wall (B) of the rotating element extends in axial direction beyond the end of the filter element. This ensures that the gas stream leaving the element rotates as a solid body (A). The droplets breaking off at the end of the channels are centrifugally separated from the gas in this rotating field, and collected in a co-rotating film on the extended wall.

2. The liquid film leaves the gas stream at the end of the extended wall (B) towards a collection volute. The liquid, still containing significant momentum, is referred to as the “feed” to the fluid film in the volute (C). Via a tangentially connected large diameter exit (D) the liquid leaves towards a collection vessel. A restriction (E) protrudes into the product gas flow to prevent re-entrainment of liquid due to splashing.

### 5.2.2 Liquid drainage

The second part of the post-separator is particularly interesting because it significantly affects the overall separation efficiency. The aim of this section is to propose a model for the liquid drainage from the post-separator liquid collection volute. Predictions should be sufficiently accurate to come to a reliable design. The main question is the amount of liquid that can be drained from the volute via a tangential outlet. The liquid flow rate is mainly determined by the fluid velocity at the tangential outlet. Because the drainage relies on the fluid momentum, the momentum dissipation in the volute is of importance. In order to determine the momentum dissipation within the collection bucket, the contact area between the fluid and the wall has to be estimated. The contact area in combination with the local velocity and friction factor determine the local momentum dissipation. The initial feed momentum, in combination with the momentum dissipation, yield the outflow conditions and thus the liquid flow rate.
5.2 Background

Liquid film profile

Within the liquid collection volute of the post-separator, a liquid film forms (C in fig. 5.3) which is supported by the outer and bottom wall of the volute. As a first step, the geometry of the liquid film interface is determined. In fig. 5.4 the coordinate definition in a cross section of the film can be found. The following relation for the pressure within the liquid at a position \((r, z)\) below the liquid-gas interface \(z_i(r)\) can be derived [44]:

\[
p(r, z) = p_g + \rho_l g (z_i(r) - z)
\]

where \(\rho_l\) is the density of the liquid phase, \(p_g\) is the pressure of the gas phase, and \(g\) the gravitational acceleration. Consequently, the derivative in radial direction becomes:

\[
\frac{\partial p}{\partial r} = \rho_l g \frac{dz_i}{dr}
\]

The pressure variation in the radial direction due to the centrifugal force, in the case of a stationary flow, can also be described by:

\[
\frac{\partial p}{\partial r} = \rho_l \frac{v_\theta^2}{r}
\]

Combining the two gives:

\[
\frac{dz_i}{dr} = \frac{v_\theta^2}{gr}
\]

Anticipating the measurements described in section 5.3.2, we state that the liquid rotates as a solid body, in which case the tangential velocity can be given by: \(v_\theta = \omega r\). Integration of eq. (5.4) then gives the shape of the liquid film interface as:

\[
z_i(r) = \frac{\omega^2 r^2}{2g} - b
\]

with \(b\) the intersection of the profile at the \(z\)-axis. This parabolic profile can be fully described in terms of the geometric variables \(H\) and \(b\) (fig. 5.4), with \(H\) the parabolic profile height:

\[
z_i(r) = H \left( \frac{r}{R} \right)^2 - b
\]
Combination of eq. (5.5) and (5.6) yields an expression for the angular velocity $\omega$

$$\omega^2 = \frac{2gH}{R^2} \quad (5.7)$$

from which the fluid tangential velocity $v_\theta$ can be calculated as

$$v_\theta = \sqrt{\frac{2gH}{R}} \quad ; r > r_b \quad (5.8)$$

with $r_b$ the inner radius of the liquid film:

$$r_b = R \sqrt{\frac{b}{H}} \quad (5.9)$$

These relations are used in the next section to determine the mass flow rate and rate of angular momentum of the liquid film in the volute.

**Mass and angular momentum**

The mass flow rate $\dot{m}$ and the rate of angular momentum $\dot{I}$ through a cross-section of the liquid layer at station $\theta$ can be determined by integration using equations (5.6)-(5.9). The transport of mass in the film equals:

$$\dot{m}(\theta) = \rho_l \int_{r_b}^{R} \int_{0}^{z_i(r)} v_\theta dz dr = \frac{1}{2} \rho_l R \sqrt{2gH} \left( \frac{H}{2} - b + \frac{b^2}{2H} \right)$$

with $H$ and $b$ a function of $\theta$. Similarly, an expression for the transfer of angular momentum can be found:

$$\dot{I}(\theta) = \rho_l \int_{r_b}^{R} \int_{0}^{z_i(r)} v_\theta^2 r dr dz = \frac{2}{9} \rho_l gHR^2 \left( \frac{H}{6} - b + \frac{b^3}{12H^2} \right)$$

Note that the shape of the liquid layer at angular position $\theta$ is determined by the parameters $H(\theta)$ and $b(\theta)$, the values of which can be calculated from equations (5.10) and (5.11) once $\dot{m}(\theta)$ and $\dot{I}(\theta)$ are known.

Using conservation of mass and angular momentum, the mass flow rate $\dot{m}$ and rate of angular momentum $\dot{I}$ at station $\theta$, can be related to transfer of mass and momentum at a previous angular position $\theta - \Delta \theta$ in combination with the feed and dissipation in the small interval $\Delta \theta$ in the following way:

$$\dot{m}(\theta) = \dot{m}(\theta - \Delta \theta) + \dot{m}_f(\Delta \theta) \quad (5.12)$$

and

$$\dot{I}(\theta) = \dot{I}(\theta - \Delta \theta) + \dot{I}_f(\Delta \theta) - \dot{I}_{ds}(\Delta \theta) - \dot{I}_{db}(\Delta \theta) \quad (5.13)$$

with $\dot{m}_f(\Delta \theta)$ and $\dot{I}_f(\Delta \theta)$ the influx of mass and angular momentum through the liquid feed, and $\dot{I}_{ds}(\Delta \theta)$ and $\dot{I}_{db}(\Delta \theta)$ the dissipation of angular momentum through
shear forces at the side and bottom walls respectively. The mass and the momentum of the feed entering at the top in the interval $\Delta \theta$, can be described by:

$$
\dot{m}_f(\Delta \theta) = \frac{\dot{m}_f}{2\pi} \Delta \theta
$$

$$
\dot{I}_f(\Delta \theta) = \dot{m}_f \omega_f R \frac{\Delta \theta}{2\pi} = \dot{m}_f(\Delta \theta) \omega_f R
$$

(5.14)

with $\dot{m}_f$ the total liquid feed to the post separator, and $\omega_f$ its tangential velocity.

In general, momentum dissipation can be described by:

$$
\dot{I}_d(\Delta A) = \int_{\Delta A} \tau r dA = \frac{1}{2} \rho l \int_{\Delta A} v_{\theta}^2 r dA
$$

(5.15)

with $\tau$ the shear stress and $f$ the friction coefficient.

For the side wall, the dissipation of angular momentum is equal to

$$
\dot{I}_{ds}(\Delta \theta) = \rho l g R^2 \int_{\theta-\Delta \theta}^{\theta} H(H - b) d\theta' \approx \rho l g R^2 \Delta \theta \left( \bar{H}^2 - \bar{H} \bar{b} \right)
$$

(5.16)

for which the final result is a first order approximation after linearisation in $H$ and $b$ (for details see appendix H).

Similarly, the dissipation at the bottom wall, $\dot{I}_{db}(\Delta \theta)$ is found in a first order approximation as:

$$
\dot{I}_{db}(\Delta \theta) = \frac{\rho l g \bar{H} \Delta \theta}{5} \left( \bar{R}^5 - \bar{r}_b^5 \right)
$$

(5.17)
with $\bar{H}$, $\bar{b}$ and $\bar{r}_b$ averages in the interval $\Delta \theta$ (for details see appendix H).

With the derived relations, the momentum and mass balance can now be solved. The liquid film is solved numerically, starting from $\dot{m} = 0$ and $\dot{I} = 0$ at $\theta = 0$. The entire film is divided into small enough segments $\Delta \theta$ to justify the approximation in equations (5.16) and (5.17). The mass flow rate $\dot{m}$ and momentum $\dot{I}$ are obtained at angular position $\theta$, using equations (5.12) and (5.13). From these values $H(\theta)$ and $b(\theta)$ are obtained using equations (5.10) and (5.11). Since the momentum dissipation depends on the average value for $H$, $b$, and $r_b$ in interval $\Delta \theta$, a sub-iteration procedure is required to solve the problem. The model gives a theoretical prediction for the film height, width and fluid velocity which together determine the theoretical liquid drainage capacity. The model will be validated with the help of lab experiments in section 5.4.

5.3 Experimental setup

An atmospheric test setup is built using air and water as working fluids. As shown in table 5.1, the test setup is based on a contaminated gas field installation with a feed of 80 MMscf/d, contaminated with 30 mole% of CO$_2$. Expansion of the natural gas (to 20 bar and $-60 \, ^\circ C$) leads to a methane-rich gas flow rate of 0.45 m$^3$/s and a CO$_2$-rich liquid flow rate of 7.5 l/s in the form of micron-sized condensate droplets entrained in the gas stream. In the test-rig we are limited by the maximum flow rate for micron-sized droplets. That is why we perform two different measurements to assess liquid drainage capacity and separation efficiency:

1. **Liquid drainage capacity**: A large amount of water (up to 9 l/s) in the form of coarse droplets is sprayed directly upon the RPS element using three large nozzles. The large droplet size does not affect loading of the post-separator volute because as with micron-sized droplets, in the end all the liquid is collected on the extended wall of the rotating element (see fig. 5.1). Drainage of this water in the post-separator volute is studied and its performance and hydrodynamic behavior are measured.

2. **Separation efficiency**: High pressure nozzles inject a mist containing a large amount of micron-sized droplets into the air stream in the inlet piping. Based on particle diffraction, the concentration and size distribution of droplets are measured in the air stream leaving the unit. From the results a separation efficiency curve is derived.

5.3.1 Description of the test setup

A schematic of the experimental setup is introduced in fig. 5.6. The components will be described in more detail in the following subsections, dealing with the feed streams, the separation unit and the discharge streams.
### 5.3 Experimental setup

<table>
<thead>
<tr>
<th></th>
<th>Field application</th>
<th>Test rig analogue</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gas</td>
<td>CH$_4$ (70 mole% of feed)</td>
<td>Air</td>
</tr>
<tr>
<td>Contaminant</td>
<td>CO$_2$ (30 mole% of feed)</td>
<td>Water</td>
</tr>
<tr>
<td>Product gas flow rate</td>
<td>0.45 m$^3$/s</td>
<td>0.50 m$^3$/s</td>
</tr>
<tr>
<td>Liquid waste loading</td>
<td>7.5 l/s</td>
<td>9 l/s</td>
</tr>
<tr>
<td>Pressure</td>
<td>20 bar</td>
<td>1 bar</td>
</tr>
<tr>
<td>Temperature</td>
<td>-60 $^\circ$C</td>
<td>20 $^\circ$C</td>
</tr>
<tr>
<td>Max. shaft speed</td>
<td>3000 rpm</td>
<td>1500 rpm</td>
</tr>
</tbody>
</table>

Table 5.1. Operating conditions of the test unit, compared to a 80 MMscf/d field application

**Figure 5.6.** Schematic of the test rig

**Feed streams**

**Air supply:** Pressurized air from a supply at 8 bar is expanded over a globe valve to approximately 1 bar. In this way a flow rate up to 0.50 m$^3$/s can be provided to the RPS unit. The valve is actuated via pressurized air, and operated manually. A Coriolis-type mass flow meter is used to monitor the flow.

**Large liquid loading:** A large storage tank (capacity 1 m$^3$) is filled with ordinary tap water. A 5.5 kW multistage centrifugal pump provides up to 9 l/s of water to three large spray nozzles, that spray the large liquid load directly upon the RPS element in axial direction. Produced droplet sizes are in the millimeter-range. The nozzles are mounted on the top flange, see fig. 5.8. The volume flow is monitored by a variable area (VA) flow meter, and can be adjusted via a frequency converter that controls the rotational speed of the pump motor.

**Fine mist supply:** Demineralized water from another, smaller storage tank is pres-
surized by a plunger pump. A pressure reducing control valve keeps the pressure in the upstream line at 70 bar, bypassing the surplus of water back to the tank. A mist is generated in a mechanical way by 28 so-called pin jet nozzles (manufactured by BETE, type PJ6), and injected in the airflow entering the RPS unit. A pin jet nozzle (fig. 5.7) forces water through an orifice of 152 µm, and the resulting jet subsequently breaks up against a metal pin that is situated right in front of the orifice. A substantial amount of generated droplets is in the micron-range. From the specifications of the nozzle manufacturer it can be estimated that water will be injected at a constant rate of approximately 3.2 l/min. To prevent material blockage of the orifice, demineralized water is used.

Figure 5.7. Pin jet nozzle

Separation unit

The core of the test rig is a vertically mounted RPS unit, with downward flow in the coagulation channels. This unit is a dedicated test version of an industrial 20 MMscf/d design. Because the separator operates at atmospheric pressure, the casing walls are thinner. Conditions are non-cryogenic and material constraints for air-water are far less severe than for CH₄ and CO₂, not to mention H₂S. We therefore use ball bearings, rubber seals, and flanges constructed out of stainless steel.

The method of manufacturing the RPS element follows the manufacturing process of corrugated paper [17]. Taking two 0.1 mm thick stainless steel foils, one foil is corrugated and the other is not. Then the two foils together are wound up many times around an axis, and welded together continuously. The end result is a complete element, with channels that have a trapezoidal shape (fig. 5.2).

A unique feature of this RPS test unit is that it has been made optically accessible. In the shell a total of six holes are cut out that have been covered by transparent PMMA windows, see fig. 5.8.

In the industrial hydrocarbon unit the gap between the element and the cylindrical housing by which it is surrounded is smaller than 1 mm. The prototype although has a gap of 4 to 5 mm, due to difficulties in making the housing. This results in a relatively large leak flow bypassing the rotating element.

In a field application, a condensed liquid mist is entrained in the inlet gas stream. Because it enters the unit tangentially, the momentum contained in the liquid is not lost but will be employed to drive the element. In our test application though, the three nozzles spray the liquid load in the axial direction upon the RPS element, and
the complete liquid load has to be brought into rotation together with the element. The consequence is that the RPS element in the test unit has to be driven by a relatively large motor. A 15 kW electric motor is mounted on top of the unit (see fig. 5.9), and a frequency converter controls its shaft speed. An advantage of this system is that the rotational speed of the RPS element is controllable.

Discharge streams

**Water discharge:** Water separated in the post-separator is discharged directly into the main storage tank, whereas water from the pre-separator ends as a dip pipe in a secondary tank (see fig. 5.6). This dip pipe acts as a siphon water
lock so that the entire airflow is forced to flow through the RPS element. The pre-separator flow rate is derived from the rising water level in the secondary tank. Subsequently, this flow rate is subtracted from the feed rate to obtain the post-separator flow rate. The pre- and post-separator discharge pipes are connected tangentially to the separator casing. Adequate drainage is ensured by using large diameter pipes over a short horizontal distance.

**Air outlet:** The air outlet pipe passes through the sample area of a Malvern Mastersizer S which measures distribution properties of droplets entrained in the airstream (see section 5.3.3). A short distance downstream, the flow is discharged into the ambient air.

### 5.3.2 Liquid drainage

Flow rates up to 9 l/s are metered and sprayed into the rotating airstream just before it enters the RPS element, see fig. 5.8. Due to the separation performance of the pre-separator, not all the liquid sprayed on the RPS element will be drained by the post-separator. A substantial amount will be drained via the pre-separator drainage system. The liquid flow rate within the post-separator only reaches up to 5 kg/s, when the pre-separator is fed with 8.5 kg/s.

Because of the proximity of the three nozzles, the element is not homogenously loaded. This does not affect loading of the post-separator because on the extended outer wall of the rotating element (fig. 5.1) the liquid is redistributed before it enters the post-separator volute. The redistribution is performed by the film within the extend sleeve of the RPS element (B in fig. 5.1).

The film rotates as a solid body (see C in fig. 5.1) and is fed from the top. The height and width of the liquid film in the post-separator can be measured through the PMMA windows. The liquid that is expelled from the rotating element collides with the wall and loses some of its rotational velocity. The angular momentum of the liquid that is fed into the film and the velocity of the film are measured (at an angle of 180°, opposite to the exit) with help of a high speed camera. The velocity of the feed (which is the liquid entering the liquid film) and film in horizontal direction are calculated from the change of position of a visually distinguishable feature within subsequent exposures. Figure 5.10 clearly shows the turbulent, unstructured flow patterns and turbulent liquid supply.

### 5.3.3 Separation efficiency measurement

To determine the separation efficiency a fine mist is injected into the airstream entering the RPS unit (see section 5.3.1). The larger droplets in the distribution are removed in the pre-separator which uses the centrifugal force generated by the swirl resulting from the tangential inlet. The remaining small droplets are entrained in the gas stream and are subsequently (partly) removed by the RPS element.

Using a Malvern Mastersizer S 300, the volume concentration and volume distribution of the droplets leaving the RPS are measured. The measurement principle is light diffraction by droplets. Particles illuminated by a laser beam scatter light
at angles that are inversely proportional to the size of the particles. Large particles scatter at small forward angles while small particles scatter the light at wider angles. The mastersizer measures both the concentration as well as the size distribution simultaneously. The total volume concentration \( c \) in vol\% is deduced from the amount of absorbed laser light, i.e. the obscuration of the laser beam, using the Lambert-Beer law \([22]\). In the further analysis only relative concentrations are of importance. The size distribution is determined with the help of concentric detector plates. Mie theory is used to establish the relation between the scattered energy distribution on the detector plates and the polydisperse particle size distribution \([37]\).

The measured distribution is presented as a histogram of volume fractions \( \Delta f_i \), distributed over 49 bins \( i \) with the lower particle diameter \( d_{pi1} \) and the upper diameter \( d_{pi2} \). The bins are equally spaced on a logarithmic scale ranging from 0.5 to 880 \( \mu \)m. The midsize of a bin equals the geometric mean of the upper and lower limits: \( d_{pi} = \sqrt{d_{pi1} d_{pi2}} \). By definition, the sum of the fractions equals unity:

\[
\sum_{i=1}^{49} \Delta f_i (d_{pi}) = 1
\]

The measurement method is based on experiments performed by Brunazzi et al. \([45]\). In their measurements the mist was measured before and after a cyclone separator. For the measurements presented in this chapter it was not possible to measure the mist prior to the separator because the pre-separator and RPS element are one integral unit. Therefore, the reference distribution and concentration \( c_0 \) are defined at the exit of a stationary separator. When the separator is rotating, the total droplet concentrations \( c \) is measured at the same position as the reference distribution. The overall efficiency of the separator is then defined by:

\[
\eta = 1 - \frac{c}{c_0},
\]
Because of the separation performance of the pre-separator, the reference concentration $c_0$ contains only a fraction of the supplied mist. The concentration of one monodisperse fraction equals $c_\Delta f_i$. The separation efficiency of the monodisperse fractions can be described by combining eq. (5.19) with the 49 volume fractions $\Delta f_i$ with droplet sizes $d_{pi}$, resulting in:

$$\eta(d_{pi}) = 1 - \frac{c_\Delta f_i}{c_0\Delta f_{0i}}. \quad (5.20)$$

Where the subscript 0 refers to a rotational velocity of 0 rpm.

Due to the low concentration and small size of the droplets in the range 1–10 $\mu m$ the measurement is sensitive to large (typically above 100 $\mu m$) re-entrained drops. To ensure an accurate measurement, the unit has to be flushed prior to each measurement, and the lens has to be cleaned regularly. The background noise has to be measured at the same air flow as is present during the actual experiment. The unit has to run at steady state for approximately 60 s to ensure a representative measurement. Both the background recording as well as the actual measurement are averaged over 20 seconds or 10000 sweeps.

### 5.4 Results and discussion

This section is divided into two parts, first the results concerning the liquid drainage are discussed and compared to the theoretical model presented in section 5.2. Subsequently the separation efficiency of the separator is discussed.

#### 5.4.1 Liquid drainage

The liquid drainage is measured with a water/air mixture to validate the theoretical model. Measurements of the liquid film have been performed for several liquid loadings at different shaft speeds $N$. The measurement data is included in appendix H.

From the measured shape of the film we conclude that the velocity profile in the film can be described by a solid body rotation. The measurements of the width $(R-r_b)$ and the height $(H-b)$ (fig. 5.10) were done at three positions along the circumference, located at 90°, 180° and 270° from the outlet. In order to calculate the velocity of the film, the measured velocity of the feed was used as an input parameter for the model. The tangential velocity $v_\theta f$ of liquid entering the liquid film was measured at 180° using the high speed camera (see section 5.3.2)

In fig. 5.11 and 5.12, the predicted velocity $v_\theta$ at $r = R$ is compared with the measured velocity at an angle of 180° from the exit pipe. Shaft speeds of the RPS element in the range of 600-1200 rpm are included. The errors of approximately 0.4 m/s are mainly due to the velocity determination with high speed exposures. The velocities within the turbulent flow vary substantially in time and place.

In the model, the friction factor $f$ is a difficult parameter to accurately predict. In the model a friction factor of 0.05 is used. It can be seen from fig. 5.11 and 5.12 that the bulk velocity is predicted rather well. The used value for the friction factor is quite high, when compared to the flat plate analogy [46]. However, since the liquid film is
5.4 Results and discussion

Figure 5.11. Tangential liquid film velocity at the outer wall as a function of mass flow rate, for shaft speeds 600 and 1000 rpm. Comparison between model predictions and measurements at an angle of 180°.

an unsteady, decelerating flow through a highly curved volute, a substantial increase of the friction factor is expected [46]. The turbulence intensity corresponding to a friction factor of 0.05 is approximately 16% which seems reasonable when compared to the high speed exposures (for details see appendix H).

From the model described above, we can also calculate the film height, and width. The model typically over-predicts the height and under-predicts the width of the film. The reason is that the model assumes a free equilibrium surface under influence of gravitational and centrifugal force only. In reality, the water is fed into the film along side the wall containing a significant vertical momentum, thus deforming the free surface of the film. This results in a lower height and a larger width of the film.

The model is intended as a design tool for the sizing of the liquid collection volute, and is not required to accurately describe the turbulent liquid film. The size of the volute is important because a volute which is too small would cause flooding of the drainage system resulting in re-entrainment of liquid CO$_2$ into the cleaned gas stream. A volute that is too large on the other hand would dissipate all the angular momentum of the liquid (due to the increased surface area), therefore an active pumping system would be required. The model is sufficiently accurate to design the liquid collection volutes. However, only a test with liquid CO$_2$ can reveal whether the liquid removal is fast and smooth enough to prevent foaming problems.
5.4.2 Separation performance

In the previous section the performance of the RPS post-separator at large liquid loads was discussed. This section concentrates on measuring the separative performance of the RPS element with regard to micron-sized mist droplets. In this section we show the measured separation efficiency of the RPS element. Finally, the measured separation curves for a number of airflow settings and rotational speeds are compared to theoretical predictions.

Pre-separator influence

The size distribution and concentration of the mist entrained within various airflows after the RPS unit operated at 0 rpm, are shown in fig. 5.13. The figure shows that the relevant droplets (see section 2.5), in the range 1–10 \(\mu\)m, are not completely separated by the pre-separator. When the airflow increases the mean droplet diameter decreases which indicates a higher pre-separator efficiency. Note that the figure shows distributions and not concentrations. For convenience the fractions have been normalized (according to [22]) with the size of the diameter intervals (bins) on a logarithmic scale, which is a constant for the Malvern Mastersizer.

The pre-separator separates droplets by two distinct effects:
- the centrifugal field within the pre-separator cyclone,
- inertia of the droplets at the entrance of the channels.

In essence the latter is comparable to the so called impactor effect. When the element
is not rotating, the mist has to follow a sharply curved path to enter the channels. The larger droplets entrained within the gas flow will not be able to follow this curve, due to their larger inertial mass, and will migrate to the wall of the channels. At low rotational velocities the separation efficiency of the impactor effect can be on the same order of magnitude as the rotating element separation efficiency. At high rotational velocities this effect becomes negligible.

Separation efficiency curve

The measured volume concentrations are in the range $1 - 6 \times 10^{-6} \, \text{V/V}$ (based on the measurement volume and outlet pipe diameter), which only accounts for 0.5% of the injected water. Most of the liquid injected into the airstream, especially the liquid contained within the large droplets, is collected prior to the measurement. Therefore, a relatively large number of nozzles (28) is needed to obtain a measurable amount of droplets in the micron range.

Figure 5.14 shows two typical bell-shaped droplet distribution curves. One curve is measured at a rotational velocity of 600 rpm and the reference curve is measured at the same airflow at a rotational velocity of 0 rpm. The corresponding volume concentration within each bin can be found on the right axis. The volume concentrations
$c \Delta f_i$ have again been normalized with interval width on a logarithmic scale.

From both bell-shaped curves a separation efficiency curve can be calculated with the help of equation (5.20). The separation efficiency curve is depicted in fig. 5.14.

The efficiency curve increases for droplets smaller than 2 $\mu$m. Whether this increase is truly physical or due to a measurement inaccuracy for small particles is not clear. Because droplets below 2 $\mu$m are not significant for the conclusions, results below 2 $\mu$m are disregarded in the further analysis.

**Comparison with theory and discussion**

As described in section 4.3.3 the theoretical performance of the RPS constructed with concentric channels and operated within the laminar flow regime can be written as [14]:

$$d_{p50\%}^2 = \frac{13.5 \mu_g d_c}{(\rho_p - \rho_g) \pi L(1 - \epsilon)(R_o^3 - R_i^3)} \frac{\phi}{\Omega^2}. \tag{5.21}$$

In this equation $\mu_g$ is dynamic viscosity of the gas, $d_c$ the width of the channels, $(\rho_p - \rho_g)$ the difference between droplet and gas density, $L$ the channel length and $R_i$ and $R_o$ the RPS element inner and outer radius. The volume flow rate is described
by φ and Ω represents the angular velocity of the RPS element. To correct for the axial flow cross-sectional area that is occupied by the element material, the correction factor \((1 - \epsilon)\) is used (see table 5.2). The performance of trapezoidal channels can be predicted with eq. (5.21) because the travel distance for most droplet also equals \(d_c\) due to the steep side walls.

The measurements have been performed with a laminar stable flow through the RPS channels. For a laminar Hagen-Poiseuille flow with superimposed rigid rotation, a bulk Reynolds numbers \(Re_z\) and a rotation Reynolds number \(Re_\Omega\) can be defined as [47]:

\[
Re_z = \frac{v_c d_c}{\nu_g}, \quad Re_\Omega = \frac{\Omega d_c^2}{4 \nu_g},
\]

with \(v_c\) the mean gas velocity in the channel and \(\nu_g = \mu_g/\rho_g\) kinematic viscosity of the gas. Previous studies performed by Mackrodt [47] show that flow instabilities can be expected at \(Re_z > 83\) when \(Re_\Omega > 27\). DNS calculation performed by Kuerten and van Esch [8; 9] show that, in case of flow instabilities, a decrease in separation efficiency for large particles will be present. In our measurements however, \(Re_z\) can be as high as 270, but \(Re_\Omega\) never exceeds 7. Interfering vortices that could prevent droplets from reaching the wall are therefore not expected.

To calculate the theoretical performance in the laminar flow regime eq. (5.21) is used. The dimensions of the test unit (table 5.2) differ from the values stated in table 4.3. Also the typical phase properties vary considerable due to the atmospheric testing with water/air mixtures (compare table 5.2 with table E.1).

<table>
<thead>
<tr>
<th>Distance between channels</th>
<th>(d_c)</th>
<th>1.9 mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Channel length</td>
<td>(L)</td>
<td>155 mm</td>
</tr>
<tr>
<td>Element inner radius</td>
<td>(R_i)</td>
<td>90 mm</td>
</tr>
<tr>
<td>Element outer radius</td>
<td>(R_o)</td>
<td>170 mm</td>
</tr>
<tr>
<td>Element surface fraction occupied by channel walls</td>
<td>(\epsilon)</td>
<td>0.1</td>
</tr>
<tr>
<td>Particle density</td>
<td>(\rho_p)</td>
<td>1000 kg/m(^3)</td>
</tr>
<tr>
<td>Gas density</td>
<td>(\rho_g)</td>
<td>1.2 kg/m(^3)</td>
</tr>
<tr>
<td>Dynamic viscosity of the gas</td>
<td>(\mu_g)</td>
<td>18·10(^{-6}) kg/ms</td>
</tr>
</tbody>
</table>

Table 5.2. Dimensions of the RPS test unit and typical values for the relevant phase properties

The separation efficiency curve was measured for 25 different operational settings. Using eq. (5.21) the corresponding values for \(d_{p50}\) were calculated and listed in table 5.3. Because the separation efficiency is only a function of the dimensionless particle diameter \(d_p/d_{p50}\) [14], there is only a single curve that predicts the theoretical performance as a function of the normalized particle size for all operational settings. This curve can be seen in fig. 5.15. The separation curve is based on triangular channel relations with a uniform flow profile across the RPS element and a Poiseuille profile within each separation channel [14]. The triangular shape is used because it is expected to predict the wall influence of the side walls on the separation performance. The uniform flow profile is due to the flow resistance of the RPS element. All curves
Table 5.3. Predicted values of $d_{p50\%}$ ($\mu$m) for airflows ranging from 30-150 g/s and rotational velocities ranging from 200-1000 rpm

<table>
<thead>
<tr>
<th>Airflow (g/s)</th>
<th>Rotational speed (rpm)</th>
<th>200</th>
<th>400</th>
<th>600</th>
<th>800</th>
<th>1000</th>
</tr>
</thead>
<tbody>
<tr>
<td>30</td>
<td></td>
<td>3.8</td>
<td>1.9</td>
<td>1.3</td>
<td>0.9</td>
<td>0.8</td>
</tr>
<tr>
<td>60</td>
<td></td>
<td>5.4</td>
<td>2.7</td>
<td>1.8</td>
<td>1.3</td>
<td>1.1</td>
</tr>
<tr>
<td>90</td>
<td></td>
<td>6.6</td>
<td>3.3</td>
<td>2.2</td>
<td>1.6</td>
<td>1.3</td>
</tr>
<tr>
<td>120</td>
<td></td>
<td>7.6</td>
<td>3.8</td>
<td>2.5</td>
<td>1.9</td>
<td>1.5</td>
</tr>
<tr>
<td>150</td>
<td></td>
<td>8.5</td>
<td>4.2</td>
<td>2.8</td>
<td>2.1</td>
<td>1.7</td>
</tr>
</tbody>
</table>

Figure 5.15. Measured separation efficiency at 600, 800 and 1000 rpm, with gas velocities of 30, 60, 90, 120 and 150 g/s. The x axis is normalized with $d_{p50\%}$

in fig. 5.15 cross the prediction line at $d_p = d_{p50\%}$, this indicates that the $d_{p50\%}$ prediction is good.

The predicted efficiencies for droplet diameters higher than $d_{p50\%}$ are 10-20% higher than the measured values. This is probably due to the significant leak flow rate around the RPS element, i.e. between housing and rotating element, see fig. 4.3. The gap of 5 mm causes a leak flow of approximately 10%. The separation of small particles also occurs within the small gap. This results in a 100% efficiency for larger
droplets of approximately 3 to 4 times $d_{p50\%}$ (see figure 5.15). When it is required that the prototype completely removes all droplets larger than 5 µm, $d_{p50\%}$ should be chosen at $5/4 = 1.25$ µm.

### 5.5 Conclusions

1. In a rotating phase separator, separated liquid can be drained with the help of a tangential liquid outlet. The drainage utilizes the intrinsic angular momentum of the fluid when it enters the liquid collection volutes.

2. Based on the cross-sectional fluid profile in the post-separator collection volute, it can be concluded that the fluid rotates as a solid body.

3. The proposed model accurately predicts the fluid outflow velocity. This enables sizing of the liquid collection device. The model can be used as a design tool, to determine the required size of the liquid collection volute such that it allows for drainage with the help of the angular momentum contained within the liquid.

4. Making use of a large number of high pressure nozzles a significant number of micron sized droplets was generated. The droplet concentration was sufficient, to accurately measure separation efficiencies for droplet sizes down to 2 µm.

5. The measured values for $d_{p50\%}$ are in agreement with theoretical predictions.

6. Measurements show that the RPS is capable of removing micron-sized droplets effectively, which distinguishes it from cyclones. It proves that the droplet separation step in the concept of removing contaminants from natural gas by condensation is feasible.

7. Measured efficiencies are 0-20% below theoretical predictions, which can probably be gained back by reducing the leak flow rate through the gap between rotating element and housing.
Chapter 6
Discussion

In this chapter the conclusions with particular relevance to the entire process development will be restated and discussed. Special attention is given to the practical applicability of the technology for future testing and application. Suggestions for further research are included.

6.1 Lab-scale measurements

The experimental setup described within this thesis is used to demonstrate the C3cep process for the first time. The lab-scale measurements were intended to give a decisive answer whether the predicted thermodynamic performance could indeed be accomplished. Experimental results showed that separation can be achieved producing the theoretical predicted equilibrium concentrations. The working principal of CRS/C3sep, i.e. the preferential condensation of CO$_2$ and subsequent centrifugal separation of the condensed phase from natural gas, has been demonstrated at lab scale.

With high CO$_2$ concentrations, the shift of the critical point to higher temperatures and the smaller two phase area within the phase diagram makes it difficult to penetrate the phase diagram deep enough by expansion cooling performed with a JT-valve. The optimal separation conditions are determined by the thermodynamic phase equilibrium. The way to arrive at the preferred conditions can vary, a turbine expander or an external refrigerating apparatus can be used. However, a different cooling scheme can result in a different droplet size distribution.

Optimal separation conditions are a trade-off between required recovery and possible enrichment. The higher the recovery, the closer one has to be to the dew point line, resulting in a lower gas product methane concentration. When a higher enrichment is required, a higher pressure is needed and more methane will be contained within the liquid CO$_2$. 
6.2 Upscale design

Efficient phase separation for C3sep applications is possible with the RPS design as presented in chapter 4. Because of the broad range of operating conditions (due to the six test cases), an optimized design could not be produced. When more detailed test conditions are established, re-evaluation of certain design choices should be considered. With more detailed information on the real field application, certain parts can be optimised. Especially the gas inlet configuration, liquid drainage facility, and mechanical construction should be reconsidered.

The optimal element orientation is vertical and the preferred flow direction is downward to ensure a fast removal of liquid from the RPS element. The enhanced risk of re-entrainment can be dealt with by a careful design of the liquid removal system.

The liquid removal system has been successfully tested (see chapter 5) at realistic liquid loads. However, the low surface tension of liquid CO$_2$ could lead to re-entrainment and/or foaming. Different liquid removal configurations are possible. Especially the steady state pressure distribution within the drainage system is important for optimal liquid removal performance. A fast and smooth removal of the liquid CO$_2$ should be provided to avoid bubble formation and cavitation.

Instead of the u-shaped drainage volutes, a downward spiralling u-shaped volute (see fig. 6.1) could be used to enhance the liquid removal. A disadvantage of this drainage is the more complex design. The stability of the drainage could be improved due to the additional gravitational acceleration of the liquid film. More importantly, this new configuration provides the opportunity to enclose the waste exit completely, preventing re-entrainment of separated liquid CO$_2$. Complete enclosure of the exit is now possible because the exit is located below the start of the volute, and does not need to serve as a liquid collection device anymore.

The presented conceptual design has to be evaluated in more detail prior to fab-

![Figure 6.1.](image-url)
6.3 Future testing

Prior to field application a complete test at a reasonable scale should be performed. Flow rates between 1-20 MMscf/d seem most promising. The testing can be done with a one-well gas stream, a slipstream from a gas processing plant or a dedicated test loop. A comparison between a loop and field test is shown in table 6.1. When a test loop is selected, high operating and capital costs and additional operation constraints should be avoided. The separator design should be geared to fit the flow rates and compositions. A once through test-rig is more favorable in terms of equipment costs and realization time (especially when an already instrumented contaminated gas well can be used).

At a larger scale the condensation process in the induction section can change. Wall influence will diminish due to the decrease of the surface to volume ratio. This will lead to a slightly different condensation process. Expansion cooling at full scale will probably lead to smaller and more homogeneously distributed droplet sizes. To understand the process of condensation in more detail, the lab scale test facility (SRTCA) can be used. The influence of higher hydrocarbons (HC’s) on process performance can also be investigated within the current set-up with only slight modifications.

The limited enrichment measured up to date is due to test loop constraints and will not be present during field application. When deeper penetration of the phase diagram is achieved with a different cooling method, operation at different pressures and temperatures becomes possible. When a large scale test facility is constructed, testing should be possible at higher enrichments. Thus, operation at higher pressures, up to 50 bars, and temperatures as low as -65 °C should be possible. It is advisable to retain optimal experimental flexibility by using an external cooling facility.

Research and development on C3sep has now come to a point where testing at real field conditions is imminent. To further develop the separator and technology of C3sep a larger scale testing facility, preferably a once-through configuration, is inevitable.

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<th>Loop configuration</th>
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Table 6.1. Loop versus field test
6.4 C3sep applications

The phase equilibrium is most favorable at low temperatures. Therefore the separation should be performed close to the freeze out curve of CO₂ which is located around -60 °C. Depending on the required recovery, the pressure can be increased to gain a higher purity gas, or decreased to increase the recovery of methane.

As previously identified by van Wissen [5], H₂S works as an “antifreeze” for CO₂ thereby making it possible to reach higher purities of methane. This higher enrichment is possible without sacrificing methane recovery due to the reduced minimum operating temperature (as low as -80 °C). The lower operating temperatures result in a higher enrichment at equal recovery rates (up to 95 mole% CH₄). Any other component (preferably non-toxic) with a similar effect on the freeze out curve will probably have the same advantage. When this “antifreeze” (e.g. H₂S) can be retrieved from the product and waste stream it can be re-used by feeding it back upstream of the separation step.

A comparable gain in enrichment can be achieved by operating within the solid/liquid/vapour area of the phase diagram. This involves a separation step which is capable of dealing with a slurry containing solid CO₂ particles. Possible blockage due to solid particles or direct freezing of CO₂ on the equipment has to be investigated. There is a possibility that, with minor changes in the design, operation within the solid regime is possible. Probably supercritical cooling to temperatures close to -60 °C and subsequent expansion by a JT valve (left of the critical point) will be favorable. For this cooling method will result in solid formation without blockage problems due to frozen CO₂ on the heat exchanger walls. Research concerning the operation within the solid area should not be done with the lab-scale separator without thorough consideration of the slurry removal mechanism.

The presence of higher HC’s will not pose a significant constraint on the process. The theoretical performance is comparable to the pure CH₄/CO₂ case. Re-optimization of pressures and temperatures will probably lead to equal or improved efficiencies.

The CH₄ recovery r₁ only concerns the preservation of CH₄ content. It is inconclusive for the higher hydrocarbons. Most of the heavier HC are completely lost with the liquid CO₂ and even for ethane (C₂) a substantial amount is contained within the waste stream. Therefore a dedicated waste treatment should be considered when the higher HC’s represent substantial value.

When a waste stream CH₄ recuperation is applied, the optimal operating point shifts to higher pressures and therefore to higher enrichments. The liquid will then contain relatively high concentrations of methane which can be recovered, and in case of insufficient purity, fed back to the inlet of the system. This is called “asymmetric cascading” analogous to the cascading processes used for isotope separation, distillation and other separation (cascade impactors) or purification processes. This waste recuperation in combination with asymmetric cascading can provide optimal purity of the methane stream whilst reducing the methane losses via the liquid stream. An economical solution would be to optimize the process for high enrichment up to a CH₄ gas purity of 85 to 90% in one step (which is possible for a feed stream concentration of 30 to 90% CH₄). This would result in a liquid waste that still contains a
considerable amount of methane. By applying a waste regeneration step, the liquid CO\(_2\) can be produced with a purity exceeding 95\% while the regenerated methane can be mixed with the contaminated gas entering the process. For a feed concentration of 50\% CH\(_4\), the process then loses less than 4\% of the supplied CH\(_4\) via the liquid waste stream, while producing the methane with a purity of 86\%. The produced methane still needs after-treatment with a relatively small absorption unit to comply with pipeline specifications. C3sep enables reduction of high H\(_2\)S/CO\(_2\) contaminations in natural gas to a level where conventional amine-based technologies can finish the upgrading at profitable conditions.


Appendix A

Measurement data
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Appendix B

Droplet separation in a bend

In this section the influence of bends located between the JT-Valve and the separator is investigated. Since a simple bend can behave as a centrifugal separator, it is necessary to investigate whether these bends have an influence on the particle distribution of the feed stream.

The separation efficiency of a bend is calculated using the Terminal Velocity of the particle $v_T$ [22],

$$\frac{dr}{d\theta} = r \frac{v_T(r)}{v_g}$$  \hspace{1cm} (B.1)

with $r$ the radius of the bend, $\theta$ the angle and $v_g$ the gas velocity. This analysis models a 1D flow geometry with constant gas velocity ($v_g$) in the bend which is an approximation of a turbulent flow profile ($Re \approx 10^4$). Influences of turbulent fluctuations and circulation zones are disregarded.

The locus of the radial position of the droplet as a function of angular position $\theta$, 

\begin{figure}[ht]
    \centering
    \includegraphics[width=0.5\textwidth]{bend_separator.png}
    \caption{Bend separator}
    \label{fig:bend_separator}
\end{figure}
for a curved rectangular duct, with inner radius $r_i$ and outer radius $r_o$ then equals:

$$r(\theta) = r(0) + \frac{(\rho_p - \rho_g)d_p^2 v_g}{18\mu} \theta$$  \hspace{1cm} (B.2)

with $\rho_p$ the density of the particle/droplet, $\rho_g$ the density of the gas, $d_p$ the diameter of the particle/droplet and $\mu$ the dynamic viscosity of the gas. With a uniform gas velocity profile in the rectangular duct, (curved over an angle $\theta_{sep}$) this can be rewritten into:

$$\eta = \frac{(\rho_p - \rho_g)d_p^2 v_g}{18\mu(r_o - r_i)} \theta_{sep}$$  \hspace{1cm} (B.3)

$$d_{p,100\%} = \sqrt{\frac{18\mu(r_o - r_i)}{(\rho_p - \rho_g)v_g \theta_{sep}}}$$  \hspace{1cm} (B.4)

With $d_{p,100\%}$ the particle diameter that will be collected with 100% probability. A similar analysis yields the efficiency for a circular bend as [14]:

$$\eta = \frac{2}{\pi} x^2 \sqrt{1 - x^4} + \frac{2}{\pi} \sin x^2 \hspace{1cm} x = \frac{d_p}{d_{p,100\%}}$$  \hspace{1cm} (B.5)

The result of equation (B.5) is plotted in fig. B.2:

![Figure B.2. Cross-section geometry](image)

When $n$ bends are mounted in series, the separation efficiency increases and can be calculated with: $\eta_n = 1 - (1 - \eta)^n$ (see figure B.2).

The particle diameter that is collected with a 100% probability ($d_{p,100\%}$), when flowing through the induction section with a gas velocity of $v_g = 3$ m/s, is approximately 25 $\mu$m. From fig. B.2, $d_{p,50\%} = 0.65d_{p,100\%} \approx 15$ $\mu$m. When the gas velocity is lowered to $v_g = 1.5$ m/s because half of the gas condenses, the $d_{p,100\%}$ goes up to approximately 35 $\mu$m, resulting in a $d_{p,50\%}$ exceeding 20 $\mu$m per bend.
Appendix C

Wall condensation in induction section

Due to the rapid cooling in the induction section, the walls of the induction pipe can cool down to a temperature below the dew point temperature of the CO$_2$ vapour. This can lead to wall condensation, which would influence the droplet size measurements. To estimate whether wall condensation can be of influence, the mass transport to the wall is estimated in this appendix.

C.1 Quasi laminar diffusion approach

For simplicity the diffusion by turbulent flow is first approximated by a quasi laminar diffusion approach i.e. the diffusion coefficient is assumed constant with a value corresponding to the turbulent diffusion coefficient at half radius, see table C.1. The turbulent diffusion coefficient can be calculated by $D = k u_* (R - r) \approx 0.04 (R - r)$ where $r$ is the radial coordinate, $R$ is the outer radius, $k$ is Von Kármán’s constant ($k \approx 0.4$) and $u_*$ the shear velocity $u_* = \sqrt{\tau_0 / \rho}$, $\tau_0$ being the mean shear stress exerted by the fluid on the wall, and $\rho$ fluid density.

<table>
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<th>Diffusion coefficients</th>
<th>$[m^2/s]$</th>
</tr>
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<tr>
<td>Laminar gas diffusion (CH$_4$/CO$_2$ at 30 bars)</td>
<td>$D_t \approx 1 \cdot 10^{-6}$</td>
</tr>
<tr>
<td>Quasi turbulent diffusion (induction section)</td>
<td>$D_q \approx 0.02 R \approx 1 \cdot 10^{-4}$</td>
</tr>
<tr>
<td>Laminar particle diffusion [22] (1 $\mu$m)</td>
<td>$D_p \approx 2.5 \cdot 10^{-11}$</td>
</tr>
<tr>
<td>Laminar particle diffusion (0.1 $\mu$m)</td>
<td>$D_p \approx 5 \cdot 10^{-10}$</td>
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</table>

Table C.1. Diffusion coefficients
C.1.1 Cartesian

In this section a coordinate system as depicted in fig. C.1 is used, where a cross section of half of the induction section is shown. The axial coordinate is denoted by \( z \) and the complete length of the induction section by \( L \). The outer wall is located at \( x = 0 \) and \( r = R \). The concentration \( c \) of CO\(_2\) at \( x = 0 \) is assumed constant and equal to zero. This is a worst case estimate because the assumption that the concentration at the wall is everywhere zero will in practice not be valid. For this cartesian coordinate system, diffusion of concentration \( c(x, z) \) in a pipe flow with uniform velocity \( U_0 \) is described by the following convection-diffusion equation:

\[
U_0 \frac{\partial c}{\partial z} = \frac{\partial}{\partial x} D \frac{\partial c}{\partial x} \Rightarrow U_0 \frac{\partial c}{\partial z} = D \frac{\partial^2 c}{\partial x^2} \tag{C.1}
\]

with boundary conditions

\[
c(x, 0) = C_0 \tag{C.2}
\]

\[
c(0, z) = 0 \tag{C.3}
\]

\[
\frac{\partial c(R, 0)}{\partial x} = 0 \tag{C.4}
\]

Suppose that separation of variables is possible according to [48]:

\[
c(x, z) = h(x)g(z) \tag{C.5}
\]

then C.1 transforms to:

\[
\frac{U_0}{g(z)} \frac{dg(z)}{dz} = \frac{D}{h(x)} \frac{d^2 h(x)}{dx^2} \tag{C.6}
\]

For arbitrary \( x \) and \( z \) it follows that

\[
\frac{U_0}{g(z)} \frac{dg(z)}{dz} = \frac{D}{h(x)} \frac{d^2 h(x)}{dx^2} = -\lambda \tag{C.7}
\]
with \( \lambda \) the eigenvalue not depending on \( x \) or \( z \). The left hand side yields:

\[
\frac{dg(z)}{dz} = -\lambda \frac{g(z)}{U_0}
\]  

(C.8)

which can be solved for \( g(z) \) using the boundary condition \( g(0) = 1 \):

\[
g(z) = e^{-\frac{\lambda z}{U_0}}
\]  

(C.9)

The right hand side results in:

\[
\frac{d^2h(x)}{dx^2} = -\lambda h(x) \frac{D_q}{D_q}
\]  

(C.10)

with the general solution:

\[
h(x) = A\sin \left( \sqrt{\frac{\lambda}{D_q}} x \right) + B\cos \left( \sqrt{\frac{\lambda}{D_q}} x \right)
\]  

(C.11)

with boundary conditions \( h(0) = 0 \) and \( \frac{dh(x)}{dx} \bigg|_{x=R} = 0 \), it follows that

\[
\sqrt{\frac{\lambda}{D_q}} R = \frac{1}{2}\pi + n\pi ; \quad n = 0, 1, 2, ...
\]  

(C.12)

so that the eigenvalue \( \lambda \) is given as

\[
\lambda = \frac{D_q(1/2\pi + n\pi)^2}{R^2}
\]  

(C.13)

Substitution in C.5 gives:

\[
c(x, z) = \sum_{n=0}^{\infty} a_n e^{-\frac{D_q(1/2\pi + n\pi)^2 x}{R^2 U_0}} \sin \left( (1/2\pi + n\pi) \frac{x}{R} \right)
\]  

(C.14)

\[
C_0 = \sum_{n=0}^{\infty} a_n \sin \left( (1/2\pi + n\pi) \frac{x}{R} \right)
\]  

(C.15)

All eigenmodes have an associated relaxation time \( \tau \) which is given by:

\[
\tau = \frac{1}{\lambda} = \frac{R^2}{D_q (1/2\pi + n\pi)^2}
\]  

(C.16)

The upper limit is given by the first eigenmode at \( n = 0 \). For \( R = 0.005 \) this results in a relaxation time of 0.1 s. When compared to the residence time \( t = \frac{L}{U_0} = 0.5 \) s, this indicates that the characteristic time of diffusion is short enough to transport almost all of the condensable mass to the wall of the induction section.
C.1.2 Cylindrical

For the cylindrical coordinate system shown in figure C.2, the convection-diffusion equation equals:

$$U_0 \frac{\partial c}{\partial z} = D_q \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial c}{\partial r} \right)$$

(C.17)

with $r$ the radial coordinate. Analogous to section C.1.1, from separation of variables

$$c(r, z) = h(r) g(z)$$

(C.18)

it follows that

$$\frac{U_0}{g(z)} \frac{dg(z)}{dz} = \frac{D_q}{h(r)} \left[ \frac{1}{r} \frac{dh(r)}{dr} \right]$$

(C.19)

With boundary condition $g(0) = 0$, the left hand side again results in:

$$g(z) = e^{-\frac{\lambda z}{D_q}}$$

(C.20)

with $\lambda$ the eigenvalue, not dependent on $r$ or $z$. The righthand side gives:

$$\frac{D_q}{h(r)} \left[ \frac{1}{r} \frac{dh(r)}{dr} \right] = -\lambda \Rightarrow \left[ \frac{d^2 h(r)}{dr^2} + \frac{1}{r} \frac{dh(r)}{dr} \right] = -\lambda h(r)$$

(C.21)

with the general solution:

$$h(r) = C_1 j_0(\sqrt{\frac{\lambda}{D_q}} r) + C_2 y_0(\sqrt{\frac{\lambda}{D_q}} r)$$

(C.22)

with $j_0$ the first Bessel function of the first kind and $y_0$ the first Bessel function of the second kind. Using the boundary conditions $\frac{dh(0)}{dr} = 0$ and $h(R) = 0$ this results in:

$$h(R) = C_1 j_0(\sqrt{\frac{\lambda}{D_q}} R) = 0$$

(C.23)
The first eigenmode is given by:

\[ \sqrt{\frac{\lambda}{D_q}} R = 2.405 \Rightarrow \lambda = \frac{5.784 D_q}{R^2} \]  

(C.24)

so that

\[ g(z) = e^{-\frac{5.78 D_q z}{R^4 U_0}} \]  

(C.25)

The characteristic time can now be derived from:

\[ \tau = \frac{1}{\lambda} = \frac{R^2}{5.78 D_q} \approx 0.05 \text{s} \]  

(C.26)

The condensation in a pipe thus occurs approximately 2 times as fast as the condensation between 2 parallel plates.

If real laminar flow is present and the diffusion coefficient is thus a lot smaller (i.e. \( D_l = 10^{-6} \)), the characteristic time of condensation is approximately equal to 4 s. This results in a wall condensation fraction of less than 10% of the total concentration.

### C.1.3 Heat analogy with laminar flow

The mathematical description of mass transfer to a wall has an analogy with heat transfer to a wall. Based on this analogy, one can use empirical results of heat transfer and convert to problems involving mass transfer. Empirical results for heat transfer are often given in terms of the Nusselt number \( Nu \)

\[ Nu = \frac{h D_h}{k} \]  

(C.27)

with \( h \) the heat transfer coefficient, \( D_h \) the hydraulic diameter and \( k \) the thermal conductivity. According to Bejan [29] the \( Nu \)-number in a cylindrical tube with diameter \( D \) in laminar case with isothermal wall is equal to:

\[ Nu = 3.66 \]  

(C.28)

For an equivalent mass transfer problem, the Sherwood number \( Sh \) is given by:

\[ Sh = \frac{h_m D}{D_q} = 3.66 \]  

(C.29)

with \( h_m \) the local mass transfer coefficient \([\text{m s}^{-1}]\). The molar flux \([\text{mole m}^{-2} \text{s}^{-1}]\) to the wall \( j_w \) is then given by

\[ h_m = \frac{j_w}{C_w - C_b} \]  

(C.30)

with \( C_w \) and \( C_b \) the concentration at the wall and in the bulk of the fluid respectively.

The change in concentration is computed using a numerical scheme involving a finite element approximation. When the problem is solved with the quasi-laminar diffusion coefficient, this results in an almost complete condensation of all condensable vapour within the 0.5 s residence time in the induction section. When real laminar flow is considered (\( D_l = 10^{-6} \)), the computation predicts less than 7% condensation.

Both estimations agree well with the calculations performed in section C.1.2.
C.2 Turbulent diffusion

A similar comparison is made for real turbulent flow where due to the high mixing in the central region the mass transport to the wall is increased. Only the laminar boundary layer close to the wall provides a significant condensation resistance limiting the wall condensation.

In the case of turbulent pipe flow, the following Sherwood relation can be found in Bejan [29] for $0.004 \leq Sc \leq 0.1$ and $10^4 \leq Re_D \leq 10^6$.

\[
Sh = 4.8 + 0.0156 Re_D^{0.85} Sc^{0.93} \quad \text{with} \quad Re_D = \frac{U_0 D_t}{\nu} \quad \text{and} \quad Sc = \frac{\nu}{D_t} \quad (C.31)
\]

with $\nu$ the kinematic viscosity of the fluid. For typical measurement conditions, the Sherwood number is approximately equal to 26. The numerical method as mentioned in section C.1.3 is used to calculate the change in concentration. For a diffusivity $D$ of $10^{-6}$, it results in a typical wall condensation fraction of approximately 40% ($Re_D = 6 \times 10^4$ and $Sc = 0.1$).
Appendix D

Flash calculations

Isothermal flash calculations can be performed using several commercial packages. These calculations do not incorporate freeze out of one of the phases. The data presented in this chapter is calculated with in-house Shell software using a modified and improved Redlich-Kwong equation of state.

D.1 System of equations

Flash calculations solve systems of equations e.g. [49]:

\[ P_v = P_l \quad \text{mechanical equilibrium} \]  
\[ T_v = T_l \quad \text{thermal equilibrium} \]  
\[ y_i = K_i x_i \quad \text{phase equilibrium} \]  
\[ F z_i = V y_i + L x_i \quad \text{component material balance} \]  
\[ F = V + L \quad \text{total material balance} \]  
\[ h_f F + Q = h_v V + h_l L \quad \text{energy balance} \]  
\[ \sum_i y_i - \sum_i x_i = 0 \quad \text{overall summations} \]

with \( P_v \) = vapour pressure, \( P_l \) = liquid pressure, \( T_v \) = vapour temperature, \( T_l \) = liquid temperature, \( x_i \) = mole fraction of component \( i \) in liquid phase (waste), \( y_i \) = mole fraction of component \( i \) in vapour phase (product), \( z_i \) = mole fraction of component \( i \) in the feed, \( K_i \) = equilibrium ratio for vapour-liquid equilibria, \( F \) = feed flow rate, \( V \) = vapour flow rate, \( L \) = liquid flow rate, \( h \) = enthalpy of stream \( F, V \) or \( L \), and \( Q \) = rate of heat transfer.
D.2 Phase diagrams and maximum enrichment

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<th>Flash pressure ($p$) [bar, abs]</th>
<th>Flash temperature [°C]</th>
<th>Product concentration ($y_1$) [mole% CH$_4$]</th>
<th>Enrichment [mole% CH$_4$]</th>
<th>Vapour fraction</th>
<th>Liquid concentration ($x_1$) [mole% CH$_4$]</th>
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<td>82.75</td>
<td>62.75</td>
<td>0.055</td>
<td>16.32</td>
<td>0.945</td>
<td>0.229</td>
</tr>
<tr>
<td>30</td>
<td>41</td>
<td>-62</td>
<td>85.64</td>
<td>55.64</td>
<td>0.098</td>
<td>23.98</td>
<td>0.902</td>
<td>0.279</td>
</tr>
<tr>
<td>40</td>
<td>45</td>
<td>-63</td>
<td>86.70</td>
<td>46.70</td>
<td>0.161</td>
<td>31.05</td>
<td>0.839</td>
<td>0.349</td>
</tr>
<tr>
<td>50</td>
<td>47</td>
<td>-64</td>
<td>87.35</td>
<td>37.35</td>
<td>0.248</td>
<td>37.70</td>
<td>0.752</td>
<td>0.433</td>
</tr>
<tr>
<td>60</td>
<td>48</td>
<td>-65</td>
<td>87.87</td>
<td>27.87</td>
<td>0.352</td>
<td>44.89</td>
<td>0.649</td>
<td>0.515</td>
</tr>
<tr>
<td>70</td>
<td>49</td>
<td>-67</td>
<td>88.89</td>
<td>18.89</td>
<td>0.256</td>
<td>63.51</td>
<td>0.744</td>
<td>0.325</td>
</tr>
<tr>
<td>80</td>
<td>50</td>
<td>-68</td>
<td>89.58</td>
<td>9.58</td>
<td>0.443</td>
<td>72.40</td>
<td>0.558</td>
<td>0.495</td>
</tr>
<tr>
<td>90</td>
<td>50</td>
<td>-74</td>
<td>93.22</td>
<td>3.22</td>
<td>0.446</td>
<td>87.41</td>
<td>0.554</td>
<td>0.462</td>
</tr>
</tbody>
</table>

Table D.1. Maximum enrichment at specified conditions in the upper left area of the phase diagram. This data is used for fig. 2.18.

The ratio between the number of moles of methane in the product stream and the number of moles of methane in the feed stream is called recovery and can be defined as [5]:

$$ r_1 = \frac{y_1(z_1 - x_1)}{z_1(y_1 - x_1)} \tag{D.8} $$

where $x_i$, $y_i$ and $z_i$ are the mole fraction of component $i$ in the liquid waste, product and feed, where $i = 1$ refers to methane. The (methane) enrichment is the change in mole fraction (methane) between feed ($z$) and product ($y$) stream ($y - z$).
D.2 Phase diagrams and maximum enrichment

**Figure D.1.** Phase diagrams CH\(_4\)(1)/CO\(_2\)(2), left: \(z_1 = 0.1\), right: \(z_1 = 0.2\)

**Figure D.2.** Phase diagrams CH\(_4\)(1)/CO\(_2\)(2), left: \(z_1 = 0.3\), right: \(z_1 = 0.4\)

**Figure D.3.** Phase diagrams CH\(_4\)(1)/CO\(_2\)(2), left: \(z_1 = 0.5\), right: \(z_1 = 0.6\)

**Figure D.4.** Phase diagrams CH\(_4\)(1)/CO\(_2\)(2), left: \(z_1 = 0.7\), right: \(z_1 = 0.8\)
Appendix E

Test cases and properties

These values correspond to the six test cases.

<table>
<thead>
<tr>
<th>Case</th>
<th>$r_1$ [%]</th>
<th>$\rho_g$ [kg/m$^3$]</th>
<th>$\rho_l$ [kg/m$^3$]</th>
<th>$\mu_g$ [Pa s]</th>
<th>$\mu_l$ [Pa s]</th>
<th>$\sigma$ [mN/m]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Case 1</td>
<td>90</td>
<td>74.0</td>
<td>627</td>
<td>1.08·10$^{-5}$</td>
<td>1.03·10$^{-4}$</td>
<td>4.115</td>
</tr>
<tr>
<td></td>
<td>95</td>
<td>61.2</td>
<td>701</td>
<td>1.05·10$^{-5}$</td>
<td>1.25·10$^{-4}$</td>
<td>7.471</td>
</tr>
<tr>
<td></td>
<td>98</td>
<td>45.4</td>
<td>757</td>
<td>1.02·10$^{-5}$</td>
<td>1.56·10$^{-4}$</td>
<td>15.17</td>
</tr>
<tr>
<td>Case 2</td>
<td>90</td>
<td>61.8</td>
<td>758</td>
<td>1.06·10$^{-5}$</td>
<td>1.29·10$^{-4}$</td>
<td>6.458</td>
</tr>
<tr>
<td></td>
<td>95</td>
<td>46.1</td>
<td>844</td>
<td>1.02·10$^{-5}$</td>
<td>1.53·10$^{-4}$</td>
<td>11.39</td>
</tr>
<tr>
<td></td>
<td>98</td>
<td>35.0</td>
<td>871</td>
<td>1.02·10$^{-5}$</td>
<td>1.72·10$^{-4}$</td>
<td>18.27</td>
</tr>
<tr>
<td>Case 3</td>
<td>90</td>
<td>31.8</td>
<td>1070</td>
<td>1.05·10$^{-5}$</td>
<td>1.67·10$^{-4}$</td>
<td>12.78</td>
</tr>
<tr>
<td></td>
<td>95</td>
<td>24.0</td>
<td>1100</td>
<td>1.07·10$^{-5}$</td>
<td>1.71·10$^{-4}$</td>
<td>15.07</td>
</tr>
<tr>
<td></td>
<td>98</td>
<td>21.7</td>
<td>1100</td>
<td>1.08·10$^{-5}$</td>
<td>1.73·10$^{-4}$</td>
<td>16.09</td>
</tr>
<tr>
<td>Case 4</td>
<td>90</td>
<td>22.6</td>
<td>1130</td>
<td>1.06·10$^{-5}$</td>
<td>1.67·10$^{-4}$</td>
<td>14.17</td>
</tr>
<tr>
<td></td>
<td>95</td>
<td>20.4</td>
<td>1140</td>
<td>1.07·10$^{-5}$</td>
<td>1.67·10$^{-4}$</td>
<td>14.64</td>
</tr>
<tr>
<td></td>
<td>98</td>
<td>18.7</td>
<td>1150</td>
<td>1.09·10$^{-5}$</td>
<td>1.65·10$^{-4}$</td>
<td>14.88</td>
</tr>
<tr>
<td>Case 5</td>
<td>90</td>
<td>37.0</td>
<td>909</td>
<td>9.68·10$^{-6}$</td>
<td>3.13·10$^{-4}$</td>
<td>19.45</td>
</tr>
<tr>
<td></td>
<td>95</td>
<td>28.1</td>
<td>924</td>
<td>9.59·10$^{-6}$</td>
<td>3.27·10$^{-4}$</td>
<td>21.86</td>
</tr>
<tr>
<td></td>
<td>98</td>
<td>21.5</td>
<td>931</td>
<td>9.57·10$^{-6}$</td>
<td>3.38·10$^{-4}$</td>
<td>23.76</td>
</tr>
<tr>
<td>Case 6</td>
<td>90</td>
<td>27.7</td>
<td>664</td>
<td>9.31·10$^{-6}$</td>
<td>1.99·10$^{-4}$</td>
<td>27.04</td>
</tr>
<tr>
<td></td>
<td>95</td>
<td>22.5</td>
<td>679</td>
<td>9.25·10$^{-6}$</td>
<td>2.12·10$^{-4}$</td>
<td>31.62</td>
</tr>
<tr>
<td></td>
<td>98</td>
<td>15.3</td>
<td>698</td>
<td>9.11·10$^{-6}$</td>
<td>2.36·10$^{-4}$</td>
<td>39.17</td>
</tr>
</tbody>
</table>

Table E.1. Mixture properties per case
For each field composition a corresponding flash temperature and pressure can be found (corresponding to the optimization performed by [5]). The operating conditions used in this thesis are given in table E.2.

<table>
<thead>
<tr>
<th>Case</th>
<th>$r_i$ [%]</th>
<th>$Q_c$ [m$^3$/s]</th>
<th>$Q_w$ [l/s]</th>
<th>$P_{sep}$ [bar]</th>
<th>$T_{sep}$ [$^\circ$C]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Case 1</td>
<td>90</td>
<td>0.062</td>
<td>4.7</td>
<td>43</td>
<td>-64</td>
</tr>
<tr>
<td></td>
<td>95</td>
<td>0.087</td>
<td>3.1</td>
<td>38</td>
<td>-62</td>
</tr>
<tr>
<td></td>
<td>98</td>
<td>0.137</td>
<td>1.7</td>
<td>30</td>
<td>-60</td>
</tr>
<tr>
<td>Case 2</td>
<td>90</td>
<td>0.073</td>
<td>4.8</td>
<td>38</td>
<td>-62</td>
</tr>
<tr>
<td></td>
<td>95</td>
<td>0.117</td>
<td>3.3</td>
<td>30</td>
<td>-61</td>
</tr>
<tr>
<td></td>
<td>98</td>
<td>0.186</td>
<td>1.9</td>
<td>23</td>
<td>-59</td>
</tr>
<tr>
<td>Case 3</td>
<td>90</td>
<td>0.145</td>
<td>5.5</td>
<td>20</td>
<td>-58</td>
</tr>
<tr>
<td></td>
<td>95</td>
<td>0.271</td>
<td>3.6</td>
<td>14</td>
<td>-57</td>
</tr>
<tr>
<td></td>
<td>98</td>
<td>0.372</td>
<td>2.2</td>
<td>12</td>
<td>-56</td>
</tr>
<tr>
<td>Case 4</td>
<td>90</td>
<td>0.181</td>
<td>6.9</td>
<td>13</td>
<td>-57</td>
</tr>
<tr>
<td></td>
<td>95</td>
<td>0.266</td>
<td>5.7</td>
<td>11</td>
<td>-56</td>
</tr>
<tr>
<td></td>
<td>98</td>
<td>0.499</td>
<td>2.2</td>
<td>9</td>
<td>-54</td>
</tr>
<tr>
<td>Case 5</td>
<td>90</td>
<td>0.099</td>
<td>5.0</td>
<td>28</td>
<td>-61</td>
</tr>
<tr>
<td></td>
<td>95</td>
<td>0.142</td>
<td>4.6</td>
<td>22</td>
<td>-59</td>
</tr>
<tr>
<td></td>
<td>98</td>
<td>0.205</td>
<td>4.1</td>
<td>17</td>
<td>-57</td>
</tr>
<tr>
<td>Case 6</td>
<td>90</td>
<td>0.088</td>
<td>10.3</td>
<td>22</td>
<td>-59</td>
</tr>
<tr>
<td></td>
<td>95</td>
<td>0.130</td>
<td>9.4</td>
<td>18</td>
<td>-57</td>
</tr>
<tr>
<td></td>
<td>98</td>
<td>0.203</td>
<td>7.9</td>
<td>12</td>
<td>-56</td>
</tr>
</tbody>
</table>

**Table E.2.** Flow rates for different mixture compositions and recoveries
The shaft rotates with a shaft speed $\Omega$ of approximately 3000 rpm. The radial loading $F_{\text{rad}}$ is given by:

$$F_{\text{rad}} = m_{\text{unbal}} \Omega R_o^2$$ (F.1)

with $m_{\text{unbal}}$ the unbalanced mass at radius $R_o$. The mass unbalance is limited to 100 gr situated at the outer radius of the RPS element. The static axial force $F_{\text{ax,stat}}$ is completely caused by the weight of the shaft and its components.

$$F_{\text{ax,stat}} = m_{\text{tot}} g$$ (F.2)

The total weight $m_{\text{tot}}$ is 211 kg which leads to 2.1 kN static axial bearing loading. The dynamic axial force $F_{\text{ax,dyn}}$ is mainly caused by the pressure drop $\Delta P$ over the channels of the RPS element with a projection area $A$, thus:

$$F_{\text{ax,dyn}} = \Delta P A$$ (F.3)

the axial dynamical loading equals 6.3 kN. When the axial and dynamic loading are combined 8.4 kN of axial force has to be neutralized by the bearing. With the forces known a selection of different bearings can take place. Use will be made of table (F.1).
## Bearing selection

### Table F.1. Bearing selection table

<table>
<thead>
<tr>
<th>Criteria</th>
<th>Roller bearings</th>
<th>Hybrid roller bearings</th>
<th>Ceramic gliding bearings</th>
<th>AMB’s bearings</th>
<th>Gas bearings</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Economical</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Initial costs [EUR]</td>
<td>$10^2$</td>
<td>$10^3$</td>
<td>$10^4$</td>
<td>$10^5$</td>
<td>$10^4$</td>
</tr>
<tr>
<td>Lifetime expectancy</td>
<td>- -</td>
<td>+</td>
<td>+</td>
<td>++</td>
<td>++</td>
</tr>
<tr>
<td>Low maintenance</td>
<td>- -</td>
<td>+</td>
<td>+</td>
<td>++</td>
<td>++</td>
</tr>
<tr>
<td><strong>Technical</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Simplicity</td>
<td>++</td>
<td>++</td>
<td>++</td>
<td>- -</td>
<td>-</td>
</tr>
<tr>
<td>Lubrication</td>
<td>- -</td>
<td>+</td>
<td>+</td>
<td>++</td>
<td>++</td>
</tr>
<tr>
<td>Auxiliary equipment</td>
<td>++</td>
<td>++</td>
<td>++</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Reliability</td>
<td>- -</td>
<td>-</td>
<td>+</td>
<td>++</td>
<td>+</td>
</tr>
<tr>
<td>Corrosion resistance</td>
<td>- -</td>
<td>++</td>
<td>++</td>
<td>++</td>
<td>++</td>
</tr>
<tr>
<td>Thermal gradient</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>++</td>
<td>-</td>
</tr>
<tr>
<td>Large shaft diameters</td>
<td>-</td>
<td>-</td>
<td>+</td>
<td>++</td>
<td>+</td>
</tr>
<tr>
<td>Clearance [mm]</td>
<td>0.01</td>
<td>0.01</td>
<td>0.1</td>
<td>0.2-0.4</td>
<td>0.01</td>
</tr>
<tr>
<td>Rotational speed</td>
<td>3000</td>
<td>6000</td>
<td>5000</td>
<td>$10^5$</td>
<td>$10^5$</td>
</tr>
<tr>
<td>Load</td>
<td>+</td>
<td>+</td>
<td>++</td>
<td>++</td>
<td>+</td>
</tr>
</tbody>
</table>
Appendix G

Material selection

In table G.1 an overview of the applicable materials for the C3sep process is presented. The table is divided into two cases, in the first case only CH$_4$/CO$_2$ are present and in the second case H$_2$S is added. The dash (-) means that there are no practical limitations present.
<table>
<thead>
<tr>
<th>Material/name</th>
<th>Temperature</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>CH\textsubscript{4}/CO\textsubscript{2}</strong>&lt;br&gt;\textit{Caskets/spring energized lip seal}</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Buna n</td>
<td>-60 to +100 ºC</td>
<td>-</td>
</tr>
<tr>
<td>Butiel rubbers</td>
<td>-10 to +150 ºC</td>
<td>-</td>
</tr>
<tr>
<td>Viton</td>
<td>-50 to +400 ºC</td>
<td>explosive decompression with CO\textsubscript{2}</td>
</tr>
<tr>
<td>Teflon/PTFE</td>
<td>-200 to +200 ºC</td>
<td>-</td>
</tr>
<tr>
<td>Graphite</td>
<td>-240 to +800 ºC</td>
<td>-</td>
</tr>
<tr>
<td>Metal</td>
<td>-240 to +200 ºC</td>
<td>-</td>
</tr>
<tr>
<td><strong>O-rings</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Silicon/Teflon</td>
<td>-60 to +90 ºC</td>
<td>-</td>
</tr>
<tr>
<td>Fluor /Silicon</td>
<td>-60 to +130 ºC</td>
<td>-</td>
</tr>
<tr>
<td><strong>Steels</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Carbon-steel and higher</td>
<td>-</td>
<td>mind brittle breaking</td>
</tr>
<tr>
<td>Stainless (304/316)</td>
<td>-</td>
<td>mind sulphur cracking corrosion</td>
</tr>
<tr>
<td>Duplex</td>
<td>down to -60 ºC</td>
<td>mind brittle breaking</td>
</tr>
<tr>
<td>UNS 625</td>
<td>down to -100 ºC</td>
<td>-</td>
</tr>
<tr>
<td>Inconel/Monel</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Material/name</th>
<th>Temperature</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>CH\textsubscript{4}/CO\textsubscript{2}/H\textsubscript{2}S</strong>&lt;br&gt;\textit{Caskets/spring energized lip seal}</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Buna n</td>
<td>-60 to +100 ºC</td>
<td>-</td>
</tr>
<tr>
<td>Butiel rubbers</td>
<td>-10 to +150 ºC</td>
<td>-</td>
</tr>
<tr>
<td>Teflon/ptfe</td>
<td>-200 to +200 ºC</td>
<td>-</td>
</tr>
<tr>
<td>Graphite</td>
<td>-240 to +800 ºC</td>
<td>-</td>
</tr>
<tr>
<td>Metal</td>
<td>-240 to +200 ºC</td>
<td>-</td>
</tr>
<tr>
<td><strong>O-rings</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Silicon/Teflon</td>
<td>-60 to +90 ºC</td>
<td>-</td>
</tr>
<tr>
<td>Fluor /Silicon</td>
<td>-60 to +130 ºC</td>
<td>-</td>
</tr>
<tr>
<td><strong>Steels</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Carbon-steel and higher</td>
<td>-</td>
<td>mind brittle breaking</td>
</tr>
</tbody>
</table>

\textbf{Table G.1.} Sealing and construction material selection table
Appendix H

Liquid drainage model

In table H.1 the raw measurement data of the liquid drainage experiments are depicted.

<table>
<thead>
<tr>
<th>$\Omega$ [rpm]</th>
<th>$m_f$ [kg/s]</th>
<th>$v_{\theta f}$ [m/s]</th>
<th>$R - r_b$ [cm]</th>
<th>$H - b$ [cm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>600</td>
<td>2.0</td>
<td>5.9</td>
<td>3, 3, 4</td>
<td>1, 2, 3</td>
</tr>
<tr>
<td>600</td>
<td>3.0</td>
<td>5.9</td>
<td>3, 3, 3</td>
<td>1, 1, 2</td>
</tr>
<tr>
<td>600</td>
<td>4.1</td>
<td>5.9</td>
<td>4, 2, 3</td>
<td>2, 5, 6</td>
</tr>
<tr>
<td>800</td>
<td>1.9</td>
<td>6.6</td>
<td>2, 3, 3</td>
<td>1, 2, 3</td>
</tr>
<tr>
<td>800</td>
<td>2.8</td>
<td>6.6</td>
<td>2, 3, 3</td>
<td>1, 2, 3</td>
</tr>
<tr>
<td>800</td>
<td>3.8</td>
<td>6.6</td>
<td>3, 3, 4</td>
<td>1, 2, 3</td>
</tr>
<tr>
<td>1000</td>
<td>1.6</td>
<td>7.4</td>
<td>3, 3, 3</td>
<td>1, 2, 3</td>
</tr>
<tr>
<td>1000</td>
<td>2.6</td>
<td>7.4</td>
<td>3, 4, 4</td>
<td>1, 2, 3</td>
</tr>
<tr>
<td>1000</td>
<td>3.6</td>
<td>7.4</td>
<td>3, 3, 3</td>
<td>1, 2, 3</td>
</tr>
<tr>
<td>1200</td>
<td>1.4</td>
<td>8.1</td>
<td>3, 4, 4</td>
<td>0, 1, 2</td>
</tr>
<tr>
<td>1200</td>
<td>2.3</td>
<td>8.8</td>
<td>3, 3, 4</td>
<td>1, 2, 3</td>
</tr>
<tr>
<td>1200</td>
<td>3.1</td>
<td>9.6</td>
<td>2, 3, 3</td>
<td>1, 3, 4</td>
</tr>
<tr>
<td>1200</td>
<td>4.1</td>
<td>9.6</td>
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<td>0, 1, 3</td>
</tr>
<tr>
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<td>1, 1, 3</td>
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<tr>
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<td>11.4</td>
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<td>4.8</td>
<td>12.3</td>
<td>3, 3, 3</td>
<td>1, 3, 4</td>
</tr>
</tbody>
</table>

Table H.1. Post-separator liquid drainage measurements. The measurements are done at three angles around the circumference (90°, 180°, 270° from the outlet).
Transport of mass and angular momentum

The transport of mass $\dot{m}$ and angular momentum $\dot{I}$ through a cross-section of the liquid layer at station $\theta$ can be calculated as follows:

\[
\dot{m}(\theta) = \rho_l \int_{r_b}^{R} \int_{0}^{z_i(r)} v_\theta \, dz \, dr = \rho_l \int_{r_b}^{R} v_\theta \int_{0}^{z_i(r)} dz \, dr
\]

which can be integrated using equations (5.6), (5.8) and (5.9):

\[
\dot{m}(\theta) = \frac{1}{2} \rho_l R \sqrt{2gH} \left( \frac{H}{2} - b + \frac{b^2}{2H} \right)
\]

with $H$ and $b$ depending on $\theta$. Similarly, an expression for the transfer of angular momentum can be found:

\[
\dot{I}(\theta) = \rho_l \int_{r_b}^{R} \int_{0}^{z_i(r)} v_\theta^2 r \, dz \, dr
\]

\[
= 2 \rho_l g H R^2 \left( \frac{H}{6} - \frac{b}{4} + \frac{b^3}{12H^2} \right)
\]

Note that the shape of the liquid layer at station $\theta$ is determined by the parameters $H(\theta)$ and $b(\theta)$, the values of which can be calculated from equations (H.2) and (H.3) once $\dot{m}(\theta)$ and $\dot{I}(\theta)$ are known.

Control volume approach

In general:

\[
\Delta \dot{I}_d(\Delta A) = \int_{\Delta A} \tau r dA = \frac{1}{2} \rho_l \int_{\Delta A} v_\theta^2 r dA
\]

At the side wall where $v_\theta^2 = 2gH$ and $r = R$ eq. (H.4) reduces to:

\[
\Delta \dot{I}_{ds}(\Delta A) = \frac{1}{2} \rho_l \int_{\Delta A_s} 2gHR dA = \rho_l g R^2 \int_{\theta-\Delta \theta}^{\theta} H(\theta-b(\theta)d\theta
\]

After linearization of $H$ and $b$ over interval $[\theta - \Delta \theta, \theta]$:  

\[
H(\theta') = H(\theta - \Delta \theta) + \frac{\Delta H}{\Delta \theta} (\theta' - (\theta - \Delta \theta)) \quad \text{(H.6)}
\]

\[
b(\theta') = b(\theta - \Delta \theta) + \frac{\Delta b}{\Delta \theta} (\theta' - (\theta - \Delta \theta)) \quad \text{(H.7)}
\]

\[
r_b(\theta') = r_b(\theta - \Delta \theta) + \frac{\Delta r_b}{\Delta \theta} (\theta' - (\theta - \Delta \theta)) \quad \text{(H.8)}
\]

with $\Delta H = H(\theta) - H(\theta - \Delta \theta)$ and $\Delta b = b(\theta) - b(\theta - \Delta \theta)$, eq. (H.5) can be written as:

\[
\Delta \dot{I}_{ds}(\Delta \theta) = \rho_l g R^2 \Delta \theta \left( H(\theta - \Delta \theta)^2 + H(\theta - \Delta \theta) \Delta H + \frac{1}{3} \Delta H^2 \right)
\]

\[
- H(\theta - \Delta \theta) b(\theta - \Delta \theta) - \frac{H(\theta - \Delta \theta) \Delta b}{2} - \frac{b(\theta - \Delta \theta) \Delta H}{2} - \frac{\Delta H \Delta b}{3}
\]

\[
\text{(H.9)}
\]
or in first order approximation:

\[
\Delta \dot{I}_{ds}(\Delta \theta) \cong f \rho_l g R^2 \Delta \theta \left( \bar{H}^2 - \bar{H} \bar{b} \right) \tag{H.10}
\]

with \( \bar{H} \) and \( \bar{b} \) averaged values of \( H \) and \( b \) in the interval \( \Delta \theta \).

\[
\bar{H} = \frac{H(\theta) + H(\theta - \Delta \theta)}{2} \tag{H.11}
\]

\[
\bar{b} = \frac{b(\theta) + b(\theta - \Delta \theta)}{2} \tag{H.12}
\]

Similarly, the dissipation at the bottom wall, \( \Delta \dot{I}_{db}(\Delta \theta) \), can be written as:

\[
\Delta \dot{I}_{db}(\Delta \theta) = \frac{1}{2} f \rho_l \int_{\theta - \Delta \theta}^{\theta} \int_{r_b(\theta')}^R 2gH \frac{r^4}{R^2} dr d\theta' = \frac{f \rho_l g R^3}{5} \int_{\theta - \Delta \theta}^{\theta} H(\theta') d\theta' - \frac{f \rho_l g}{5R^2} \int_{\theta - \Delta \theta}^{\theta} H(\theta') r_b^5(\theta') d\theta' \tag{H.13}
\]

Using linearization of \( H \) and \( b \) in interval \( \Delta \theta \), eq. (H.13) can be written as:

\[
\Delta \dot{I}_{db}(\Delta \theta) = - \frac{(\rho_l f g \Delta \theta)}{210R^2} (-21\Delta H R^5 - 42H(\theta - \Delta \theta) R^5
+ 6\Delta r_b^5 \Delta H + 7\Delta r_b^5 H(\theta - \Delta \theta) + 35r_b(\theta - \Delta \theta) \Delta r_b^4 \Delta H + 42r_b(\theta - \Delta \theta) \Delta r_b^4 H(\theta - \Delta \theta)
+ 84r_b(\theta - \Delta \theta)^2 \Delta r_b^3 \Delta H + 105r_b(\theta - \Delta \theta)^2 \Delta r_b^3 H(\theta - \Delta \theta)
+ 105r_b(\theta - \Delta \theta)^3 \Delta r_b^2 \Delta H + 140r_b(\theta - \Delta \theta)^3 \Delta r_b^2 H(\theta - \Delta \theta)
+ 70r_b(\theta - \Delta \theta)^4 \Delta r_b \Delta H + 105r_b(\theta - \Delta \theta)^4 \Delta r_b H(\theta - \Delta \theta)
+ 21r_b(\theta - \Delta \theta)^5 \Delta H + 42r_b(\theta - \Delta \theta)^5 H(\theta - \Delta \theta)) \tag{H.14}
\]

or in first order approximation as:

\[
\Delta \dot{I}_{db}(\Delta \theta) \cong \left( \frac{\rho_l f g R^3}{5} H \Delta \theta \right) - \left( \frac{\rho_l f g \bar{H} \Delta \theta}{5R^2} \right) \left( R^5 - \bar{r}_b^5 \right) \tag{H.15}
\]

with \( \bar{r}_b \) the average value of \( r \) in the interval \( \Delta \theta \).

\[
\bar{r}_b = \frac{r_b(\theta) + r_b(\theta - \Delta \theta)}{2} \tag{H.16}
\]
Friction factor

Within the model the friction factor $f$ is a difficult parameter to accurately predict. It can be seen from fig. 5.11 that the bulk velocity is rather well predicted when a friction factor of 0.05 is used in the model. This friction factor is quite high, when compared to the drag force of a turbulent flow over a flat plate [46].

$$D = \frac{1}{2} \rho_l U^2 w L \left( 0.072 Re_L^{-0.2} \right) \quad (H.17)$$

where $w$ is the width of the plate, $L$ the length, $U$ the bulk velocity, $\rho_l$ the density of the fluid, and $f$ the average friction factor over the plate described by $f = 0.072 Re_L^{-0.2}$ where

$$Re_L = \frac{\rho_l U L}{\mu} \quad (H.18)$$

The liquid film is not steady because of the non-uniform feed, the film decelerates and flows through a heavily curved volute, therefore a substantial increase of the friction factor is expected [46]. The turbulent fluctuations within the film that lead to a friction factor of 0.05 can be estimated by [50]:

$$f = \frac{\tau}{\frac{1}{2} \rho U_0^2} \text{ with } \tau = \rho u_*^2 \rightarrow \frac{u_*}{U_0} \sim \sqrt{\frac{1}{2} f} = 0.16 \quad (H.19)$$

with $u_*$ the shear velocity which is approximately the RMS of the turbulent velocity fluctuations. This level of turbulent fluctuation seems also present within the high speed exposures. The fluctuations corresponding to the calculated values of the flat plate are in the order of 5%.
Appendix I

Fouling

<table>
<thead>
<tr>
<th>Contaminants in feed gas</th>
<th>Possible upstream removal</th>
<th>Compromises/results in</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sand (size: 10-1000 µm)</td>
<td>De-sanding</td>
<td>Erosion/wear resistance</td>
</tr>
<tr>
<td>Asphaltenes</td>
<td>Removal by acids-washing with naphtha</td>
<td>Paste-layer forming (asphaltenes coagulate at −50 °C)</td>
</tr>
<tr>
<td>Aromatics (liquid)</td>
<td>Liquid knockout</td>
<td>O-ring integrity</td>
</tr>
<tr>
<td>Mercury</td>
<td>Mercury removal bed</td>
<td>Aluminium parts/environmental</td>
</tr>
<tr>
<td>Chlorides (vapour-phase)</td>
<td>Water scrubber</td>
<td>Chlorides in combination with H₂O lead to corrosion</td>
</tr>
<tr>
<td>H₂O</td>
<td>De-hydration</td>
<td>Corrosion/hydrate forming</td>
</tr>
<tr>
<td>Condensate-liquids</td>
<td>Liquid knock-out</td>
<td>Potential loss of value (direct and CO₂)</td>
</tr>
<tr>
<td>Wax</td>
<td>Liquid knockout - wax is removed with condensates</td>
<td>Potential blockage</td>
</tr>
<tr>
<td>Salts (size: 3-2000 µm)</td>
<td>Filtration</td>
<td>Deposition and erosion</td>
</tr>
<tr>
<td>CO₂</td>
<td>n/a</td>
<td>CO₂ in combination with H₂O leads to corrosion</td>
</tr>
<tr>
<td>H₂S</td>
<td>n/a</td>
<td>H₂S in combination with H₂O leads corrosion</td>
</tr>
</tbody>
</table>

Table I.1. Fouling problems and their counter measures
Supprimer le risque et la concurrence dans les entreprises industrielles, ferait tarir tout les progrès de la civilisation.

"Hier et Demain"
by Gustave le Bon
Dankwoord

Dit proefschrift is de sluitsteen van 4 jaar plezierig werk. Om het proefschrift tot een goed einde te brengen heb ik hulp gehad van vele personen die ik hier wil bedanken.

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Guy
Guy Willems was born in Heerlen, on July 03, 1981. From 1992 to 1998 he followed his secondary education (Havo) at the scholengemeenschap Sint Michiel in Geleen. In 1998 he started his BEng study Mechanical Engineering at the Hogeschool Zuyd in Heerlen. During this study he did several internships at Wegom, Philips, DSM and SP special products.

In September 2002 he started his MSc study in Mechanical Engineering at the Technische Universiteit Eindhoven. Guy started his graduation project in 2004 in the Process Technology Group of prof.dr.ir. J.J.H. Brouwers at the Eindhoven University of Technology. The goal of the project was to determine whether a submerged steam fed Coanda nozzle could be used for ship propulsion purposes.

After graduating in December 2005, Guy started to work on his Ph.D. project, in the group of prof.dr.ir. J.J.H. Brouwers. The project concerned the further development of a novel concept for condensed rotational separation. The results of this work are described in this thesis. Parts of this work are published in: Chemical Engineering Science, volume 63, 2008, p. 3358 – 3365 (chapter 3), AIChE J., 2009, accepted for publication (chapter 2), ASME J. Fluids Eng., 2009, submitted for publication (chapter 5).