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## ENGINEERING ASPECTS OF THE PLANT DESIGN TO SEPARATE VOLATILE HYDROCARBONS BY VAPOR PERMEATION

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### ABSTRACT

The paper deals with engineering aspects of the design of membrane systems for the separation and recovery of volatile hydrocarbons from off-gases. The membrane which is used for this application is a thin film composite membrane with an elastomeric selective top layer. This membrane has a high permeability for various hydrocarbon vapors and a low permeability for oxygen and nitrogen. The membrane configuration is a flat sheet membrane manufactured to an envelope with a round shape which is installed in the so-called GS module. The energy impact in accordance with the condensation mode and operating pressures is shown. Case studies on the influence of different process parameters, e.g., plant capacity, recovery rate, pressure ratio, stage cut and retentate concentration were carried out on the basis of given feed compressor and vacuum pump capacities. Finally, the investment costs of vapor recovery units in gasoline tank farms are discussed.

### INTRODUCTION

Membrane separation is on the way to becoming an established technique to separate and recover volatile organic hydrocarbon vapors from off-gas streams. The membrane which was developed in the GKSS research center is a thin film composite membrane which consists of an elastomeric permselective coating, a microporous substrate with a given pore size and a given pore distribution and a nonwoven fabric to provide the required mechanical strength. (1) Figure 2 shows the selectivity of the membrane of various hydrocarbon vapors vs. nitrogen. It can be seen that most of the substances have a selectivity hydrocarbon vapor vs. nitrogen of more than 10 which is sufficient to design an economic separations process.

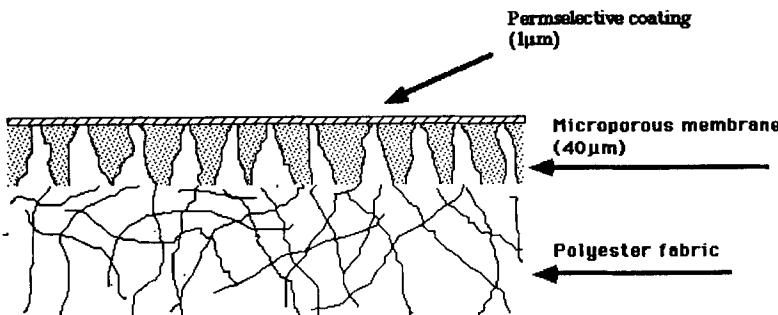


FIGURE 1. Schematic diagram of a composite membrane.

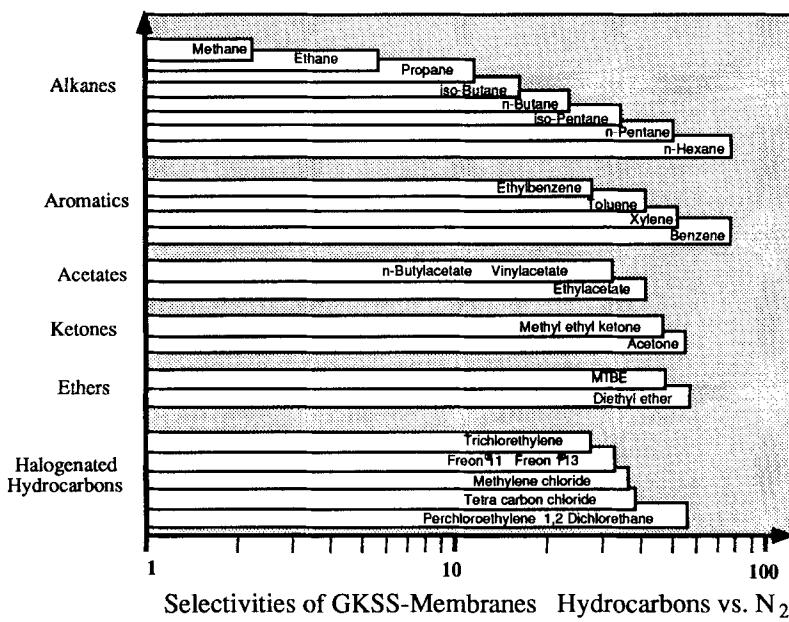


FIGURE 2. Membrane selectivity of various hydrocarbon vapors vs. nitrogen.

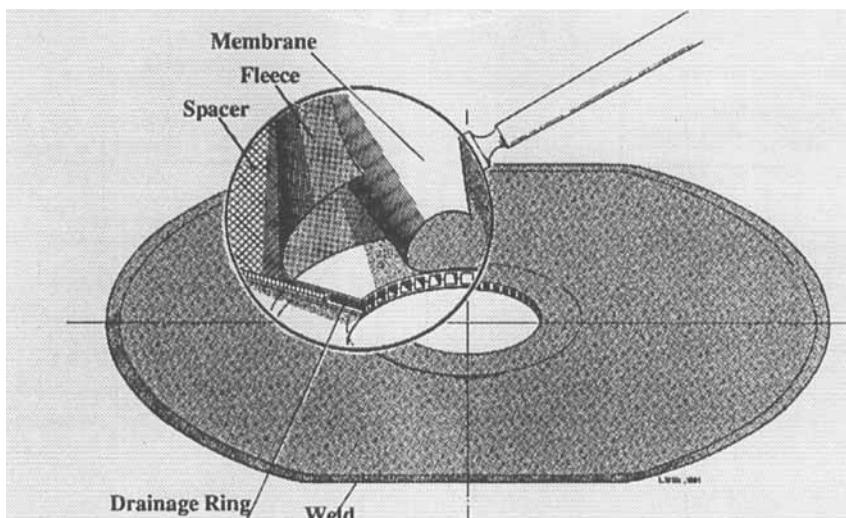


FIGURE 3. Membrane envelope.

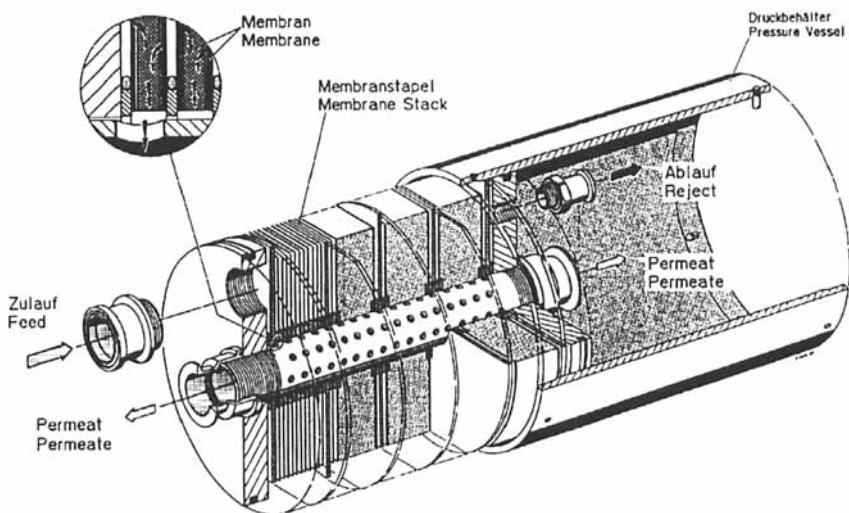


FIGURE 4. GS-Module.

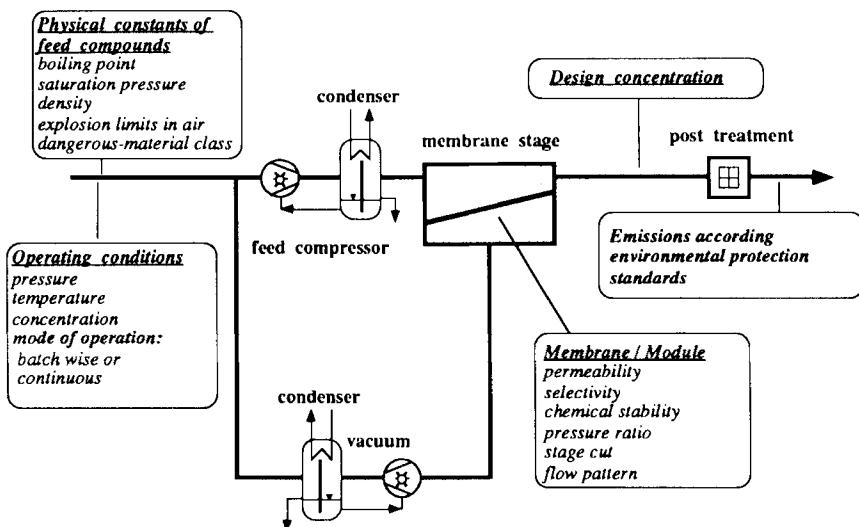


FIGURE 5. Flow scheme of a membrane separation process.

### GS-Module

The GS-Module is a modified plate and frame configuration developed by GKSS engineers. (2) Flat sheet membranes are manufactured into round envelopes of a round shape which are thermally welded at the cutting edges. Nonwoven fleeces and spacer materials are placed between two membranes to provide unrestrained permeate drainage to the central bore of the membrane envelope. The inner materials are fixed by an injection-molded drainage ring which is also used as a support for the O-ring seals of the membrane stack packed in the module.

The membrane/module arrangement is depicted in Figure 4. A standard module is approximately 500 mm long and has a diameter of 320 mm. The installed membrane area of a membrane stack varies from 8 to 10 m<sup>2</sup> and is divided into compartments by means of baffle plates. The feed flow is introduced into the module at the front flange, changes direction at the first baffle plate, flows over the membrane surface and leaves the module at the outlet at the back flange. The feed stream is split into a hydrocarbon enriched permeate stream which penetrates into the membrane envelope and a depleted stream at the upstream side of the membrane. The permeate flow is fed to the central permeate tube (Fig. 4).

### Input Parameters for the Process Design

The layout of a membrane separation process is governed by the physical constants of the feed compounds, the operating conditions, the performance of the membrane/module configuration and the required purity of the vent stream released to the atmosphere (Fig. 5).

TABLE 1. FEED GAS COMPOSITION

0.01 % Methane	0.03 % Ethane
0.69 % Propane	3.69 % i-Butane
6.66 % Butane	4.43 % i-Pentane
2.60 % Pentane	1.51 % Hexane
0.14 % C <sub>7</sub> <sup>+</sup>	0.29 % Benzene
16.71 % Oxygen	63.24 % Nitrogen

TABLE 2. PROCESS DATA

Feed flow		100 m <sup>3</sup> /h
Feed concentration		20 vol% HC
Condensation temperature		30 °C
Feed pressure	Case 1	6.6 bar
	Case 2	6 bar
	Case 3	4 bar
Permeate pressure	Case 1	atmospheric
	Case 2	400 mbar
	Case 3	400 mbar
Pressure difference	Case 1	5.6 bar
	Case 2	5.6 bar
	Case 3	3.6 bar
Pressure ratio	Case 1	6.6
	Case 2	12.5
	Case 3	10
Condensation mode	Case 1	condensation by pressure
	Case 2	condensation by pressure
	Case 3	condensation in lean gasoline
Required retentate concentration		approx. 10 g HC/m <sup>3</sup> air

The boiling point of the feed compounds and their concentration, in addition to the condensation pressure and the condensation temperature, determine the location of the condenser unit. At high hydrocarbon concentrations it is more economical to install the condenser before the membrane stage whereas at low intake concentrations an arrangement where the condenser is situated in the permeate line is more efficient.

#### Comparison of Condensation and Operation Modes

A case study was carried out to compare different condensation and operation modes. The calculations are based on a typical off-gas composition from gasoline tank farms with approximately 20 vol% HC.

Figures 6 and 7 show the dependence of installed membrane area on retentate HC concentration, required power of the total process, recovery rate, and stage cut regarding Case 1.

Case 2 and Case 3 are depicted in Figures 8 to 11.

Case 1

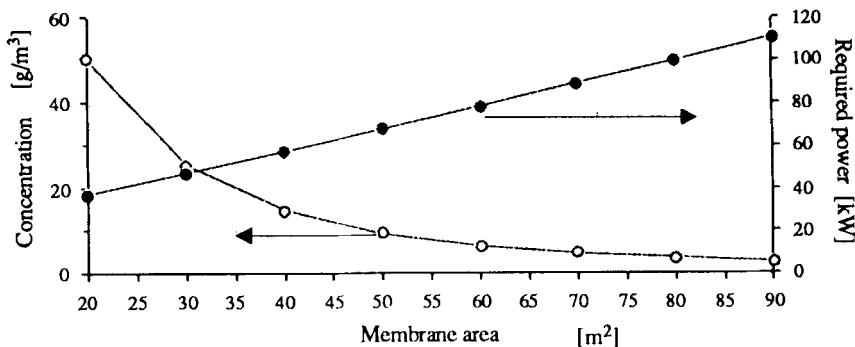


FIGURE 6. Membrane area vs. retentate HC concentration and required power.

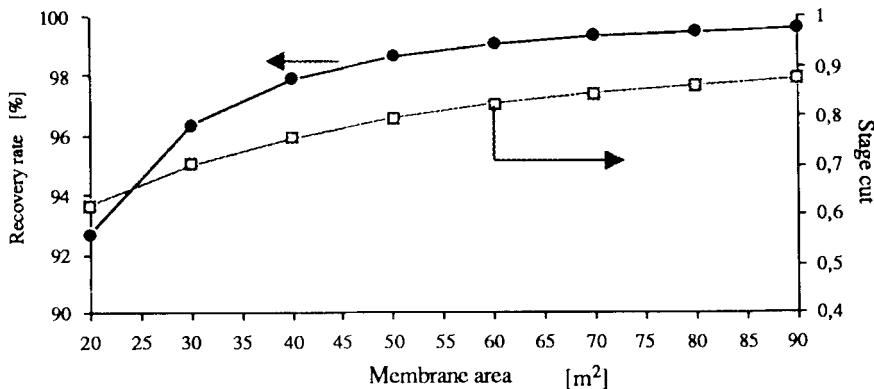


FIGURE 7. Membrane area vs. recovery rate and stage cut.

Case 2

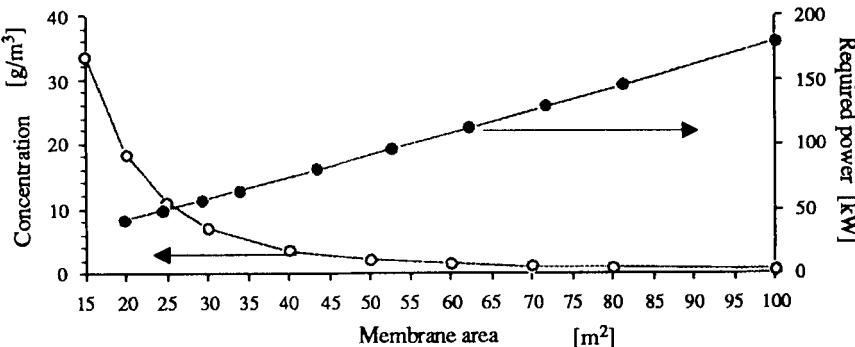


FIGURE 8. Membrane area vs. retentate HC concentration and required power.

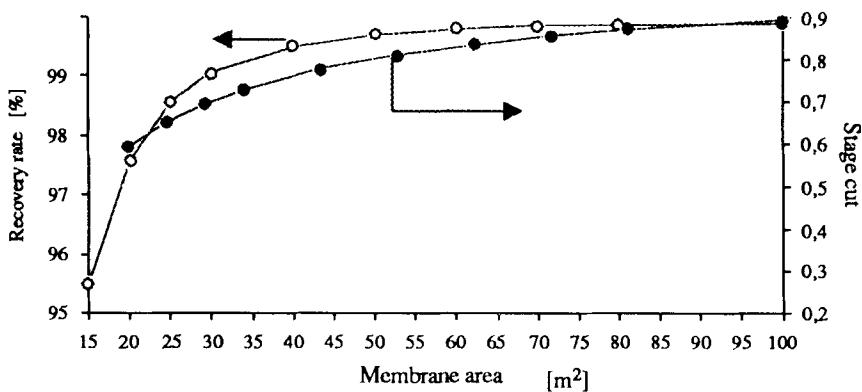


FIGURE 9. Membrane area vs. recovery rate and stage cut.

Case 3

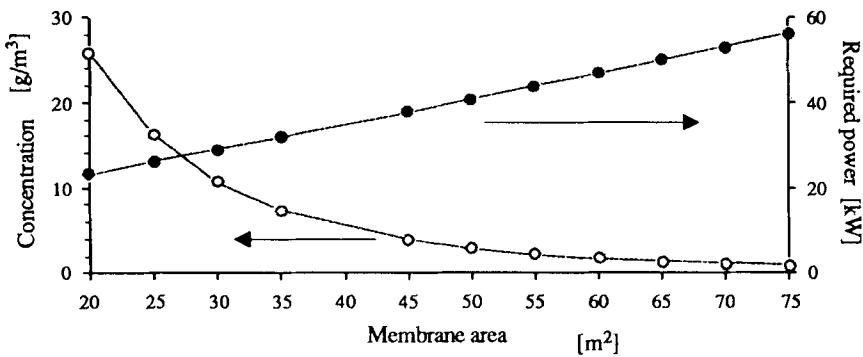


FIGURE 10. Membrane area vs. retentate HC concentration and required power.

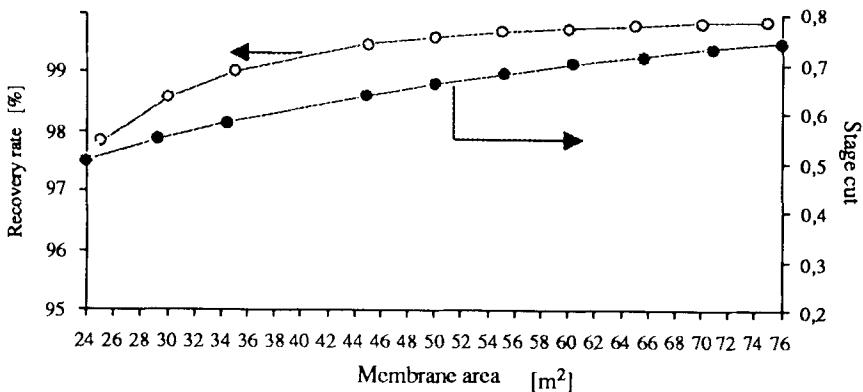


FIGURE 11. Membrane area vs. recovery rate and stage cut.

TABLE 3. CONDENSATION ALTERNATIVES

Case	System	Retentate concentration	Recovery efficiency	Stage cut	Membrane area	Required power
1	Condensation by pressure	9.4 g/m <sup>3</sup>	98.66%	80%	50 m <sup>2</sup>	67.35 kW
2	Condensation by pressure (vacuum mode)	10.92 g/m <sup>3</sup>	98.55%	69%	25 m <sup>2</sup>	56.8 kW
3	Condensation by absorption (vacuum mode)	10.66 g/m <sup>3</sup>	98.58%	54.4%	30 m <sup>2</sup>	28.88 kW

TABLE 4. DATA FOR PROCESS CALCULATIONS

Intake concentration	approx. 20 vol% HC composition, see Table 1
Compressor capacity (liquid ring compressor)	800 m <sup>3</sup> /h
Feed and condensation pressure	4 bar
Condensation temperature	30 °C
Mode of condensation	condensation in lean gasoline
Vacuum pump capacity (rotary vane pump)	1900 m <sup>3</sup> /h
Vent gas concentration (single stage unit)	35 g HC/m <sup>3</sup> air = 95 % recovery
Vent gas concentration (hybrid system)	150 mg HC/m <sup>3</sup> air organic compounds 5 mg benzene/m <sup>3</sup> air outlet concentration of the first stage 10 g HC/m <sup>3</sup> air

The results of the calculations are summarized in Table 3.

It can be concluded that absorption in lean gasoline and operation in vacuum mode is the most economic process.

Case 2 shows a slight decrease in membrane area because of the higher feed pressure but the energy demand is twice that of Case 3. Case 1 is the last choice because of the high energy demand and the large membrane area. This is caused by the disadvantageous pressure ratio which has a direct influence on membrane areas and stage cut at given retentate concentrations. (3)

#### Limitations at Given Compressor and Vacuum Pump Capacities

Pump suppliers offer their set of machines with a defined suction capacity. The calculation of the suction capacity is based on the following conditions: 20 °C dry air according suction pressure of vacuum pumps or 20 °C dry air at atmospheric pressure (1013 mbar) dealing with compressors. These units are available under certain gradings. The sizing of a vapor recovery unit (VRU) depends on the various layout data e.g., volume off-gases to be treated, required vent

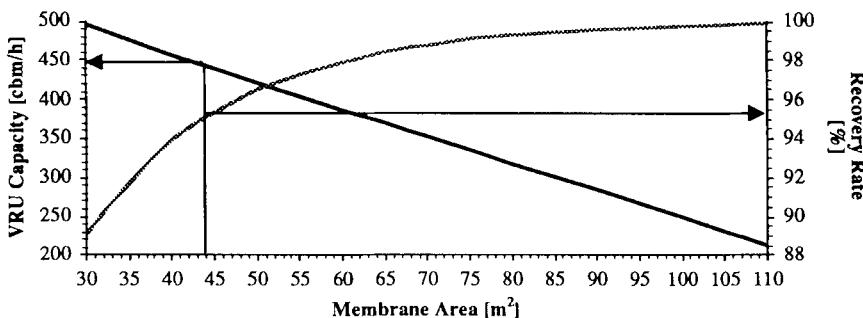


FIGURE 12. Membrane area vs. VRU capacity and recovery rate.

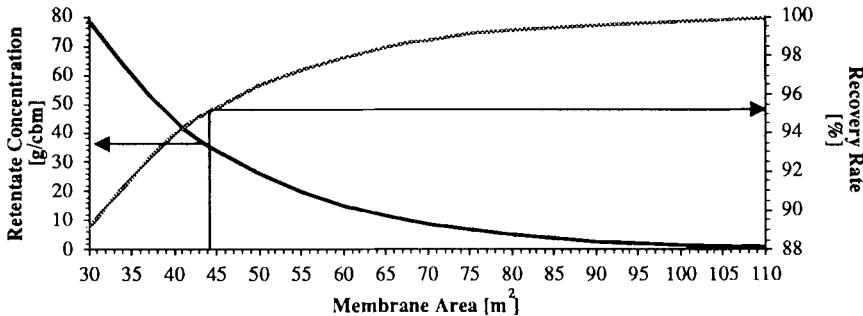


FIGURE 13. Membrane area vs. retentate concentration and recovery rate.

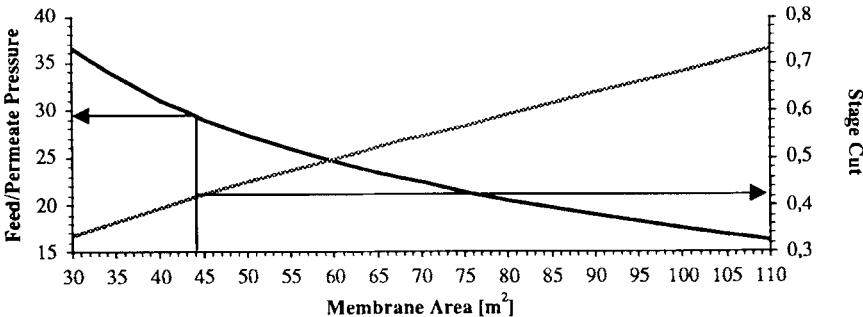


FIGURE 14. Membrane area vs. pressure ratio and stage cut.

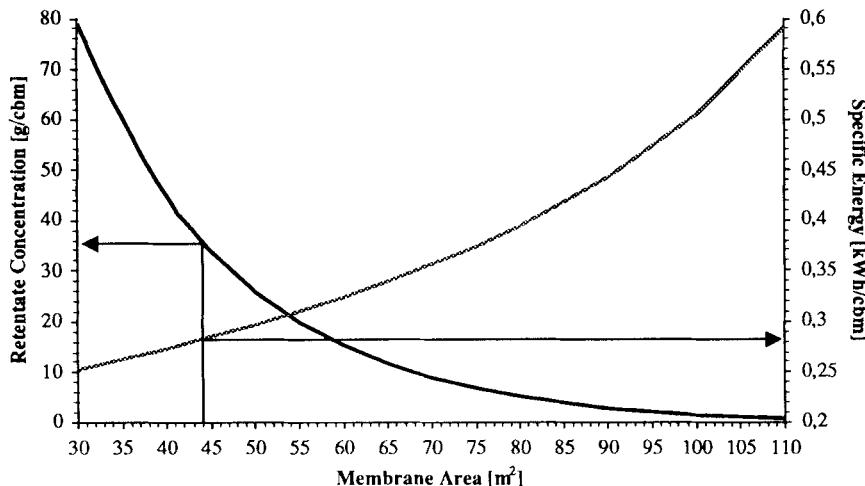


FIGURE 15. Membrane area vs. retentate concentration and specific energy.

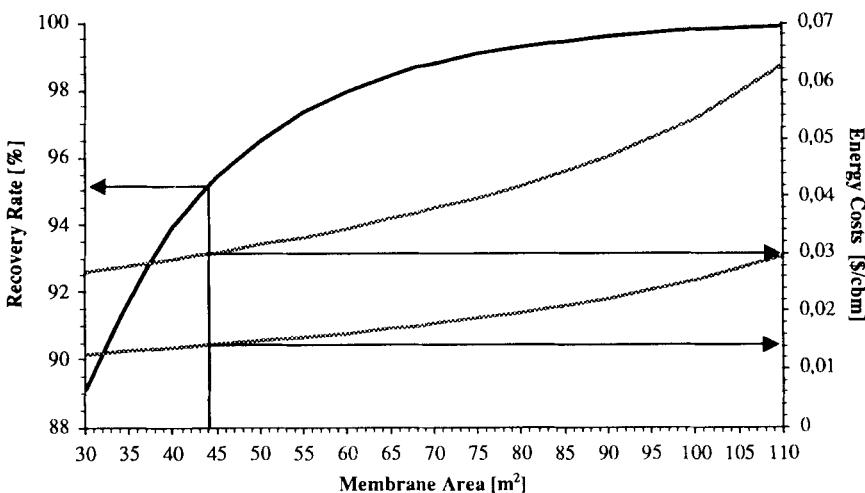


FIGURE 16. Membrane area vs. recovery rate and energy costs.

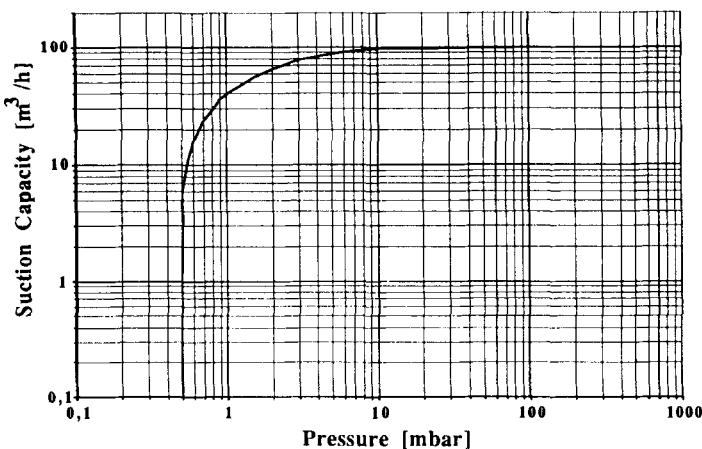


FIGURE 17. Typical characteristic of a rotary vane pump.

stream purity, and the process parameters which are discussed earlier under "Input Parameters for the Process Design". The following graphs show the influence of layout requirements on the process design at given pump capacities which result in inevitable limits.

Figures 12 - 16 show the dependence of VRU capacity, recovery rate, retentate concentration, stage cut, pressure ratio, specific energy consumption, and energy costs on installed membrane area.

The recovery rate [%] is a dependent quantity and is governed by the average intake concentration. The total retentate concentration [g HC/m<sup>3</sup> air] is determined by the permeability of the feed compounds through the membrane and their absorption behavior in lean gasoline.

The stage cut is the relation of permeate volume flow to feed volume flow introduced in the membrane stage. The pressure ratio is defined as quotient of feed pressure to permeate pressure. The nonlinear slope down the curve, membrane area vs. stage cut, is caused by the characteristic of the rotary vane pump which is shown as a typical example in Figure 17.

The specific energy [kWh/m<sup>3</sup>] summarizes the energy consumption of the feed compressor and the vacuum pump. The energy costs are based on a price of 0.05 \$/kWh in the USA and 0.106 \$/kWh in Germany.

Because of the fixed suction capacity of the feed compressor, the VRU capacity changes (volume of off-gas from the plant to be treated) in dependence on the required retentate concentration. The increase in membrane area causes a decrease of the retentate concentration. This is associated with a rise of the stage cut which has a linear influence on the reduction of the VRU

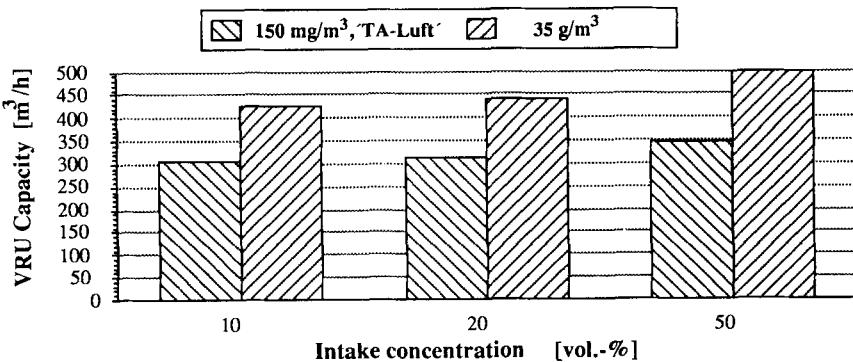


FIGURE 18. Intake concentration vs. VRU capacity.

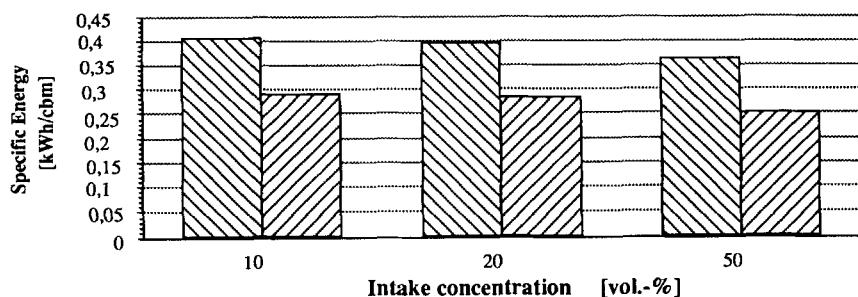


FIGURE 19. Intake concentration vs. specific energy.

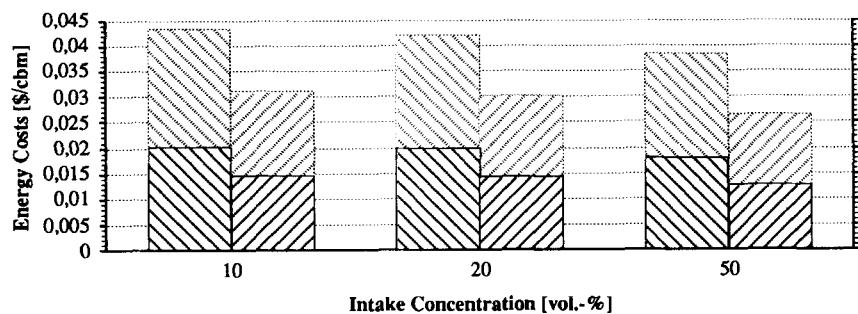


FIGURE 20. Intake concentration vs. energy costs.

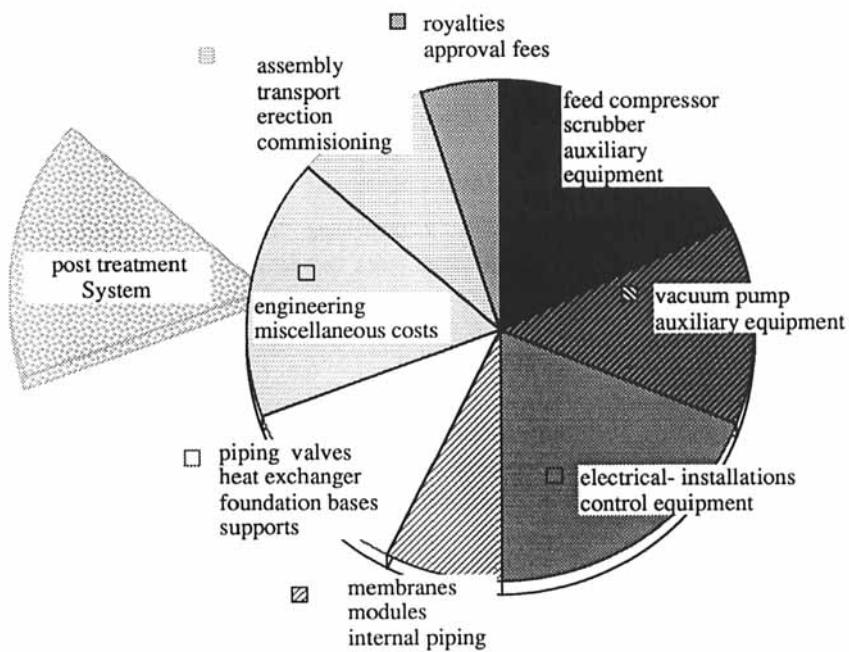


FIGURE 21. Cost distribution of a vapor recovery unit.

capacity. Consequently the specific energy demand and the energy costs increase with the decrease of hydrocarbon content of the retentate stream. The influence of the hydrocarbon concentration of the off-gas versus VRU capacity, specific energy and energy costs in dependence on vent gas purity are depicted as block diagrams in Figures 18 to 20.

It has been shown that the volume to be treated can be increased at higher intake concentrations. Because of the decrease in volume which is absorbed and condensed in the condensation/absorption column, the recycled permeate flow is also reduced. Consequently, the specific energy demand and the energy costs decrease with an increase of HC content of the off-gas.

### Conclusions

Six VRU's based on membrane technology with a capacity of 300 to 1500 m<sup>3</sup>/h have been installed in Germany or the Netherlands. The suppliers of these units are optimistic that they will get more orders in the near future. Figure 21 shows the cost distribution of the total system. Feed compressors and vacuum pumps take over approximately  $1/3$  of the total costs. Electrical installations and control equipment share also a big part of the costs because of the stringent safety stan-

dards for the operation of such units in an explosive environment. Engineering and miscellaneous costs have a cost reduction potential with an increasing number of installed units. If the environmental protection standards require extremely high vent gas purities, a post-treatment system is necessary. This adds an additional 15 % to investment costs.

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