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Thermodynamic and economic evaluation of a CO_2 membrane separation unit integrated into a supercritical coal-fired heat and power plant

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Abstract

This paper presents the results of thermodynamic and economic analysis of a coal-fired combined heat and power plant (CHP) working at supercritical parameters, integrated with a carbon dioxide capture unit based on membrane technologies. Two membrane system configurations are described, compared and optimized. Both consist of a two-stage membrane unit, but in the first variant (Case 1) no recirculation is performed while in the second one (Case 2), retentate from behind the second membrane is recirculated before the first membrane. The economic analysis includes a comparison of the systems with a unit working without CO_2 capture (reference unit). The main thermodynamic (annual generation of the products, efficiencies) and economic (break-even price of electricity, break-even price of membranes) indices are presented in this paper. The results show that the profitability of the investment in CHP units integrated with CO_2 capture is strongly dependent on the annual operation time and price of emission allowances. Better thermodynamic and economic characteristics are obtained for the system with retentate recirculation than for the system without recirculation.

Keywords: Membrane CO_2 separation, Combined heat and power plant, Thermodynamic and economic analysis

1. Introduction

The energy policy of the European Union is becoming increasingly restrictive with regard to carbon dioxide emissions. The latest documents include a declaration of intent to reduce the emissions of greenhouse gases by as much as 90% by the year 2050 [1, 2]. In order to fulfill the obligations of the European Union it is needed to increase the efficiency of generation (also by using combined heat and power production) and to introduce new energy production technologies. In view of the important share of fossil fuels in the energy generation mix, it is insufficient to merely increase the share of renewable energy sources in the structure of primary energy consumption. There is a need to develop and implement technologies that use fossil fuels but do not cause emissions. One such solution is a group of Clean Coal Technologies. An indispensible element of such units is their integration with carbon dioxide capture technologies, which can be realized in three main groups: post-combustion, pre-combustion or oxy-fuel combustion [3–8].

These technologies embrace several methods of carbon dioxide capture from flue gases (or process gas in the case of pre-combustion technology). Among the most frequently used there are absorption, adsorption, cryogenic and membrane methods, but also, being in the early stage of development, thermoacoustic methods [6, 7, 9–13]. These solutions find application in different types of power systems. The most technologically mature are methods based on chemical absorption, but they need a large amount of heat for conduction of the desorption process of carbon dioxide from the sorbent solution.

The special nature of operation of a combined heat and power plant in temperate climate zones means that the extraction of steam for the desorption process can signifi-

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cantly restrict the possibility of heat production for useful purposes. Thus, an interesting way ahead might be to use an external source of heat for steam generation to power the capture process, or to use methods that do not require heat to be supplied to the process. Especially attractive seem to be membrane methods, which have been intensively developed in recent years. The merit of using membranes for gas separation consists in the selective permeation of a chosen component through the membrane material. The effectiveness of the process depends on the membrane material and on the partial pressure difference of the separated gas on both sides of the membrane. This difference is caused by compressors and/or vacuum pumps driven by electricity. The results of experimental and computational research show, as presented e.g. in [14], that this technology can be significantly less energy intensive than other methods of separation.

The main aim of this paper was to evaluate the possibility of integrating a carbon dioxide membrane separation unit with a supercritical coal-fired combined heat and power plant. Analysis and choice of the structure of the separation unit, as well as the assessment of the impact of integration on thermodynamic and economic indices of the whole CHP plant was performed to achieve the desired goal.

2. Methodology of calculations

A membrane is a phase separator, allowing for selective flow of chosen components of the mixture between the boundary of two liquid or gaseous phases. There are many types of membranes applied in the separation process, but the most commonly used are inorganic polymer membranes [14, 15]. Their big advantage is the ability to produce them in the form of hollow fibers with a large surface area, which significantly reduces their size and thus the cost of production. Inorganic membranes are also often characterized by resistance to high temperature, resistance to the presence of water, a fixed, stable pore structure and chemical inertness.

The most important parameters of the membranes from the point of view of their suitability for the gas separation process are: permeability (stream transported through the membrane per unit of pressure and thickness of the membrane) and selectivity (ratio of permeabilities of the individual components of the mixture flowing through the membrane). The properties of the membranes have been widely discussed, inter alia, in [7, 10, 12, 13].

A system for gas separation using membranes in the simplest case consists of one module. In such a system

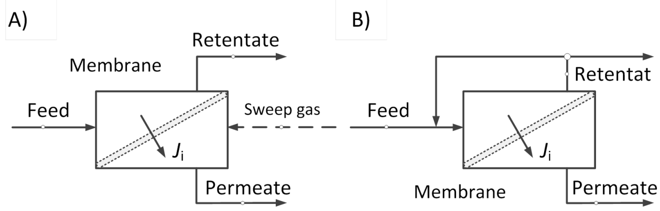

Figure 1: Configurations of a single-stage membrane system; A—countercurrent cross flow; B—system with retentate recirculation

the feed stream is supplied to the membrane, where it is separated into two streams—permeate (part of stream permeating through the membrane) and retentate (part of a stream which remains before the membrane). A diagram of this system is shown in Fig. 1A. The flow of gas is caused mostly by a fan (compressor) installed upstream of the membrane module (on the feed stream). The partial pressure difference across the membrane, which is the driving force of the separation process, is caused by a vacuum pump (optionally, the compressor installed in place of the fan) and the difference in the molar shares of the same component on both sides of the membrane. The flow of the different components through the membrane may proceed in various configurations, including in particular downstream or upstream (Fig. 1A), with the use of a sweep gas (Fig. 1A, dashed line) and with recycling of a part of the retentate stream before the membrane (Fig. 1B) .

Single-stage installations can be combined into larger systems consisting of two or more membrane modules in different configurations. In order to improve the parameters of the obtained gas, recirculation of a stream—typically of the retentate—is also sometimes applied.

The model of a membrane used in the analysis was built in the Aspen Custom Modeler and then imported into Aspen Plus. In the calculations a real gas model called Peng–Robinson [16] was applied. In the modeling process, certain assumptions were made. It was assumed, among others, that the permeability of the membrane is independent of the pressure and concentration of gas components, the pressure loss during a flow through the membrane is negligible, and that no mass transfer resistance occurs at either side of the membrane. A detailed description of the model can be found in [7, 12].

2.1. Evaluation indices of the separation process

Evaluation of the quality of the process of gas separation using membrane methods can be made with the use of two indicators: purity and recovery rate. Purity is the mole fraction of carbon dioxide in the permeate stream (and therefore the stream directed to further compression and transport to the place of storage). The recovery rate (R_i) determines which part of the stream of carbon dioxide from flue gas permeated to the permeate stream and is defined by the relationship:

$$R_\mathrm{i} = \frac{\dot{n}_\mathrm{i,P}}{\dot{n}_\mathrm{i,F}} = \frac{\dot{n}_\mathrm{P}(Y_\mathrm{i})}{\dot{n}_\mathrm{F}(X_\mathrm{i})} \tag{1}$$

where $\dot{n}_\mathrm{i,P}$, $\dot{n}_\mathrm{i,F}$ is the molar stream of i-component in the permeate and feed stream, respectively, kmol/s; $\dot{n}_\mathrm{P}/\dot{n}_\mathrm{F}$is the molar stream of permeate/feed, kmol/s; Y_i/X_i is the molar share of i-component in the permeate/feed, respectively.

Purity and recovery rate can be used to assess the quality of the separation process and to indicate whether the resulting stream parameters are consistent with guidelines, but say nothing about the energy inputs that must be incurred in the process of separation. The energy intensity index E_{CO2} gives information of this type, defining the electrical power needed for the process of carbon dioxide capture, and is calculated according to the formula:

$$E_\mathrm{CO2} = \frac{N_\mathrm{el}}{\dot{m}_\mathrm{CO2,P}} \tag{2}$$

where N_{el} is the electric power needed for the process, kW; $\dot{m}_\mathrm{CO2,P}$ is the mass stream of CO_2 in the permeate stream, kg/s.

The energy intensity index is useful when comparing different methods of carbon dioxide separation, but for a full evaluation of the separation process all the evaluation indices must be determined (including purity and the recovery rate).

2.2. Characteristics of the analyzed membrane systems

As shown in the analysis presented inter alia, in [17], a system with one membrane section usually fails to obtain sufficient purity of the separated carbon dioxide. Therefore, this paper analyzes a cascade system, consisting of two membrane modules in two configurations—without recirculation of retentate (Fig. 2) (Case 1) and with recirculation of retentate (Fig. 3) (Case 2).

In both variants, after each membrane module with the same parameters (shown in Table 1 [18]), there is a two-section vacuum pump (VP) with intersection cooling of the gas in heat exchangers (HE) to a temperature of 30°C. In Case 2 retentate stream is recycled from behind the second membrane before the first membrane, where it is mixed with the feed stream. This choice resulted from preliminary calculations, which showed that recirculation of the retentate after the second and not the first membrane achieves better effects, because the stream is characterized by a higher molar share of carbon dioxide. In the calculations the energy consumption required for recirculation of the retentate was not taken into account. The nominal parameters of the CO_2 membrane separation unit assumed for the analysis are presented in Table 1.

Table 1: Nominal parameters of the CO_2 membrane separation unit

Quantity	Value
Feed temperature, °C	30
Feed stream, kg/s	100
Feed pressure, bar	1.0
Permeate pressure, bar	0.03..1.00
CO_2 permeability, $m^3/(m \cdot h \cdot bar)$	3.3825
N_2 permeability, $m^3/(m \cdot h \cdot bar)$	0.072
H_2O permeability, $m^3/(m \cdot h \cdot bar)$	0
O_2 permeability, $m^3/(m \cdot h \cdot bar)$	0.3
Membrane thickness, μm	60
Ideal selectivity coefficient	47

2.3. Characterization of the analyzed CHP plant

The power plant was assumed to have gross power of 320 MW (during condensation operation) and to work for the needs of the district heating network with a maximum heat demand of 500 MW. The plant consists of a supercritical coal boiler (SCB) powered by hard coal (with the following composition: c = 0.599, h = 0.038, o = 0.050, n = 0.012, p = 0.010, w = 0.090, ash = 0.200), an extracting-condensing steam turbine (with a high—(H), intermediate—(I) and low pressure (L) part), a condenser (CND), four low-pressure regenerative heat exchangers (LR) and three high-pressure (HR), a steam cooler (SC) and a deaerator (DEA). A schematic diagram of the CHP unit integrated with the gas separation membrane module is shown in Fig. 4.

It was assumed that the plant includes a pulverized coal boiler, operating at 94.5% efficiency. The boiler produces live steam with parameters of 653°C/30.3 MPa (at the turbine inlet 650°C/30.0 MPa) and reheated steam with parameters of 672°C/6.0 MPa (at the turbine inlet 670°C/5.9 MPa). As a result of the combustion processes, flue gas with the following molar composition is produced: (CO_2) = 0.1418; (H_2O) = 0.0775; (N_2) = 0.7381; ($Ar+O_2$) = 0.0417; (SO_2) = 0.0009.

In the first stage of the analysis operational indicators of the power plant were determined, including both instantaneous and annual average values. Detailed data concerning the assumptions and results of analyses of the operation of the system are presented, among others, in [12, 19, 20].

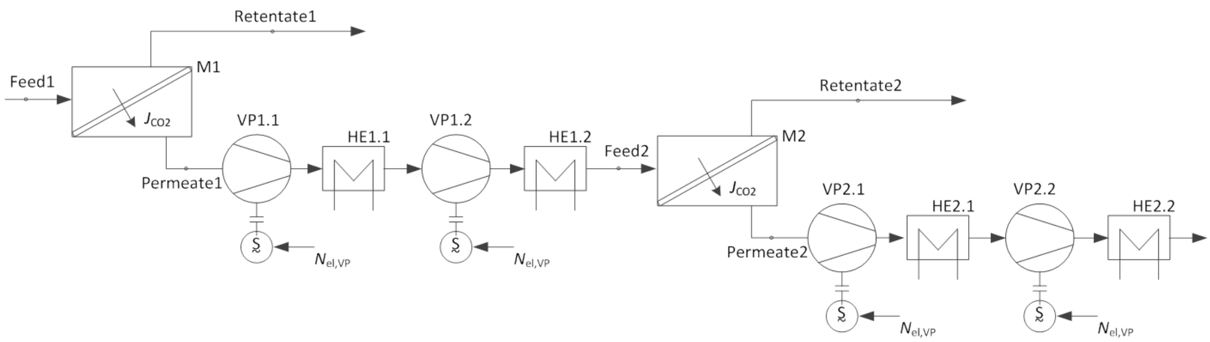

Figure 2: Schematic diagram of a two-stage membrane system with a two-section vacuum pump

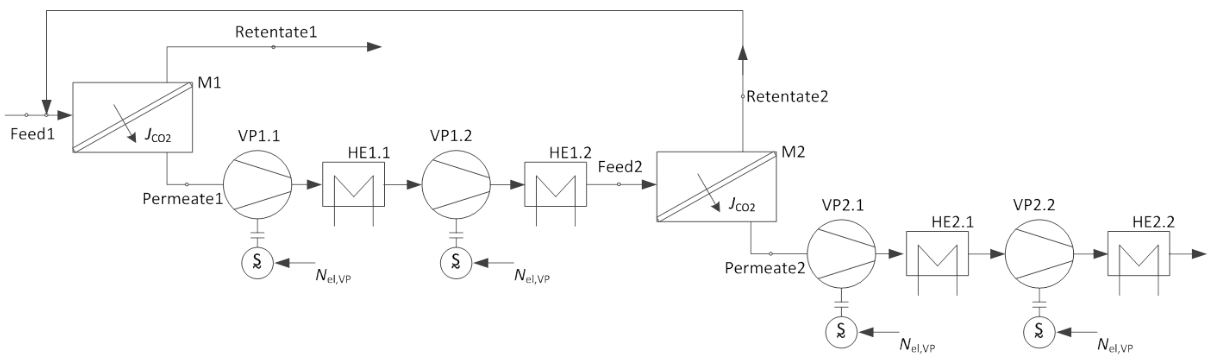

Figure 3: Schematic diagram of a two-stage membrane system with retentate recirculation and a two-section vacuum pump

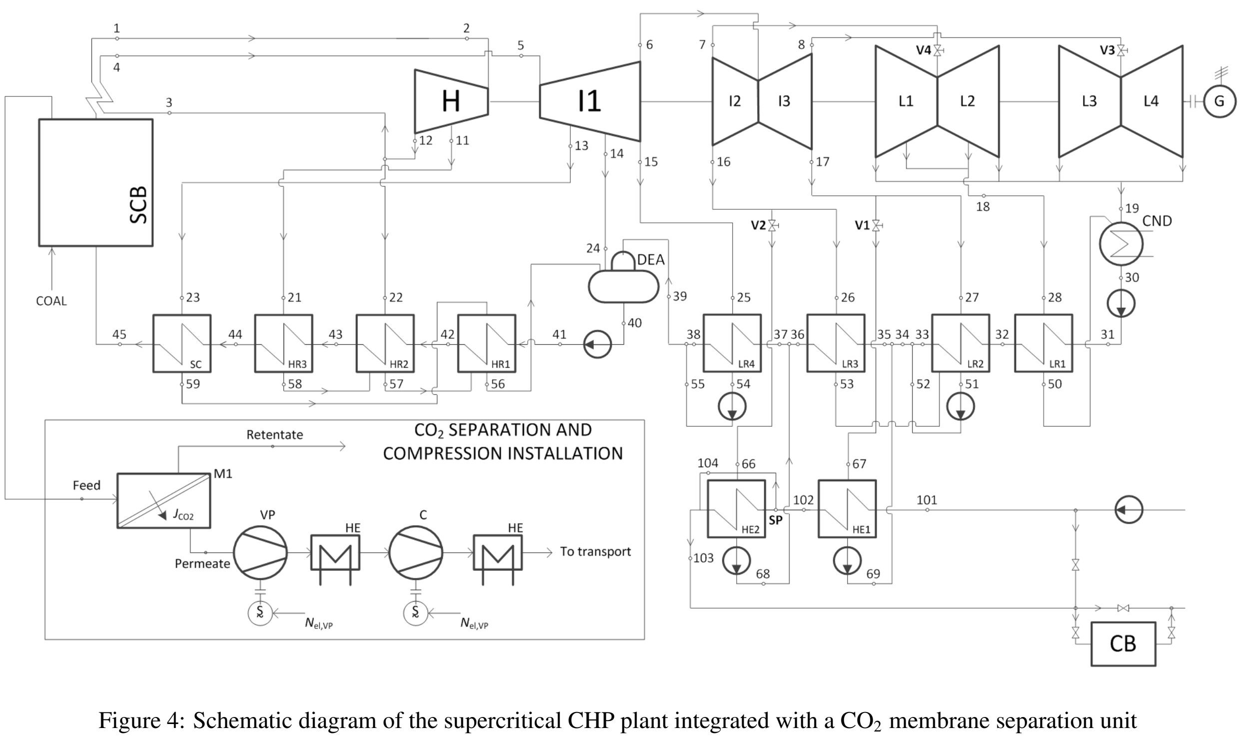

Figure 4: Schematic diagram of the supercritical CHP plant integrated with a CO_2 membrane separation unit

Table 2: Key assumptions for economic analyses

Quantity	Value
Total investment cost , M PLN	2 090
Nominal annual operations time, h/a	8000
Share of internal funds/commercial loan, %	20/80
Real interest on loan, %	6
Loan repayment time, years	10
Operation time, years	20
Discount rate, %	6.2
Average amortization rate, %	6.67
Unit operating and maintenance costs, PLN/MWh	25.0
Price of heat, PLN/GJ	33.9
Coal price, PLN/GJ	15.3
Nominal CO_2 emission allowance price, PLN/Mg	91.5
Liquidation value related to the investment, %	20
Exchange rate, PLN/€	4.185

2.4. Assumptions for economic analysis

Economic analysis of the two variants is based on the method of determining the break-even point, here in the form of the break-even price of electricity c_{el}^{gr}. This quantity is expressed in monetary units, related to 1 MWh of electricity produced, which in the defined time of operation will provide a return of investment, and therefore the condition of zeroing of the net present value (NPV = 0) will be fulfilled. Detailed methodology for determining the NPV value and break-even price of electricity was presented, among others, in [12, 13].

Economic calculations were carried out using the authors' own computational algorithms, built in the Excel environment. For particular variants of the systems (with and without CO_2 capture) the same assumptions about fuel prices, interest rates and the time of repayment of loans, operating periods, etc, were made. The methodology for evaluating the investment in CHP system is presented in detail in [12]. It was decided not to diversify the size of the investment on different systems (with and without capture), but instead to determine the break-even (maximum) price of 1 m^2 of the membrane, which would allow for a return on investment in relation to the variant without capture (but in light of the existence of the emissions trading scheme). This is due to the fact that, given the still insufficient maturity of the membranes, their purchase cost in the literature covers a very wide range (from several to several hundred US$, in e.g. [21–23]), thus, it is difficult to adopt reliable values. Key assumptions for the economic analysis are summarized in Table 2.

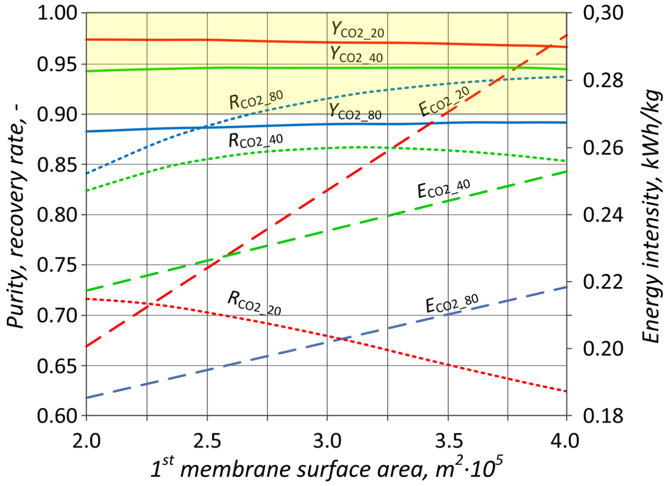

Figure 5: Purity, recovery rate and energy intensity of the separation process as a function of first membrane surface area for three values of the second membrane surface area for the system without recirculation

3. Results of analyses

In the first stage of work a carbon dioxide separation system as a stand-alone installation was analyzed. This was primarily aimed at comparing the two considered concepts, analyzing the impact of various quantities on the evaluation indices and at optimizing the systems. As part of further analyses, calculations relating to the system integrated with a supercritical coal-fired CHP unit were performed which included thermodynamic and economic analyses.

3.1. Analysis of the CO_2 membrane separation unit

The first step was to analyze the influence of the selected parameters on evaluation indices of a membrane separation system, i.e., purity, recovery rate and energy intensity. This analysis was carried out for the system (i) without and (ii) with retentate recirculation (as shown in Fig. 2 and 3, respectively). However, as the characteristics are very similar, only one case is presented here. Exemplary results of the evaluation are shown in Fig. 5. The yellow background marks the area in which purity and the recovery rate are 0.9 and above. It was assumed that the surface of the first membrane varies in the range 200 000..400 000 m^2, while the surface of the second module is defined at three levels: 20 000 m^2, 40 000 m^2 and 80 000 m^2. In the case of the system with recirculation (not shown here), it is possible to obtain a slightly higher purity of permeate at a lower power consumption. For a given surface of the first membrane, better evaluation indices can be achieved by increasing the surface area of the second membrane.

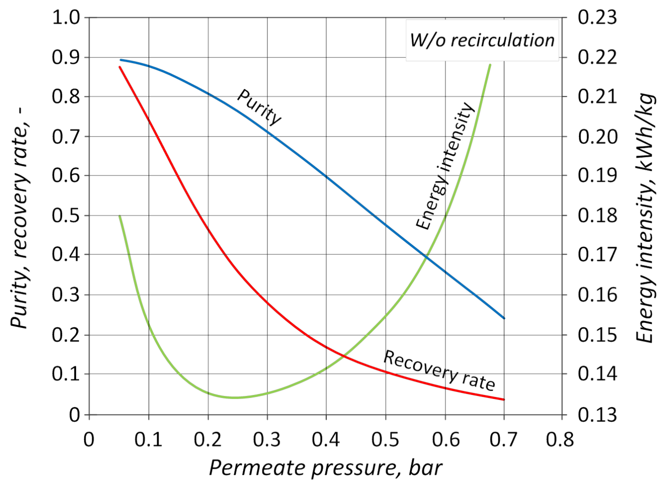

Figure 6: Purity, recovery rate and energy intensity of the separation process as a function of permeate pressure after the first membrane for Case 1

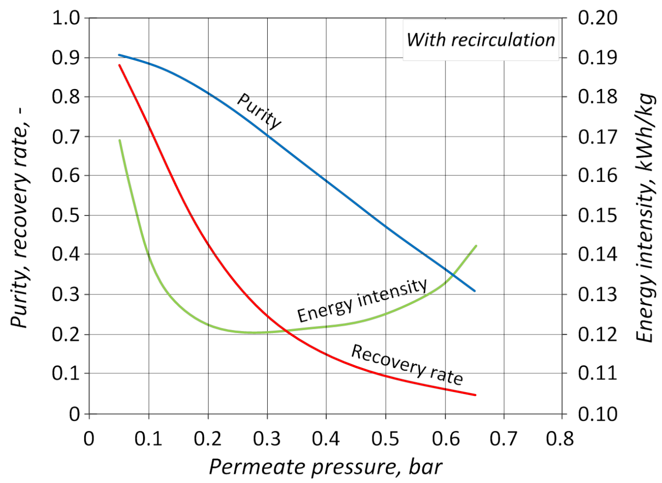

Figure 7: Purity, recovery rate and energy intensity of the separation process as a function of permeate pressure after the first membrane for Case 2

Then the influence of pressure on the membrane separation process evaluation indicators was analyzed. The results of this analysis are shown in Figs 6 and 7. It is assumed that the surface area of the first membrane is 350 000 m^2, while the surface area of the second membrane is 80 000 m^2. The analysis shows that there is a certain minimum energy consumption, which in the presented cases lies in the range in which both purity and the recovery rate are below the established minimum values.

3.2. Optimizing the selection of membranes for the CHP plant

The next stage of the work involved optimization calculations. The assumption made was that the main objective is to minimize the energy consumption of the separation process, while maintaining the minimum requirements of 0.9 for purity and the recovery rate. In the system with two membranes there are multiple decision variables affecting the final result of separation. The analysis assumed decision variables of: (i) permeate pressure after the first and second membrane, and (ii) surface area of the first and second membrane. Table 3 summarizes the most important results of the calculations for two-stage optimization with and without recirculation of the retentate from behind the second membrane.

The optimization calculations provided key evaluation indices, including in particular the energy consumption of the capture and compression process, which were used to determine the basic thermodynamic quantities characterizing the analyzed plant. It was necessary to determine annual amounts of fuel burned, the amount of electricity and heat sold, and, consequently, the average annual gross and net efficiency as well as overall efficiency. These quantities, calculated for different operating times of the year (in Table 4: 5000 h, 6500 h and 8000 h), were used as input data for the economic analysis.

3.3. Results of the economic analyses of the considered variants of CHP plants

The results of optimization calculations, thermodynamic analysis and economic assumptions were used to determine the economic evaluation indices. In view of the current EU Emissions Trading System, in the analyses presented in this paper, the cost of purchasing carbon dioxide emission allowances (C_{UE}) is the main quantity that was subjected to variation. Moreover, due to uncertainties related to the unit cost of the purchase and operation of the membranes, it was assumed that investments in the units are the same regardless of the option, but varied the net power and efficiency of the system.

The profitability of carbon dioxide capture systems in relation to the construction of the reference systems (without capture) is largely affected by the costs associated with the charges for carbon dioxide emission allowances. Making assumptions regarding prices of emission allowances introduces a high degree of uncertainty. The calculations used the nominal value of 91.5 PLN/Mg, which is the expected average price from the price path forecast in [23] for investments beginning in 2012. However, in view of the low emissions energy policy and low correlation in recent years between forecasts and actual values, the predicted value is likely to be wrong. Accordingly, the influence of the prices of carbon dioxide emission allowances

Table 3: Results of optimization of a two-stage system without and with recirculation

Quantity	Case 1	Case 2
Feed stream, kg/s	288.45	
CO_2 stream in the feed, kg/s	60.60	
CO_2 stream after two stages of separation, kg/s	54.54	
CO_2 purity after first membrane module	0.6315	0.6416
CO_2 recovery rate after first membrane module	0.9209	0.9683
First membrane surface area, m^2	854175	829845
Pressure after first membrane, bar	0.0310	0.0310
Second membrane surface area, m^2	829845	250986
Pressure after second membrane, bar	0.1271	0.247
CO_2 purity	0.9000	0.9000
CO_2 recovery rate	0.9000	0.9000
Energy intensity of the separation process, kWh/kg	0.1930	0.1797
Energy intensity of the compression process, kWh/kg	0.1343	0.1350
Energy intensity of separation and compression, kWh/kg	0.3273	0.3147

Table 4: Annual quantities of the products obtained and annual fuel consumption for variants without CO_2 capture (reference) and with CO_2 capture (Case 1 and Case 2) for different annual operations time

Quantity	Reference case			Case 1			Case 2		
	5000 h	6500 h	8000 h	5000 h	6500 h	8000 h	5000 h	6500 h	8000 h
Gross production of electricity, GWh	1445.6	1919.51	2392.9	1445.6	1919.5	2392.9	1445.6	1919.5	2392.9
Net production of electricity, GWh	1337.2	1778.47	2219.4	1016.3	1361.1	1706.0	1028.2	1376.8	1725.0
Useful heat production, GWh	1276.7	1351.7	1426.7	1276.7	1351.7	1426.7	1276.7	1326.7	1426.7
Use of chemical energy of fuel, GWh	3301.5	4292.27	5282.6	3301.5	4292.27	5282.6	3301.5	4292.27	5282.6
Annual average gross efficiency of electricity generation	0.4379	0.4472	0.4530	0.4379	0.4472	0.4530	0.4379	0.4472	0.4530
Annual average net efficiency of electricity generation	0.4050	0.4143	0.4201	0.3078	0.3171	0.3230	0.3111	0.3208	0.3266

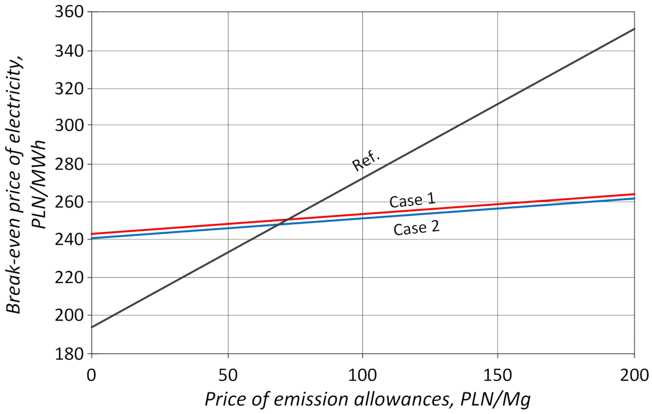

Figure 8: Break-even price of electricity as a function of emission allowances price for the analyzed systems

Table 5: Break-even price of membrane (in PLN/m^2) for different values of annual operation time and two prices of CO_2 emission allowances

	Case 1		Case 2	
t, h \ C_{ue}, PLN/Mg	91.5	200	91.5	200
5000	0.57	73.44	3.66	77.52
6000	7.62	95.15	11.05	99.73
7000	14.55	117.61	18.33	122.72
8000	21.33	139.41	25.50	145.10

on the value of the break-even price of electricity was analyzed.

In the first step for nominal assumptions (as shown in Table 5) the value of the break-even price of electricity as a function of the price of emission allowances was determined. The results of this analysis are shown in Fig. 8. The analysis shows that the point of intersection of the curves, and therefore the point at which the the break-even price of electricity is the same for the variant without capture and with capture, is determined by the price of emission allowances at the level of 65 PLN/Mg, and corresponds to the break-even price of electricity close to 250 PLN/MWh. It should be underlined, however, that in the cash flows of the units with capture installation, the costs associated with membranes were not included. In sum, the system with recirculation of the retentate had slightly better profitability.

An important factor influencing both the thermodynamic and economic characteristics of the CHP plant is annual operating time. Therefore, the characteristics shown in Fig. 9 demonstrate the influence of annual operating on the break-even price of electricity for two values of emission allowances C_{UE} (91.5 PLN/Mg and 200 PLN/Mg).

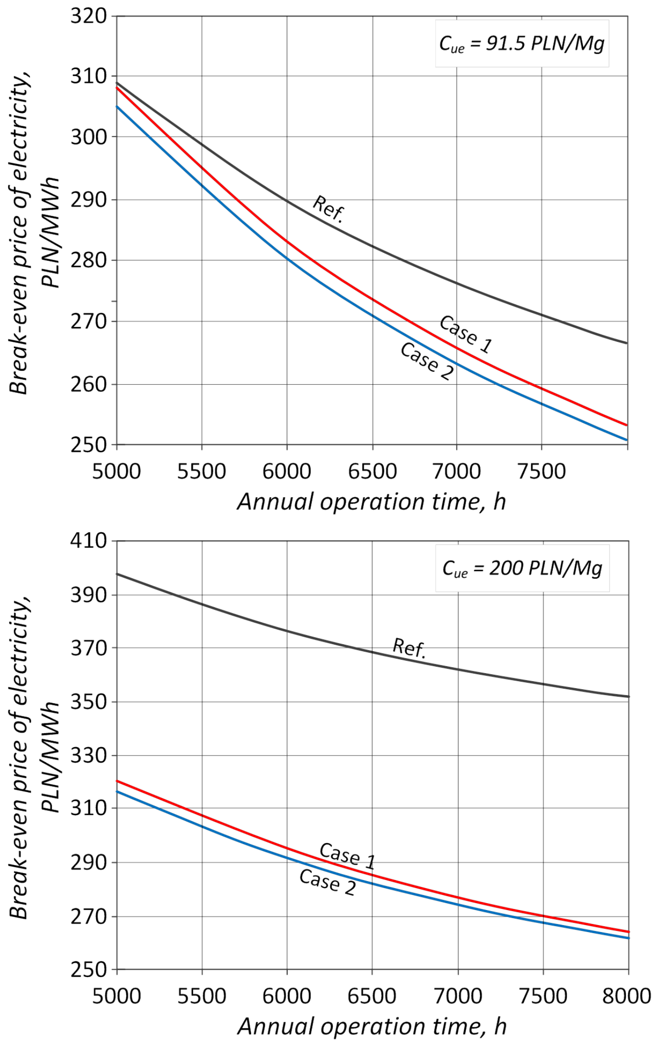

Figure 9: Break-even price of electricity as a function of annual operation time for emission allowances price at 91.5 PLN (top) and 200 PLN/Mg (bottom); Ref—reference case, i.e. without CO_2 capture

The characteristics shown in Figs 8 and 9 were determined without taking into account the costs associated with implementation of the membrane system. This approach is motivated by the desire to determine the break-even price of the membrane (per 1 m^2) which would provide equal profitability for the system with and without capture. The results of this analysis are shown in Table 5, for different annual operation times of the CHP unit for two prices of CO_2 allowances and for the system without and with retentate recirculation.

Regardless of the type of system, for the assumed CO_2 emission allowance prices, the break-even cost of the membrane obtained a positive value. However, for the CO_2 emission allowance price of 91.5 PLN/Mg the values obtained seem too low to ensure the profitability of the system compared to the system without capture. In light of the membrane costs cited in the literature (usually up to 20 \$/m^2), for CO_2 emission allowance prices of 200 PLN/ Mg, especially for longer annual operating times, units integrated with a membrane system may achieve better profitability than units without CO_2 capture.

The analysis shows the system with retentate recirculation to have better thermodynamic indices (lower energy consumption, higher net efficiency of electricity generation) and a slightly higher break-even price of membrane, although a larger total membrane area is required to achieve a defined separation effect in this system.

4. Summary and Conclusions

Carbon dioxide capture from flue gases in a CHP plant is problematic due to there being insufficient steam for use in the capture process at the time of operation of the unit for the needs of the district heating system. This paper presents the results of an analysis of systems which may serve as an alternative to the most commonly promoted CO_2 capture methods based on chemical absorption. The proposed solutions are based on a system consisting of a cascade of two membranes, in which one of the systems also uses recirculation of the retentate from behind the second membrane.

The analysis showed that the system with recirculation enjoys lower energy consumption than the system without recirculation. After optimizing the energy consumption of the two systems, the difference was 7%. Factoring in the energy needed to compress the carbon dioxide to the required pressure reduces this difference to 4% (due to the increased flow of compressed carbon dioxide for the system with recirculation).

The economic efficiency analysis suggests that systems with membrane separation of carbon dioxide can be competitive compared to the systems without capture. However, the cost-effectiveness of the use of membranes depends largely on the price of allowances for carbon dioxide emissions, the cost of membranes and annual operation time of the CHP unit. For allowances costing 91.5 PLN/Mg with an annual operating time of 8000 h, systems without capture will achieve the same profitability at a price of membrane system equal to 139 PLN/m^2 (without recirculation system) or 145 PLN/m$_2$ (system with recirculation).

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