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D11.4 **Membrane-assisted CO₂-liquefaction scale up to TRL7–8**

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Abstract
<p>The membrane-assisted CO₂-liquefaction carbon capture process has been investigated by process simulations and laboratory experiments to validate its performance. To increase the TRL level to 7-8, a pilot facility with real flue gas from a cement plant in an operational environment is required.</p> <p>The membrane-assisted CO₂-liquefaction process is a hybrid between two carbon capture technologies, each of which are unfit for post-combustion carbon capture by itself. By combining the two technologies, they can both operate in their respective favourable regime of operation. The process consists of a single membrane stage for bulk separation of the flue gas from a CO₂ concentration in the range 17–20 mol%, delivering crude CO₂ with a purity up to 60–70 mol%. This bulk separation is followed by CO₂ liquefaction and two stages of phase-separation for purification up to above 99 mol% purity. The final liquid CO₂ stream can be pumped to the transport pressure and reheated in the case of pipeline transport, or directly extracted as liquid CO₂ for ship transport.</p> <p>In this work, a preliminary design of a combined membrane and CO₂-liquefaction test plant has been proposed. The baseline mass balance flowsheet has been simulated in Aspen HYSYS, and the design is further supported by recent experience from a laboratory pilot plant with a capacity of around 10 tons CO₂ per day. The simulated plant is designed for 60 % CCR and capacity to capture 100 tons CO₂ per day. A membrane area of 10 000 m² is required according to our model results. The plant delivers liquid CO₂ with at least 99.4 % purity, but higher purity may be attainable through further reduction of pressure in the low-pressure CO₂ purification separator.</p> <p>The necessary main components, equipment types and availability of off-the-shelf equipment have been investigated. Suggestions on how to control the process, based on experience from the laboratory experiments, are also provided.</p>

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1 INTRODUCTION

1.1 Background and motivation

The use of a hybrid membrane-liquefaction process for post-combustion CO₂ capture can potentially be more cost effective compared to two-stage membrane processes or standard MEA absorption processes [1].

In the membrane-assisted CO₂ liquefaction (MAL) process, the two different separation technologies can each carry out a partial separation within its favourable regime of operation. The membrane separation is generally suited for bulk separation with moderate product purity, while the low-temperature liquefaction process is well suited for purification of the CO₂ stream, from moderate purity to a high-purity product by phase separation, as described in CEMCAP deliverable number D11.3 [2]. An advantage of this process is that there are no requirements for process steam, which is normally not available in cement plants.

The MAL process has been investigated by process simulations and laboratory experiments to validate its performance. Focus has been on obtainable carbon capture ratio (CCR), CO₂ product purity and main process parameters (e.g. temperature, pressure and retention time in separation vessels) for synthesized binary membrane permeate-gas compositions. To increase the TRL level of the MAL capture technology for cement plants to 7–8, a test plant with real flue gas from a cement plant in an operational environment is required.

In this work, a test plant design has been proposed and simulated in Aspen HYSYS. The necessary main components, equipment types and availability of off-the-shelf equipment has been investigated. Suggestions on how the plant can be operated, based on experience from the laboratory experiments are also provided.

1.2 Limitations

The pilot-scale process simulation is based on the 60 % CO₂ capture case with "Typical air leak"-derived flue gas described in deliverable D11.3 [2]. The main limitations of this work are given in the following.

It is assumed that the membrane units can be scaled up to a certain modular size of spiral-wound geometry, and that the same permeability and selectivity as seen in bench-scale laboratory experiments and/or supplier data can be achieved in the large-scale pilot plant.

Several parameters, particularly in the pre-treatment part of the process, are dependent on membrane testing. This includes requirement for de-dusting, NO_x and SO_x removal, water knock-out and similar steps before the membrane units, as well as degradation and changes in permeability and selectivity of the membranes over time. As discussed in section 3.1, relevant membranes should be tested in a simpler set-up exposed to real flue gas conditions before a pilot MAL plant is built on-site.

NO_x and SO_x removal is not included in the simulation. If required, it is assumed that the same methods that are used in the MEA reference capture process can be used, described in D4.3 [2].

Simulations have been done in steady-state and equilibrium mode in Aspen HYSYS using the Peng-Robinson cubical equation of state.

1.3 Polymeric membrane unit

The same membrane model that was used for the full-scale MAL process simulations in D11.3 [2] have been used in this work. The description of the model is repeated here.

A multicomponent membrane model developed by SINTEF Energy Research has been implemented in the simulation software environment and is used to simulate the membrane separation process. See [3] for uses of this model for other post-combustion CO₂ capture applications. The membrane model is a 1-D distributed cross-flow model that makes calculations based on the following inputs:

- Feed gas mass flow, pressure and composition
- Permeate gas pressure
- Permeance of all components through membrane and membrane area, or permeance for a primary component and selectivity of all other components relative to the primary component
- Membrane surface area

Based on these inputs, the model calculates the respective chemical compositions as well as the mass flows of permeate and retentate streams. The membrane data used in the simulations are listed in Table 1.1.

Table 1.1: Membrane permeance data used in simulations.

Component	Permeance [m ³ (STP)/m ² sPa]	Permeance [m ³ (STP)/m ² bar-h]	CO ₂ selectivity over other components
CO ₂	7.5·10 ⁻⁹	2.7	1
N ₂	1.5·10 ⁻¹⁰	0.054	50
O ₂	6.0·10 ⁻¹⁰	0.216	12.5
H ₂ O	1.5·10 ⁻⁷	54	0.05

The feed pressure level for the membrane unit depends on the main blower pressure ratio, as well as the pressure drop in the direct-contact coolers. Pressure drop through the membrane units is assumed to be 10 kPa. The permeate pressure level is set to 0.2 bar(a) and is obtained by using vacuum pumps on the permeate side.

1.4 Description of the proposed MAL test plant process

The simulated pilot-scale membrane-assisted CO₂ liquefaction plant process is similar to a scaled-down version of the 60 % CO₂ capture case with "typical air leak" described in deliverable D11.3 [4]. The mass flow rate at the inlet to the simulated pilot plant is 26 662 kg/h, which is 6.9 % of the full-scale case. Flue gas feed stream conditions used in this work are given in Table 2.1. The dust fraction has been omitted from the composition in the simulation.

A process diagram of the proposed pilot-scale plant is shown in Figure 1.1. It is assumed that SO_x and NO_x removal is performed prior to this process if required. The flue gas enters the plant at 110 °C and atmospheric pressure. It is then cooled and pressurised to 34 °C and 1.60 bar(a) by

two direct contact coolers with an intermediate blower. Before entering the membrane units, the flue gas is mixed with a recycle stream from the main separator in the CO₂ liquefaction unit, which enriches the CO₂ concentration slightly, from 19.1 mol% to 19.5 mol%. The volume flowrate of the membrane stream increases by 7 % due to this recirculation. With the membrane properties given in Table 1.1, feed- and permeate-side pressure levels of 1.60 and 0.20 bar(a), respectively, 60 % CCR and a capacity to capture 100 tons of CO₂ per day, the total required membrane area is estimated to 10 000 m².

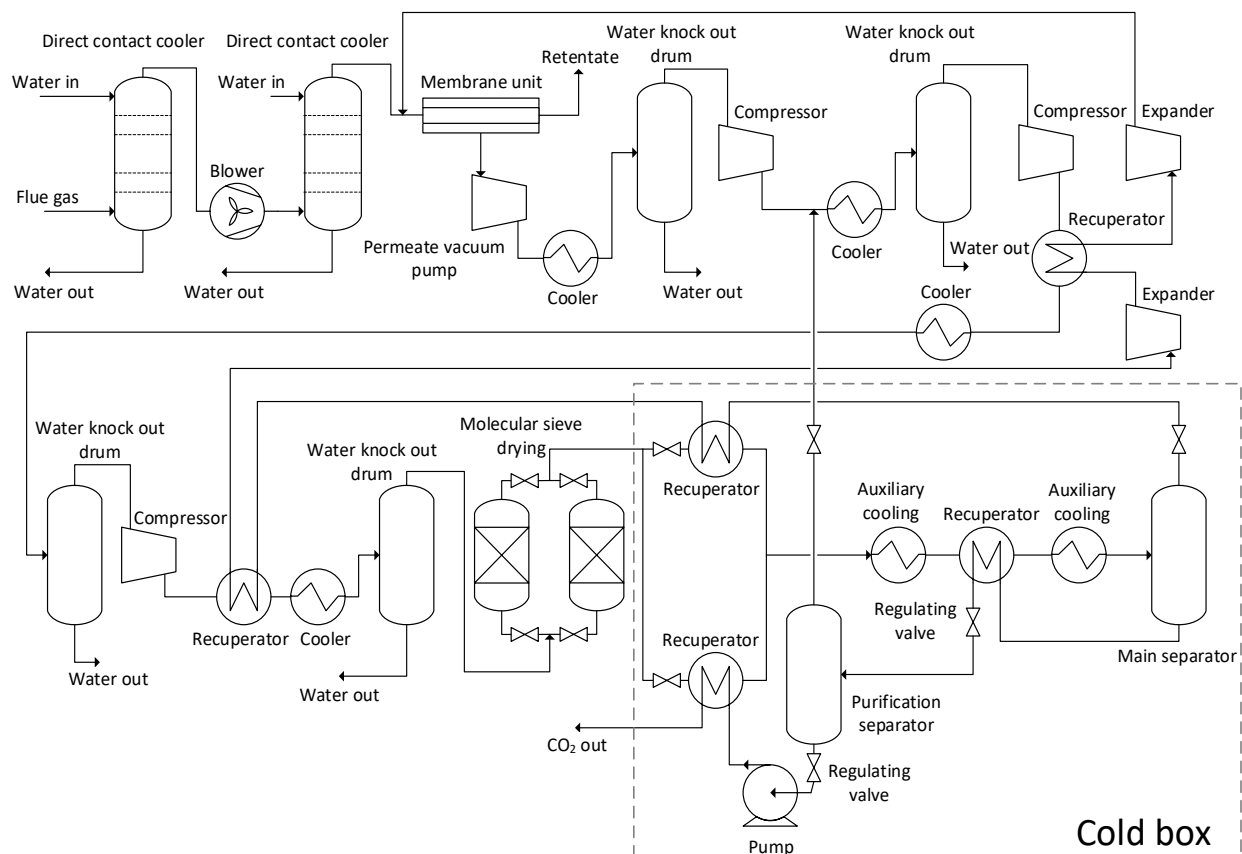


Figure 1.1: Process diagram for the membrane-assisted CO₂ liquefaction process.

The membrane enriches the gas mixture to an estimated permeate CO₂ concentration of about 62 mol%. At the vacuum pump outlet, the pressure is 1.034 bar(a) and the temperature 213 °C. In a full-scale plant, part of the generated heat from the vacuum-pump pressurisation could be utilised by heating the retentate stream in a gas-to-gas heat exchanger, before expanding it to near-atmospheric pressure in a power recovery turbine. To reduce costs and complexity, the power recovery turbine is omitted in the proposed pilot plant. The permeate stream is therefore cooled in a gas-cooler to 28 °C. All condensed water is separated in a water knockout drum before the permeate stream enters a three-stage intercooled permeate gas compressor train with water knockout after each stage. After the first compressor, the flue gas is mixed with a recycle stream from the purification separator in the CO₂ liquefaction unit, increasing the volume flow by 4.3 %.

After the final compression stage, the pressure is 31.9 bar(a) and the residual water fraction is removed in a molecular sieve desiccant bed. The dry permeate stream with a CO₂ concentration of 70.7 mol% is thereafter fed to the cold box and cooled to -54 °C in a heat exchanger network made up of internal heat recuperators as well as auxiliary refrigeration heat exchangers. At -54 °C

the gas mixture is partially condensed, and a CO₂-rich liquid phase is separated from the CO₂-depleted gaseous phase, which can be denoted tail gas, in the main separator. The latter stream has a CO₂ concentration of 24.8 mol%, which is higher than that of the flue gas feed stream, and it contains about 13 % of the CO₂ in the permeate stream. Hence, to enhance the overall CO₂ capture ratio, the tail gas stream is recycled back to the membrane inlet and mixed with the pressurised flue gas. To improve the power recovery and thus energy efficiency in the process, the tail gas is heated and expanded to a pressure level equal to that of the membrane feed in two expander stages.

The CO₂-rich liquid phase from the main separator has a purity of 95.4 mol%, provided enough retention time to approach equilibrium composition. To further increase the purity, the stream is heated to about -42 °C and throttled to 8.48 bar(a) and -55 °C before it is further separated in a secondary separator. The flash gas occurring in the throttling process contains an estimated 8.1 % of the CO₂ from the throttled stream and has an estimated CO₂ concentration of 65.6 mol%. This flash gas stream is throttled and mixed with the gas stream after the first compressor stage in the permeate compressor train.

The purified liquid CO₂ stream from the second separator has a purity of 99.4 mol%, provided enough retention time to approach equilibrium composition. Experimental results show that even higher purity is achievable by further lowering the pressure of the second separator. See section 9 for how the second separator pressure level is controlled. To pressurise this stream from 8.5 bar(a) to 110 bar(a) delivery pressure in the case of high-pressure pipeline transport, the liquid CO₂ stream is pumped in two stages. First the stream is pumped to 60 bar(a) before being heated to 3.5 °C in a recuperator. At the outlet, the liquid CO₂ is still subcooled and can thus be further pumped to 110 bar(a) in a second pumping stage. The location and discharge pressure of each pump can be configured differently if the maximum operational pressure of the exposed heat exchangers give tighter or more relaxed pressure constraints.

Some performance indicators for the simulated pilot MAL process are given in Table 1.2. The adiabatic efficiency is assumed to be 0.85 for the compressors, blower and expanders, 0.80 for the CO₂ pumps and 0.75 for the vacuum pumps.

Table 1.2: Performance indicators for the MAL pilot plant

General	Carbon capture ratio (CCR)	60 %
	CO ₂ purity	0.994 ^a
	CO ₂ captured	4170 kg/h
Refrigeration	Refrigeration duty	297 kJ/kg _{CO2}
	Refrigeration power consumption	164 kJ/kg _{CO2}
	Refrigeration system cooling demand	462 kJ/kg _{CO2}
Cooling water	Process	924 kJ/kg _{CO2}
	Refrigeration system	462 kJ/kg _{CO2}
Power consumption	Process	1058 kJ/kg _{CO2}
	Refrigeration system	164 kJ/kg _{CO2}

^a Higher purity achievable through lowered separator pressure

The power consumption and production for the components in the process are given in Table 1.3. The power demand is dominated by the duties of the blower, vacuum pumps, permeate compressors and auxiliary refrigeration. Some of the power can be recovered from turbines to reduce the net power requirement. The power requirements for pressurisation of liquid CO₂ are extremely low due to the high density and incompressibility in the liquid state.

Table 1.3: Power requirements for pilot MAL plant

Component	Power (kW)	Specific power (kJ/kg _{CO2})
Blower	371	321
Vacuum pump	330	285
Permeate compressor stage 1	173	149
Permeate compressor stage 2	181	156
Permeate compressor stage 3	176	152
CO ₂ pump stage 1	6	6
CO ₂ pump stage 2	8	7
Auxiliary refrigeration	190	164
Cooling water pumps	32	28
Tail gas expander 1	-49	43
Tail gas expander 2	-53	46
Net power (electric-to-mechanical efficiency 0.95)	1448	1250

2 BOUNDARY CONDITIONS

2.1 Incoming flue gas boundary conditions

Flue gas feed stream conditions used in this work are given in Table 2.1 below and originate from the CEMCAP framework D3.2 [5].

Table 2.1: Flue gas feed data for "Typical air leak".

		Typical air leak
Total flow rate	kg/h	388 098 (26 662)
Temperature	°C	110
CO₂	vol%	18
N₂	vol%	63
O₂	vol%	10
H₂O	vol%	9
Dust	mg/m ³ STP	10

2.2 Outgoing boundary conditions

2.2.1 CO₂ product

The CO₂ is conditioned for pipeline transport, as described in D3.2 [5], with the main requirements being:

- Pressure: 110 bar
- Temperature: < 30 °C
- > 95 vol% CO₂

To dispose of the captured CO₂, it can possibly be returned and mixed with the main flue gas from the cement plant in a catch-and-release configuration, after being heated and throttled down to around atmospheric pressure. This must be verified with the cement plant operators. Low temperatures are achieved when the high pressure captured CO₂ is throttled down. To avoid freeze out of dry-ice, the throttling should be done in two stages with intermediate heating. This could be integrated with cooling of the incoming permeate stream in the cold box to relieve the auxiliary refrigeration duty, or with other available waste heat sources in the MAL pilot plant or surrounding cement plant.

2.2.2 Decarbonised flue gas

The decarbonised flue gas leaves the membrane units at an estimated 32 °C and 1.5 bar(a). This stream can most likely be returned and mixed with the main flue gas, and released to the atmosphere through the main stack. This must be verified with the cement plant operators.

2.3 Utilities

The MAL process requires electrical power, cooling water and auxiliary refrigeration. The duties of these are given in Table 1.2 and Table 1.3. An advantage of the process is that there is no requirement for process steam, which is normally not available in cement plants.

To avoid or minimise the extent of EX zones, the refrigeration cycle may use CO₂ as working fluid. A low-pressure, low-temperature CO₂ refrigeration cycle is to be commissioned and tested in SINTEF Energy Research's laboratory pilot facility shortly after concluding the CEMCAP project. Alternatively, to reach temperatures of -55 °C or below, hydrocarbons such as ethane may be suitable as refrigerant.

3 FLUE GAS PRE-TREATMENT AND REQUIRED TESTING OF MEMBRANE EXPOSURE TO REAL CONDITIONS

The need for flue gas pre-treatment is highly dependent on the actual membrane material. Therefore, a definitive layout of the front-end pre-treatment, if needed, cannot be settled until the actual membrane material is defined.

Qualification of a given membrane material for the given application requires ageing experiments in which the decay of performance and integrity due to exposure is monitored over time. The acceptable lifetime and thus frequency of replacement will depend on e.g. the membrane material and installation and replacement cost.

This section describes a possible set-up for membrane testing and defines the most prominent pre-treatment steps that may be required.

3.1 Membrane testing

Before a fully integrated pilot test plant is built, the relevant membranes should be tested in real flue gas conditions. A principle schematic of a membrane test section is shown in Figure 3.1.

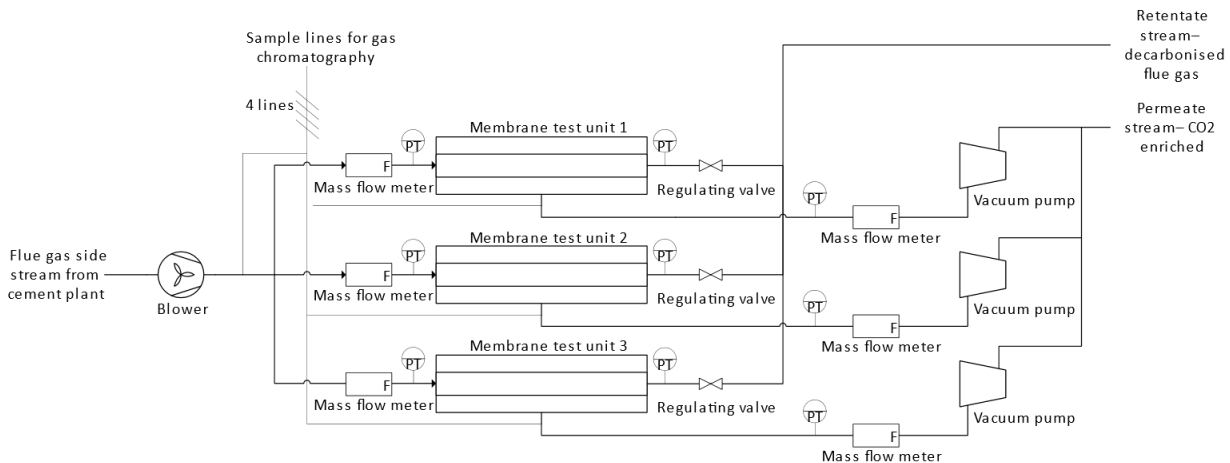


Figure 3.1: Principal schematic of membrane test section

Upon cooling, a side stream from the cement plant flue gas line enters a blower, where the gas is compressed to the appropriate feed side pressure in the membrane unit. Multiple membranes can be tested in parallel. This could be different membrane materials, or the same membrane under different operating conditions. Vacuum pumps keep the pressures on the permeate sides at the targeted levels. The test section should be well instrumented with mass flow meters, pressure sensors, temperature sensors and sample lines for gas chromatography as indicated in the schematic.

Membrane testing is necessary to see how the membranes performance parameters such as selectivity, permeability and integrity decay over time. Sensitivity to impurities such as SO_x , NO_x and dust can also be tested to determine which flue gas pre-treatment steps are necessary before the flue gas enters the membrane units.

3.2 NO_x and SO_x removal

Although polymeric membranes are assumed to have a tolerance to SO_x and NO_x, de-SO_x and de-NO_x units may be required upstream the gas separation system to curb degradation. In this case, NO_x and SO_x removal of the same type as those used in the MEA reference case modelling [6] can probably be used. NO_x is removed by increasing the de-NO_x process performed by the selective non-catalytic reduction (SNCR) system that is assumed to already be installed in the cement plant, while SO_x is removed by NaOH scrubbing in the direct-contact flue gas cooler. If NO_x concentrations below what is achieved with SNCR is required, a selective catalytic reduction system could be installed.

3.3 Water removal

The membranes' tolerance of water is highly membrane-specific. Additional process steps such as dehydration beyond regular water knock-out upstream of the membrane units needs to be decided after membrane testing and selection. It should be added that certain types, such as fixed-site carrier membranes, require water presence for activating the principal transport mechanisms [7].

3.4 De-dusting

As for water removal, the requirement for de-dusting is membrane-specific, and must be decided after membrane testing and selection. Due to environmental regulations, the dust content in the flue gas is reduced by a filter before the stack. The flue gas will also be cooled in direct contact coolers before entering the membrane section. The dust content will therefore already be very low in the flue gas entering the membranes, without additional de-dusting steps.

3.5 Removal of other trace impurities

Depending on the flue gas composition and the membrane type used, additional processing steps to remove impurities may be required (e.g. mercury).

4 FLUE GAS PRE-CONDITIONING

This section describes the processing steps after the possible removal processes for NO_x, SO_x and other impurities that may be needed, and before the flue gas enters the membrane units.

4.1 Precooling, aftercooling (and water knockout)

In the simulated pilot-scale MAL facility, the flue gas is assumed to enter the direct-contact cooler at close to atmospheric pressure and 110 °C temperature. Due to the high temperature the gas is cooled to around 36 °C before entering a blower (discussed in section 4.2) where the temperature and pressure rises to an estimated 86.5 °C and 1.65 bar(a). The flue gas is then cooled to 34 °C in a second direct-contact cooler before entering the mixer and membrane units.

4.2 Pre-compression

The volume flow out of the first direct-contact cooler is 21 533 m³/h, and the pressure level is to be increased from atmospheric pressure to approximately 1.65 bar(a). This is necessary to increase the absolute pressure difference (driving force for flux) and pressure ratio (for selectivity) across the membrane units. To increase the pressure to 1.65 bar(a) at this flowrate, a blower is suitable. There are several blower types that can be used, and manufacturers who can deliver these. The most relevant types [8] for these conditions and examples of manufacturers are provided below:

- Multistage centrifugal blower [9-11]
- Single stage centrifugal blower [12, 13]
- Rotary lobe positive displacement blower – may not be suitable if there is dust in the gas [14, 15].

4.3 Mixer of feed gas and recycled tail gas from CO₂ separation

Before entering the membrane units, the flue gas is mixed with a recycle stream of tail gas from the main separator in the CO₂ liquefaction unit. The CO₂ concentration in the recycled gas stream depends on the conditions in the main separator. With the calculations used as basis in the present design, a slight enrichment of the CO₂ concentration in the aggregate membrane feed stream is expected – from 19.1 mol% to 19.5 mol%. Correspondingly, the volume flowrate increases by around 7 %. A static mixer can be used to assure a homogenous mixture enters the membrane units.

5 MEMBRANE UNIT

In the simulated pilot facility, the gas stream enters the membrane units with a flowrate of 7.68 kg/s (about 14 000 m³/h volume flowrate) at around 33 °C and 1.6 bar(a). The gas composition is given in Table 5.1.

Table 5.1: Membrane inlet composition

Component	Mole fraction
CO ₂	0.195
Nitrogen	0.656
H ₂ O	0.0317
Oxygen	0.118

To achieve 60 % CCR in the simulated pilot facility, with the membrane properties given in Table 1.1 combined with respective feed- and permeate-side pressure levels of 1.60 and 0.20 bar(a), a membrane area of around 10 000 m² is required. Membrane type and shape depends on availability from manufacturers and must be determined after membrane testing. The number of membrane units necessary depends on the packing density, and a wide interval, typically 200–1000 m²/m³) for spiral-wound membranes is indicated in the literature. As an example, if high-density spiral-wound membrane units with a length of 1.0 m, a diameter of 0.25 m, packing density around 1000 m²/m³ and a membrane area of about 50 m² per unit are used, approximately 200 membrane units will be necessary. If the available packing density is around 500 m²/m³, this will double the required units. The units can be stacked and bundled in racks as principally illustrated in Figure 5.1.

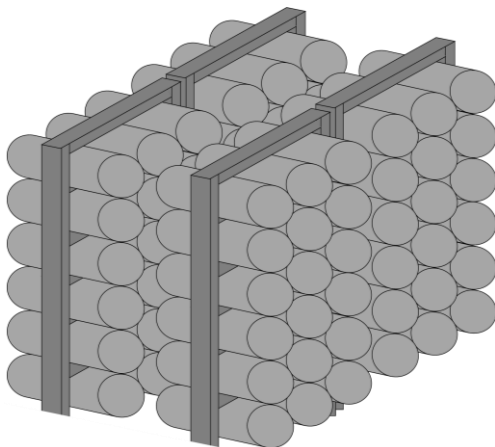


Figure 5.1: Possible principal configuration of membrane units stacked in racks

6 POST-CONDITIONING OF FLUE-GAS RETENTATE

To partly recover pressure and heat energy through conversion to shaft power, the retentate stream can be heated against the permeate stream after the vacuum pump and expanded in a power-generating turbine before being vented to the atmosphere. This design is proposed for the full-scale facility described in [4]. For the pilot scale facility, however, the general stream flowrates are substantially smaller, and it is therefore proposed to omit these components for economic reasons and since it is debatable whether a final retentate-gas expander adds significant value to the demonstration aspects. A short review of the main components is nevertheless given below.

In the simulated pilot facility, the retentate stream exits the membrane units with a flowrate of 5.87 kg/s (about 12 000 m³/h) at around 33 °C and 1.5 bar(a). The retentate gas composition is given in Table 6.1.

Table 6.1: Membrane retentate composition

Component	Mole fraction
CO ₂	0.09
Nitrogen	0.78
H ₂ O	0.01
Oxygen	0.12

6.1 Retentate heating

On the opposite permeate side, the outlet temperature of the vacuum pumps is high due to the high compression ratio. To partially recover this thermal energy, the permeate gas can be cooled against the retentate gas stream in a gas-to-gas heat exchanger. The retentate stream, which contains a considerable amount of pressure energy, can then be expanded to recover also parts of the thermal energy transferred from the hot permeate gas. This also increases the final outlet temperature of the retentate gas and contributes to mitigating potential buoyancy issues when the gas is released to the atmosphere. In a pilot plant, without heating of the retentate gas, these issues could be mitigated by returning the retentate stream to the main flue gas line of the cement plant before entering the main stack. This should be unproblematic, as it is a small stream of 5.87 kg/s, and since it has considerable gauge pressure compared to the flue gas. This must however be verified with the cement plant operator before an integration can be made.

6.2 Retentate expansion

In the simulation basis for the pilot facility, the retentate gas exits the membrane units at around 1.5 bar(a) pressure. The pressure could be somewhat higher depending on the selected membrane and desired carbon capture ratio. It can be considered to use a turbine, for instance directly coupled with an inlet fan stage, to recover some of the power used for compression of the flue gas feed. The expected power output, with or without heating the stream prior to expansion, is expected to be in the range 150–200 kW for the suggested capacity of the pilot facility.

7 PERMEATE CONDITIONING

A permeate-side pressure of 0.2 bar(a) is used as a basis for the membrane units in the pilot facility simulations. The permeate stream has a flowrate of 1.81 kg/s (about 22 400 m³/h volume flowrate at vacuum conditions) and around 33 °C temperature. The composition of the stream derived from the basis simulations is given in *Table 7.1*.

Table 7.1: Membrane permeate composition

Component	Mole fraction
CO ₂	0.62
Nitrogen	0.15
H ₂ O	0.13
Oxygen	0.10

7.1 Vacuum pump

The vacuum pump is required to provide a moderate vacuum level of about 0.2 bar(a) on the permeate side of the membrane units. It must also be able to handle the relatively large flow rate of 22 400 m³/h. The membrane units will be highly modularised, so a modularised vacuum pumping system could also be used if found advantageous in the selection of vacuum-pumping machinery. There are however several available vacuum pump types that can handle the flowrate and vacuum levels required for the pilot scale facility. Some of the vacuum pump types and examples of suppliers are given here:

- Multistage centrifugal blower [11] [9]
- Liquid ring pumps [9]
- Screw pumps [9]

7.2 Cooler and water knockout

Once brought to a pressure slightly above atmospheric pressure, the permeate gas stream has a volume flowrate of around 6900 m³/h. The cooler inlet temperature is estimated to 213 °C and is cooled to 28 °C. The duty of the gas cooler is estimated to 550 kW. An absolute pressure loss of 0.02 bar is assumed in the simulation of the gas cooler. A shell-and-tube type heat exchanger with cooling water can be suitable for this purpose. A knockout drum with sufficient demisting capacity is installed after the cooler to remove condensed water and droplets before the permeate gas enters the first compressor stage.

8 LOW-TEMPERATURE PERMEATE PROCESSING

Before entering the cold box, the permeate stream must be compressed, and the water concentration reduced to below 1 ppm.

8.1 Permeate compression, intercooling and water knock-out

The compression is performed by three compressors with intercooling and water knockout after each compressor. The permeate stream enters the first compression stage at 28 °C, about atmospheric pressure and is compressed to 32 bar(a). Both reciprocating and centrifugal compressors can be suitable for the flow rate and pressure ratio desired in the pilot scale facility. Centrifugal compressors are recommended due to cost and maintenance, although the efficiency is expected to be lower for this flow rate. Centrifugal compressors will also be suitable in a full-scale facility, and it is therefore beneficial to use the same machinery type in a pilot, to gain experience on controlling the process. With centrifugal compressors, three compressors with two stages in each and a pressure ratio of 1.77 per stage can be suitable. The compressor train can also be made up of one single-shaft compressor with sequences of outflows from the compressor to the intercoolers and knockout drums, and back to the compressor.

After the first compressor stage, the gas stream is mixed with a recycle stream from the purification separator in the CO₂ liquefaction unit, which increases the volume flow by 4.3 %. This stream is throttled down from 8.5 bar(a). In a full-scale facility this stream would be recompressed and recycled to the inlet of the cold box. This is not recommended in the pilot-scale facility to save the cost of an additional recycle compressor.

8.2 Permeate Dehydration

The permeate stream enters the dehydration unit with a mass flow rate of 1.837 kg/s (118.1 m³/h) at 28 °C and 31.27 bar(a). The stream still contains a water mole fraction of 0.0019. If water ice formation is to be completely avoided, the water concentration must be reduced to around 0.6 ppm or lower as seen in Figure 8.1, since temperatures as low as -55 °C are applied in the liquefaction and phase separation part of the process. To achieve this, solid desiccant bed adsorption is required. The dehydration unit removes about 1.6 g/s of water.

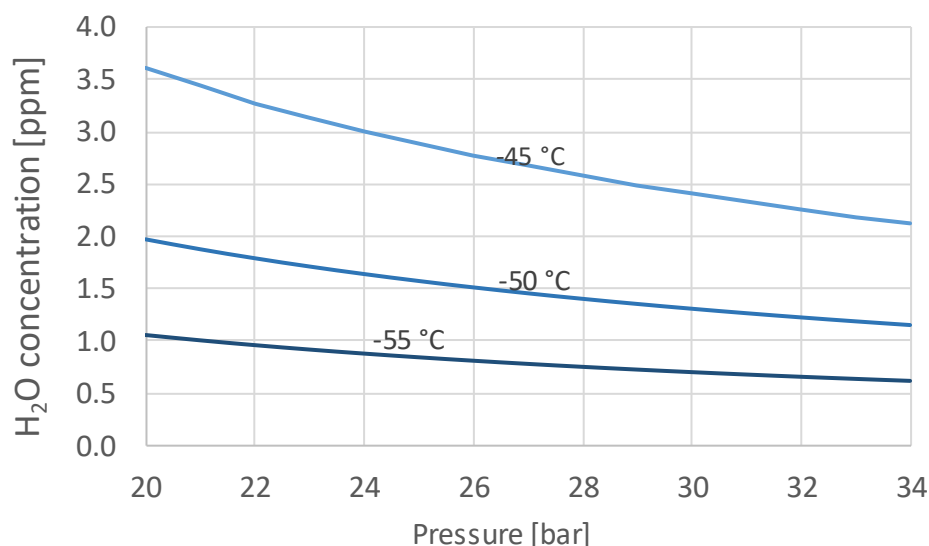


Figure 8.1 Maximum water concentration to avoid ice formation for different temperature and pressure levels.

Molecular sieve adsorbents that can provide dew point levels down to below $-70\text{ }^{\circ}\text{C}$ is conventional equipment and can be purchased from multiple suppliers. Two units in parallel are necessary such that one can be regenerated while the other is active.

A slip stream of captured liquid CO_2 can be heated with electric heating elements and used as sweep gas to regenerate the desiccant beds. Alternatively, or as backup to the self-sufficient regeneration system based on captured CO_2 , nitrogen or CO_2 can be purchased from a gas supplier and installed in sufficient quantities to be used as regeneration gas.

8.3 Cooling and condensation

The heat exchanger network for cooling and condensation of the permeate gas – recuperators and auxiliary refrigeration heat exchangers – can be configured as a network of two-stream heat exchangers, as indicated in Figure 1.1. In the proposed scale of the pilot CO_2 capture plant, the duties of these two-stream heat exchangers are between approximately 30 kW and 270 kW. These duties are well within the range of plate heat exchangers, which is standard off-the-shelf equipment available from multiple suppliers [16-18]. Brazed or welded plate heat exchangers should be used.

8.4 Auxiliary refrigeration

The auxiliary refrigeration unit, with a cooling duty of 344 kW is conventional technology and can be ordered based on a tender with specifications. A prototype refrigeration unit with CO_2 as working fluid will be tested in SINTEF Energy Research's laboratory pilot plant. It is preferable to use a non-combustible working fluid to avoid ATEX requirements and since there will be direct heat exchange with an oxygen-containing process fluid. If temperatures below what can be provided with CO_2 as refrigerant fluid is required in the refrigeration cycle, hydrocarbons (e.g. ethane) can be considered.

8.5 Tail gas heating and expansion

The proposed pilot plant consists of two power recovery turbines for converting pressure and thermal energy in the tail gas stream into shaft power. The pressure ratio across each expander is approximately 1:4, and the specific enthalpy drop is 90–100 kJ/kg. The molar mass of the tail gas is about 33.1. Radial gas turbines are well known and mature technology within several cryogenic industries such as helium liquefaction, hydrogen liquefaction and air separation. The power output from each turbine is about 50 kW and depending on the complexity and cost considerations, the shaft power could be either dissipated by brakes, converted to electric power by generators or coupled with a single-stage permeate compressor. If, like for the retentate gas leaving the membrane unit, the deployment of power recovery expanders is considered to be too complex for the pilot plant design, they can simply be excluded from the scheme and replaced with throttling valves. This will simplify the process control structure.

9 PROCESS CONTROL STRATEGIES

A control system for the membrane-assisted liquefaction pilot plant is proposed in the following sections. The description of the control system is divided into two sections. One for the membrane part and one for the liquefaction and phase separation part of the process.

The flue gas entering the pilot plant can be controlled by a control valve at the inlet to the plant. The valve should be installed before the flue gas pre-treatment steps.

9.1 Membrane separation unit

The feed volume flow can be controlled by the control valve on the line entering the pilot plant, as mentioned above. To control the feed gas pressure to the membrane unit, a control valve on the retentate outlet line can be used, as illustrated in Figure 9.1. The vacuum pump should be set to keep the vacuum pressure level (0.2 bara for our case).

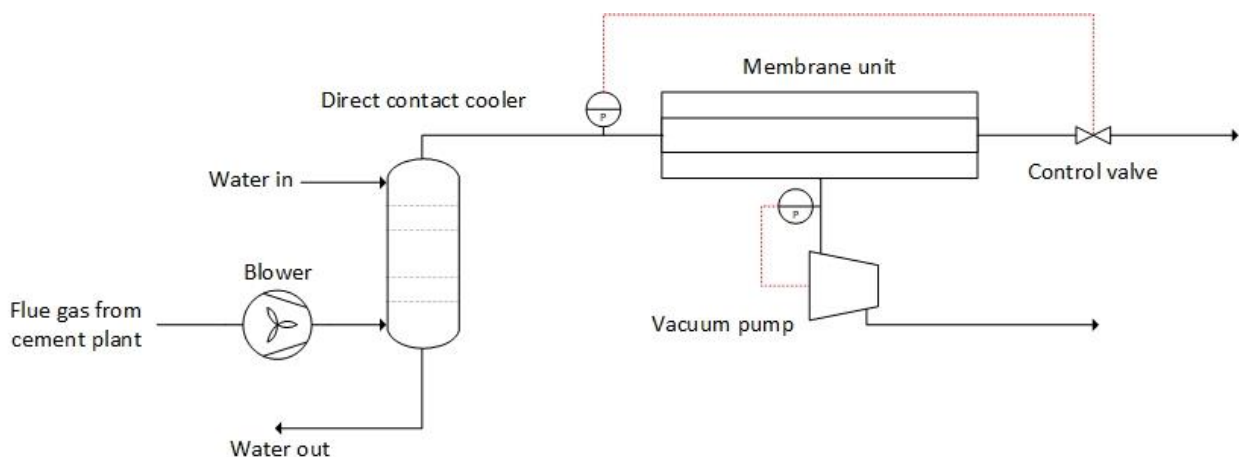


Figure 9.1: Principle schematic of control system for the membrane part of the pilot plant

9.2 Liquefaction and phase separation unit

The liquefaction and phase separation unit can have a similar control system to that used in the laboratory pilot unit, described in deliverable D11.2 [19]. A brief description of the laboratory pilot control system is summarized in section 9.2.1 before a proposal for a control system for a pilot plant is given in section 9.2.2.

9.2.1 Laboratory rig control system

A principle schematic of the laboratory pilot with control system is shown in Figure 9.2.

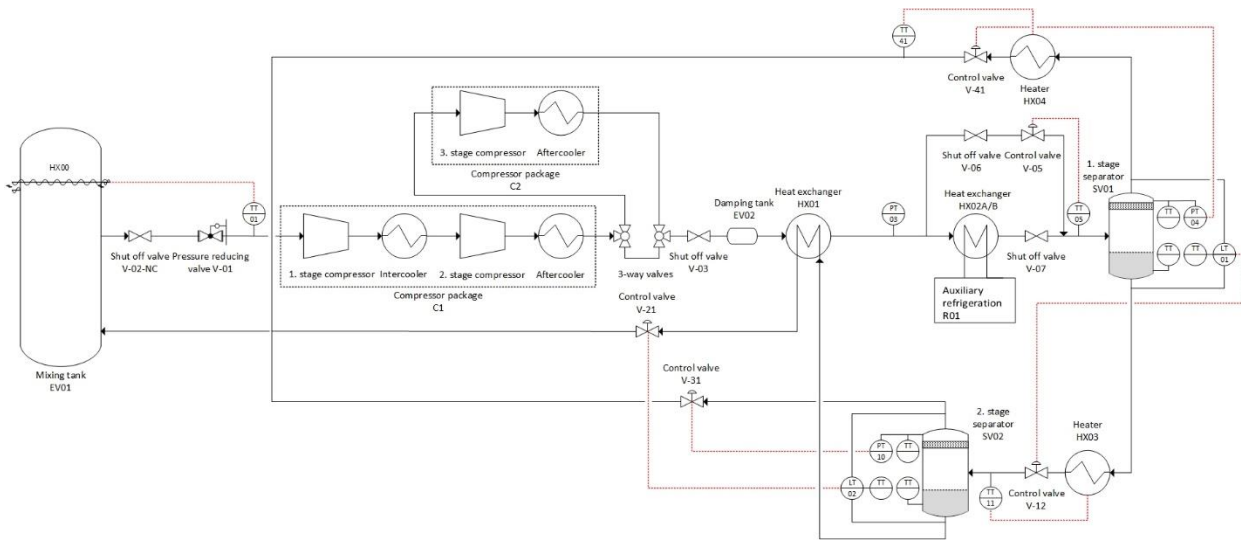


Figure 9.2: Principle schematic of CO₂ liquefaction laboratory rig with control system

The compressors have variable frequency drives (VFD) which are used to control the mass flow rate in the rig. The operating range of the compressors are between 50 % and 100 % of full capacity. A pressure reducing valve ensures that the suction pressure remains at the desired level. The trace heater, HX00, controlled by temperature transmitter TT01, keeps the inlet temperature to the low-pressure compressor above the specified minimum suction temperature. Control valves on the cooling water lines, controlled by temperature sensors at the gas outlets of the inter- and aftercoolers are used to control the outlet temperature from the compressor packages.

Control valve V-05, controlled by temperature transmitter TT05, keeps the inlet temperature to separator SV01 at the desired temperature. An auxiliary refrigeration unit will be connected to the rig at a later stage and used to cool the gas mixture to the separation temperature. The pressure in the separator is controlled by control valve V-41, connected to pressure transmitter PT04. A trace heater (HX04), controlled by temperature transmitter TT41, is used to heat the gas stream returning to the mixing tank EV01, and ensures that there is no formation of dry ice when the gas is throttled to the tank pressure. Control valve V-12, connected to the liquid level meter LT01, controls the liquid level in SV01.

Immersion heater HX03, controlled by temperature transmitter TT11, keeps the temperature at the inlet to SV02 at the desired level, and ensures no dry ice is formed in the throttling process. Control valve V-31, connected to pressure transmitter PT10, controls the pressure in SV02. The liquid level is controlled by V-21, connected to liquid level meter LT02.

National Instruments LabVIEW software is used to control the rig. The control system is set up with PID regulators where the controlling signal (e.g. pressure or temperature transmitter) can be changed, and the proportional, integral and derivative terms specified. The derivative terms have currently been set to zero for all regulators. Stable operation with liquid levels in both separators has been achieved for both a constant composition at the inlet to SV01, and for varying composition.

9.2.2 Pilot plant control system

The liquefaction unit in the pilot plant can be controlled by a similar system to that described for the laboratory rig in section 9.2.1. A principal schematic of the liquefaction unit with control system is given in Figure 9.3. Control valves on the liquid outlet lines from the process gas main and purification separator control the liquid levels, and control valves on the gas outlet lines control the pressure levels. A recuperator on the liquid line before throttling to the second stage separation pressure is used to keep the stream at the inlet to the purification separator at the desired temperature and avoid freeze out of dry ice. The capacity of the recuperator, and the temperature at the inlet to the purification separator, can be controlled by controlling the capacity of the auxiliary refrigeration.

The capacity of the liquefaction part of the plant can be controlled either by using centrifugal compressors with variable frequency drives or fixed-speed motors combined with throttling valves at the compressor inlets to reduce the suction pressure and thereby the capacity. Using compressors with VSD provides good operating flexibility to run the plant at different capacities. The fixed-speed motor option has a lower capital expenditure compared to VSD, but is more power demanding. Fixed-speed motors are assumed in Figure 9.3. The gas temperatures out of the compressors can be controlled by controlling the cooling water flow rate in the compressor inter- and aftercoolers.

Switching between the parallel molecular sieve beds happens when the active bed is nearly saturated. The gas flow is then redirected to a regenerated molecular sieve bed, while the active bed is switched into regeneration mode.

The distribution of gas between the two parallel recuperators directly behind the molecular sieve beds are controlled by control valves on the inlet lines to the recuperators. These can be set manually to suitable constant values.

The refrigeration duty of the first auxiliary refrigerator is controlled by a temperature transmitter on the inlet to the purification separator. The second auxiliary refrigerator is controlled by a temperature transmitter on the inlet to the main separator.

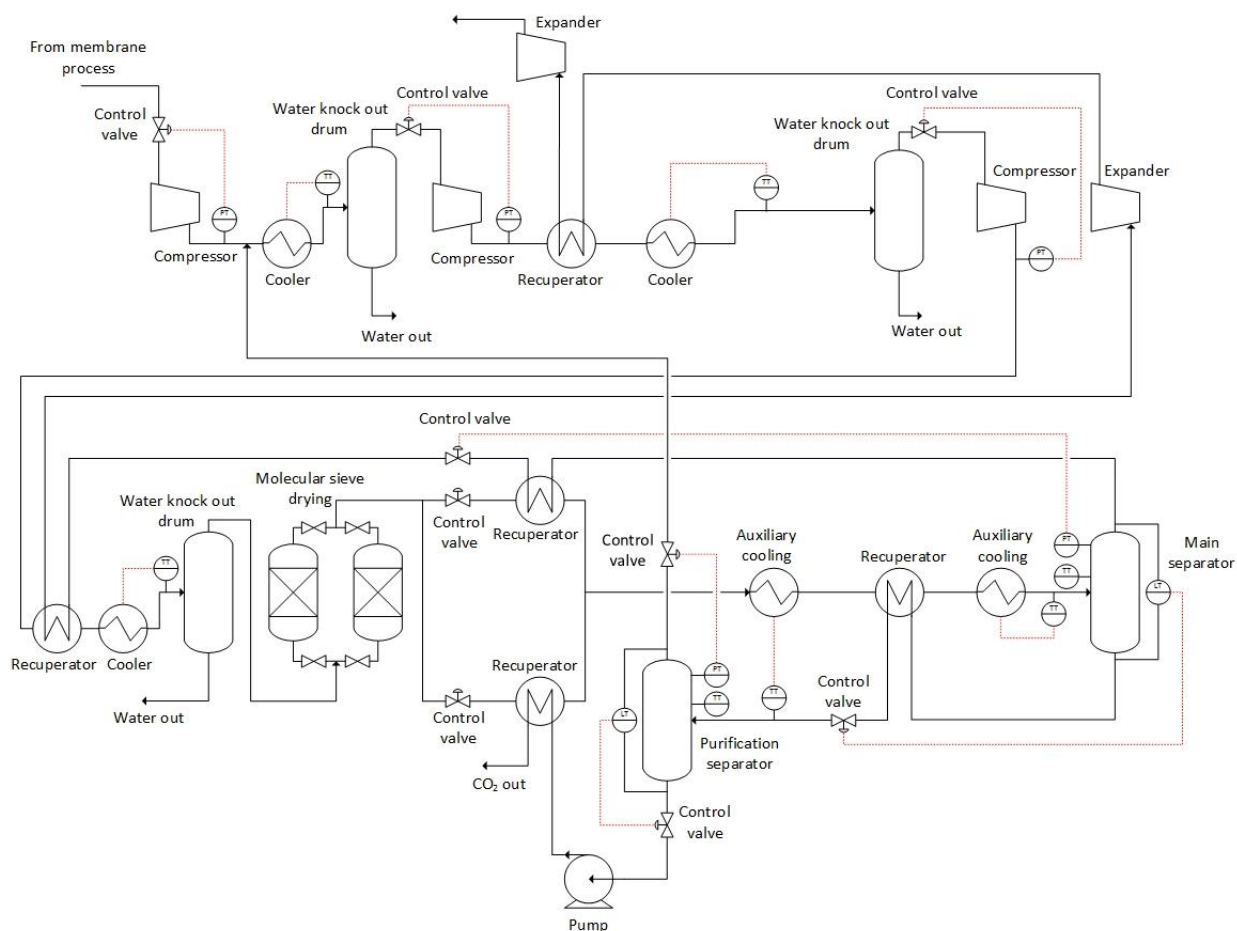


Figure 9.3: Compression and liquefaction unit with control system for pilot plant

10 INTERFERENCE WITH THE CEMENT PRODUCTION PROCESS

The membrane-assisted liquefaction process is an end-of-pipe technology which only requires electric power as energy input and cooling water or alternatively air as thermal energy sink. Modifications of the cement production process is therefore not necessary. The efficiency of the carbon capture process can however be increased considerably by increasing the CO₂ concentration of the incoming flue gas. This can be done by reducing the false air leakage into the cement production process or by enriching the air for the combustion with oxygen. These options should be evaluated for full-scale plants.

The 10 ton-per-day laboratory pilot for CO₂ liquefaction and phase separation unit has been operated with varying feed composition and mass flow rate. The process remained stable, which indicates that this part of the MAL process is well suited to handle fluctuations in the flue gas feed from the cement plant.

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A SUPPLEMENTARY PROCESS DATA

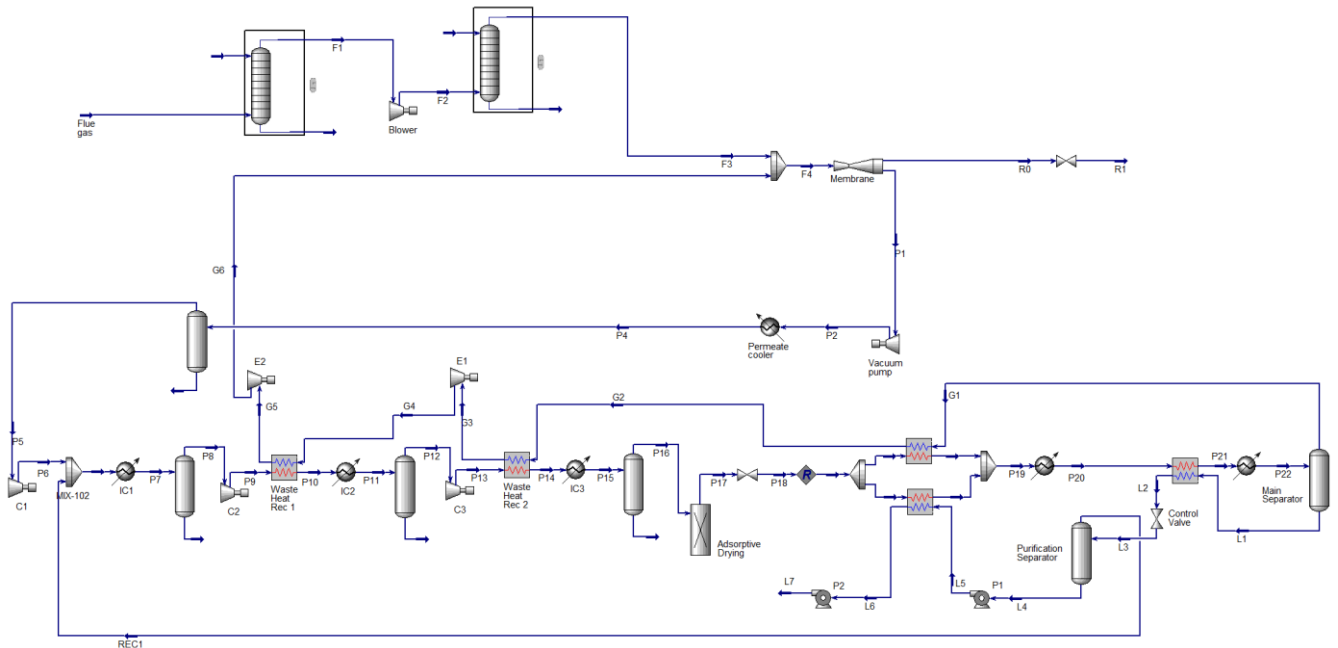


Figure A.1: Simulation PFD

Table A.1: Basic stream data

Stream	Temperature [°C]	Pressure [bara]	Mass Flow [kg/s]
F1	36.4	1.013	7.264
F2	86.5	1.650	7.264
F3	34.3	1.600	7.149
F4	32.9	1.600	7.682
R0	32.8	1.600	5.870
R1	32.8	1.500	5.870
P1	32.8	0.200	1.812
P2	213.2	1.034	1.812
P4	28.0	1.014	1.812
P6	134.8	3.245	1.727
P7	28.0	3.180	1.865
P9	135.3	10.18	1.846
P12	28.0	9.97	1.839
P13	137.1	31.91	1.839
P17	28.0	31.27	1.835
P18	27.6	30.77	1.835
P19	-22.2	30.37	1.835
P20	-39.2	29.97	1.835
P21	-42.9	29.57	1.835

P22	-54.0	29.17	1.835
G1	-54.0	29.17	0.533
G2	4.1	28.59	0.533
G3	112.1	28.02	0.533
G4	12.0	7.195	0.533
G5	110.3	7.051	0.533
G6	4.9	1.600	0.533
L1	-54.0	29.17	1.302
L2	-42.2	28.77	1.302
L3	-55.0	8.481	1.302
L4	-55.0	8.481	1.163
L5	-52.6	60.00	1.163
L6	3.5	59.60	1.163
L7	9.1	110.0	1.163
REC1	-55.0	8.481	0.138

Table A.2: High and low estimates for utilities

		Low estimate	High estimate
Total electric power requirement	kW _{el}	1500	2000
Ambient coolers	kW _{th}	1600	1800
Direct contact cooling duty	kW _{th}	Appr. 2500	