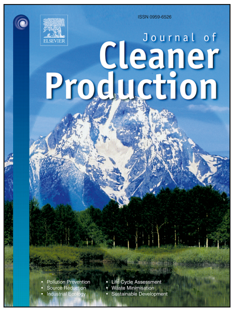

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Abstract

Pulp and paper is considered to be the fourth most energy-intensive industry (EII) worldwide. However, as most of the CO_2 emissions are of biomass origin, this sector has the potential to become a carbon-negative industry. This study proposes a new concept for conversion of the pulp and paper industry to carbon negative that relies on the inherent CO_2 capture capability of the Kraft process. The techno-economic performance of the proposed carbon-negative system, based on calcium looping (CaL) retrofitted to a pulp and paper plant, was evaluated. The effect of CaL design specifications and cost assumptions on the thermodynamic and economic performance were evaluated. Under the initial design assumptions, the reference pulp and paper plant was shown to turn from electricity importer to electricity exporter with the cost of CO_2 avoided equal to 39.0 €/tCO_2. The parametric study showed that an increase in the fresh limestone make-up rate resulted in a linear increase of the specific primary energy consumption for CO_2 avoided (*SPECCA*) and a reduction in the amount of electricity exported to the electric grid. This translates into an increase in the price of pulp and newsprint, and the cost of CO_2 avoided. This study has also demonstrated that the pulp and paper industry has high potential to become carbon negative. It has been shown that carbon capture and storage would become economically viable in this industry if the negative CO_2 emissions are recognised and a negative CO_2 emissions credit of at least 41.8 €/tCO_2 is implemented.

Keywords: Pulp and paper, calcium looping, carbon capture, techno-economic analysis, negative CO_2 emissions credit

1. Introduction

The pulp and paper industry generated 0.2 Gt of direct carbon dioxide (CO_2) emissions in 2017, accounting for 6% of industrial greenhouse gas (GHG) emissions in the UK (Griffin et al., 2018). It is considered as one of the main energy-intensive industries (EIIs), consuming 31,659 ktoe of primary energy in 2014 (Eurostat, 2016). However, the pulp and paper industry can become carbon negative due to the origin of its CO_2 emissions, which are mainly from biomass (Möllersten et al., 2004). This can be achieved by capturing and storing CO_2 or by using it as a raw material in other industries (Kuparinen et al., 2019). As biomass is the primary source of energy in the pulp and paper plant, the CO_2 emissions are considered carbon neutral, assuming the biomass is sustainably sourced, and integrated in a closed carbon cycle. During plant photosynthesis, biogenic CO_2 is captured from the atmosphere. Importantly, biogenic emissions are accountable in agriculture, forestry and other land-use and not in the energy sector (Intergovernmental Panel on Climate Change, 2014). Therefore, these emissions are not currently included in the European Union Emissions Trading System (EU ETS). Consequently, there is no incentive to implement carbon capture and storage (CCS) technologies by this industry (Onarheim et al., 2017a).

CCS is considered as a feasible route to deep decarbonisation of EIIs (Gerres et al., 2019). The techno-economic feasibility of industrial CCS technologies has been thoroughly studied in the iron and steel (Garðarsdóttir et al., 2018; Tian et al., 2018), cement (De Lena et al., 2019; Rolfe et al., 2018) and petrochemical (Fernández-Dacosta et al., 2017; Yao et al., 2018) industries. Nevertheless, economic assessments of CCS integration to pulp and paper plants are limited (Table 1).

Table 1. Literature data on economic assessments of CCS integration to pulp and paper plants

Reference	CCS integration	CO_2 capture cost [€/tCO_2]
Möllersten et al. (2006)	Pre-combustion physical absorption + CHP	18–27 €/tCO_2. 43 €/tCO_2 (long CO_2 transport distances)
Hektor and Berntsson (2009)	Amine scrubbing + CHP	29–51 €/tCO_2 (pulp plant) 20–66 €/tCO_2 (integrated pulp and paper plant)
McGrail et al. (2012).	Post-combustion amine scrubbing	52.5 €/tCO_2.
Onarheim et al. (2017a)	Post-combustion amine scrubbing	71–89 €/tCO_2 (integrated pulp and paper plant) 52–66 €/tCO_2 (pulp plant)
Nwaoha and Tontiwachwuthikul (2019)	Post-combustion amine scrubbing	114.8–117.4 €/tCO_2 (AMP) 122.5–131 €/tCO_2 (MEA)
Kuparinen et al. (2019)	CO_2 capture and utilisation + MEA	≤50 €/tCO_2

Möllersten et al. (2006) have performed a preliminary assessment of the potential integration of pre-combustion physical absorption in both the pulp plant and integrated pulp and paper plant with combined heat and power (CHP) generation. They found that the cost of CO_2 capture and storage, if the points of capture and storage are located in the same place, is in the range of 18–27 €/tCO_2. However, the CCS cost can increase to 43 €/tCO_2 for long transport distances (above 1000 km). Application of amine scrubbing for CO_2 capture from flue gas of the recovery boiler was assessed by Hektor and Berntsson (2009). They studied five possible configurations that combined CO_2 capture with CHP to overcome the additional steam demand. The extra energy demand was achieved by the following alternatives: upgrade biomass boiler, replace biomass boiler by natural gas combined cycle (NGCC), upgrade low-grade heat from the plant with a heat pump, process integration with a larger biomass boiler, or process integration combined with NGCC. Considering these scenarios, their study reported values of 29–51 €/tCO_2 for the pulp plant and 20–66 €/tCO_2 for the integrated pulp and paper plant. The techno-economic feasibility of retrofitting a pulp and paper plant with post-combustion amine scrubbing was also evaluated by McGrail et al. (2012). They have proposed replacing two existing natural gas-fired boiler and hog boiler with a larger biomass boiler. The latter would meet the additional demand for steam of the CCS unit. They concluded that the CO_2 capture cost was around 52.5 €/tCO_2. Onarheim et al. (2017a) have performed a comprehensive study to assess the techno-economic performance of retrofitting post-combustion amine scrubbing to both a pulp plant and integrated pulp and paper plant. They found that the cost associated with the integrated pulp and paper plant (71–89 €/tCO_2) was higher than for a standalone pulp plant (52–66 €/tCO_2) when 60–90% of the total CO_2 emissions were captured for both plants. For CO_2 capture rates below 60%, which implied only the CO_2 from the multi-fuel boiler and lime kiln flue gases was captured, these costs increased to 92 €/tCO_2 for the standalone pulp plant and 93 €/tCO_2 for the integrated pulp and paper plant. The costs associated with a retrofit of the pulp plant with post-combustion amine scrubbing were also estimated by Nwaoha and Tontiwachwuthikul (2019). They also compared the use of a conventional monoethanolamine (MEA) solvent with 2-amino-2-methyl-1-propanol (AMP) solvent for different process configurations. The use of AMP-MEA resulted in lower costs, in the range of 114.8–117.4 €/tCO_2, compared to 122.5–131 €/tCO_2 when MEA was used. Kuparinen et al. (2019) evaluated CO_2 capture and its potential on-site utilisation in the pulp plant and pulp and paper plant. They concluded that CO_2 capture is a feasible option for the pulp and paper industry, and estimated that the cost of CO_2 avoided can be below 50 €/tCO_2 if MEA was used as solvent (Kuparinen et al., 2017). However, such a low cost of CO_2 avoided was obtained because CO_2 was utilised on-site for production of bioproducts, such as tall oil, lignin and precipitated $CaCO_3$.

The review of the current literature has indicated that, to date, amine scrubbing was the only CCS technology considered for retrofits in pulp and paper plants. However, this technology has presented some challenges in the power industry, such as thermal degradation and adverse reactions of solvent with flue gas impurities such as NO_2, SO_2 and O_2 (Dean et al., 2011), the cost of solvent (Rao and Rubin, 2002), solvent concentration limited to 30 wt% (MEA) (Shao and Stangeland, 2009), high efficiency penalties of 9.5 to 12.5% points (Xu et al., 2010), and high volumes of waste generated (Dean et al., 2011). Therefore, more energy efficient and less expensive capture technologies have been explored. Calcium looping (CaL) has emerged as one of the promising technologies for decarbonisation of the power and industrial sectors. Importantly, the cost of CO_2 avoided from the CaL process has been shown to be as low as 20.5 €/tCO_2 when implemented in the cement industry (Rodríguez et al., 2012). This figure is one-third to one-sixth that reported for amine scrubbing retrofits in the cement industry (60.5–107 €/tCO_2) (Barker et al., 2009; Ho et al., 2011). Ca-based sorbents, such as limestone (~95%$_{wt}$ $CaCO_3$), are the most considered sorbents for CaL. Importantly, $CaCO_3$ is the main compound of lime mud, a bio-waste from the Kraft process in the pulp and paper industry. Furthermore, Sun et al. (2013) have shown that lime mud from the lime kiln can be successfully used as CO_2 adsorbent in CaL. Therefore, it is pertinent to assess the feasibility of using CaL for CO_2 capture in the pulp and paper industry.

The aim of this work is to assess the techno-economic feasibility of CaL retrofitted to a pulp and paper plant. The concept of the Kraft process with inherent CO_2 capture was proposed by integrating CaL in the existing lime cycle. In order to investigate the influence of CaL design specifications and economic assumptions on the techno-economic performance of the retrofitted process, a sensitivity analysis was carried out. The economic performance of the retrofitted pulp and paper plant was benchmarked against amine scrubbing. The impact of recognising negative CO_2 emissions on the cost of CO_2 avoided was also evaluated.

2. Process and model description

In this work, an integrated pulp and paper plant was selected as a reference plant, considering a process model developed in CADSIM Plus®. It was assumed that the reference plant produces 1000 ADt (air-dried tonnes, 90% dry content) of bleached pulp per day, 375 ADt/d of thermomechanical pulp and 450 ADt/d of newsprint. As some of the Kraft pulp and the thermomechanical pulp are consumed on-site, only 925 ADt/d of bleached pulp and newsprint are sold to the market.

2.1. Kraft process

The Kraft process involves conversion of raw wood into pulp, mainly cellulose fibres, which occurs in digesters with a solution of NaOH and Na_2S, so-called white liquor. The pulp is then separated from the solution, called black liquor, and forwarded to the fibre line. In order to recover the inorganic chemicals and produce steam for the entire process, the black liquor is burnt in a recovery boiler. This stage can generate 75% of the total CO_2 released in the plant (Garðarsdóttir et al., 2018). The molten black liquor (mainly Na_2CO_3 and Na_2S) is then dissolved in water to produce the green liquor. This solution is sent to a slaker where it is mixed with the lime (CaO) burnt in the lime kiln. At this point CaO is converted to $Ca(OH)_2$, which reacts with Na_2CO_3 to produce NaOH and $CaCO_3$. While the white liquor (mainly NaOH and Na_2S) is sent to the pulp digester to restart the cycle, the precipitated $CaCO_3$, called lime mud, is calcined in the lime kiln. Since this reaction is endothermic, the heat required to sustain it is generated by combustion of fossil fuels. The flue gas released during this step has the highest CO_2 concentration (~20%$_{vol}$), which is partially biogenic due to CO_2 formed during lime mud calcination. Importantly, the CO_2 emissions related to the calcination of the fresh limestone make-up are of fossil origin. The remaining CO_2 emissions come from the multi-fuel boiler that produces steam and/or electricity for the plant. Depending on the type of fuel used, the CO_2 emissions produced by this unit can be biogenic or of fossil origin. In that case the multi-fuel boiler refers to both the hog and power boilers. Unlike flue gas from the lime kiln, the multi-fuel and recovery boilers generate flue gas with lower CO_2 concentration (between 10–13%$_{vol}$). Table 2 shows the breakdown of the total CO_2 emissions produced by the pulp and paper plant. It can be observed that 4% of the total CO_2 emissions are of fossil origin and come primarily from the lime kiln. Around 34% of CO_2 from the lime kiln is of fossil origin, mostly because of the requirement for natural gas combustion, which is valid for the specific case study considered in this work. Although the main fuels are of fossil origin, methanol, tall oil, hydrogen, turpentine and strong odorous gases may also be burnt in the lime kilns (Kuparinen and Vakkilainen, 2017). Although the main fuels in the recovery and hog boilers are black liquor and wood, natural gas is also burnt during start-up. As around 96% of the total CO_2 emissions are biogenic, this industry has high potential to become a carbon-negative industry if CCS is implemented.

Table 2. CO_2 emissions breakdown for the pulp and paper plant without CCS.

Parameter	Recovery boiler	Hog boiler	Power boiler	Lime kiln
Biogenic CO_2 [t/d]	2299.7	823.0	-	191.4
Fossil CO_2 [t/d]	0.2	0.2	48.1	99.1
Total CO_2 [t/d]	2299.9	823.2	48.1	290.5

The reference pulp and paper plant consists mainly of a fibre line, the recovery and lime cycles, and the biomass and power boilers used to generate steam, which is then combined with that produced by the recovery boiler and sent to the steam turbine island. Part of steam is used in the process and the remaining part is converted to electricity that is used on-site. Due to the integration of paper production, which is an energy intensive process (Kuparinen et al., 2019; Möllersten et al., 2006; Onarheim et al., 2017a), the considered plant needs to import additional electricity from the electric grid. Although not depicted in the diagram (Figure 1), an air separation unit and a bleach chemical plant are also part of the plant to provide O_2 and ClO_2 to the fibre line. In this work, the concept of the Kraft process with inherent CO_2 capture is proposed for the integrated pulp and paper plant, as presented in the simplified block diagram in Figure 1. The proposed concept considers CO_2 capture by integration of CaL in the lime cycle. The existing lime kiln is replaced by a kiln of larger capacity, interconnected with the add-on carbonator, as shown in Figure 1. It is important to note that this is a general concept and that in a real application the process should be designed considering the causticisation requirements.

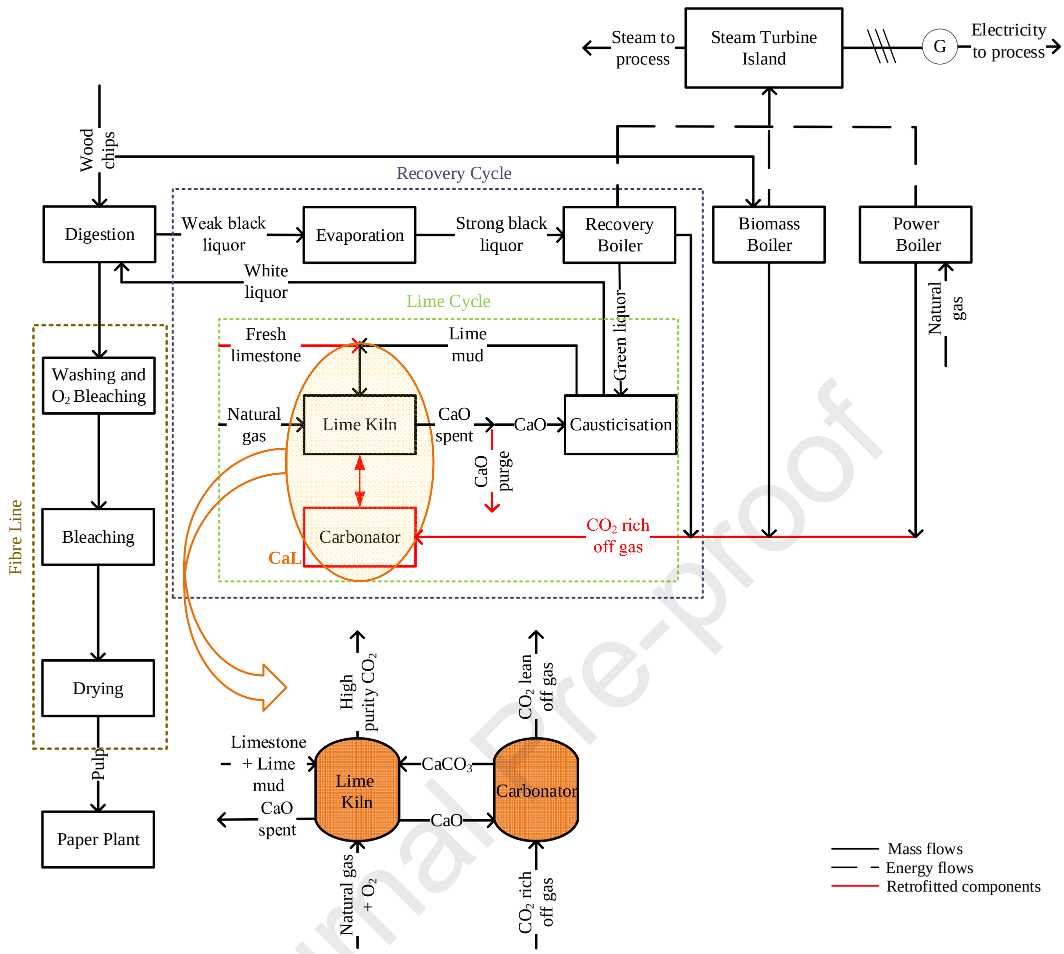


Figure 1. Kraft process concept with inherent CO_2 capture.

2.2. Reference pulp and paper plant model development

The performance of the reference pulp and paper plant was assessed using existing CADSIM Plus® model. Since the lime kiln was represented as a black box in CADSIM Plus®, it was modelled in Aspen Plus® to obtain the flue gas composition. The lime kiln was represented by two Gibbs reactors connected in series, where the Gibbs free-energy minimisation model was used to predict the equilibrium composition of the gas product. Natural gas combustion with air, which occurs in the first reactor, supplies the energy to achieve the desired temperature for calcination. The fuel rate was assumed based on the specific energy requirement of 6.5 GJ/tCaO, which was fixed in the CADSIM Plus® model (Schorcht et al., 2013). To ensure complete combustion of fuel, 12% excess air was assumed. In the second reactor, the lime mud is heated with the combustion gas to the calcination temperature (900°C). The mass and energy balances of the reference pulp and paper plant, which were used as input data to the CaL model, were validated against literature data (Table 3) (Nwaoha and Tontiwachwuthikul, 2019; Onarheim et al., 2017b). The results of the lime kiln modelled in Aspen

Plus® are also shown in Table 3. It can be concluded that the characteristics of the flue gas from the recovery boiler, power boiler, biomass boiler and lime kiln are comparable to those reported in the literature. The differences observed between the flue gas compositions can be attributed to the different operating conditions and type of fuel burnt in the plant. As mentioned previously, the power boiler can burn biogenic or fossil fuels and, depending on that, the flue gas composition will vary. Therefore, the power and biomass boiler results, whose fuel is natural gas and hog, respectively, are compared with the multi-fuel boiler. Natural gas is also burnt in the recovery and biomass boilers during start-up.

Table 3. Properties and composition of the flue gases in the reference pulp and paper plant.

Parameter	**This work**				**Literature data** (Nwaoha and Tontiwachwuthikul, 2019; Onarheim et al., 2017b)					
					Recovery boiler		**Multi-fuel boiler**		**Lime kiln**	
	Recovery boiler	**Power boiler**	**Hog boiler**	**Lime Kiln**	**Data** (Onarheim et al., 2017b)	**Data** (Nwaoha and Tontiwachwuthikul, 2019	**Biomass** (Onarheim et al., 2017b)	**Biomass + NG** (Nwaoha and Tontiwachwuthikul, 2019**)**	**Data** (Onarheim et al., 2017b)	**Data** (Nwaoha and Tontiwachwuthikul, 2019**)**
Temperature [°C]	210.0	150.0	130.0	204.0	184.0	185.0/ 192.0	189.0	163.0	250.0	196.0/ 207.0
Mass flow [MTPD]	11070.5	330.3	3957.0	1035.5	-	-	-	-	-	-
Mass flow [t/ADt]	11.1	0.3	4.0	1.0	10.2	2.0/ 4.5	1.9	3.1	0.9	0.4/ 0.4
N_2 [$\%_{vol}$]	63.5	71.7	56.6	43.3	67.6	58.1/ 54.9	53.4	59.7	47.4	53.6/ 55.8
O_2 [$\%_{vol}$]	1.7	0.7	0.8	1.2	2.3	8.3/ 6.1	1.7	10.1	1.2	9.6/ 8.9
H_2O [$\%_{vol}$]	21.4	18.4	29.8	37.7	17.0	20.9/ 24.1	32.7	19.3	30.9	22.9/ 21.6
CO_2 [$\%_{vol}$]	13.3	9.2	12.8	17.3	13.0	12.7/ 14.9	12.1	10.9	20.4	13.9/ 13.7
SO_x [ppm]	152.1	0.0	18.9	N/A	60.0	27.2/ 31.0	40.0	N/A	50.0	19.9/ 24.2
NO_x [ppm]	N/A	N/A	N/A	31.7	125.0	N/A	150.0	N/A	175.0	0.7/ 1.2

2.3. Calcium looping model development and integration

The retrofit of the reference pulp and paper plant with CO_2 capture can be achieved in the lime production without affecting the rest of the Kraft process. The flue gas streams from recovery, power and biomass boilers are merged and directed to the carbonator where the carbonation reaction, Eq. (1), takes place. During this step, CO_2 is captured by lime produced in the lime kiln. The decomposition of $CaCO_3$ into CaO and CO_2, Eq. (2), which occurs in the lime kiln (calciner), requires heat that is generated by oxy-fuel combustion. Thus, the sorbent circulates between the two reactors in alternate cycles of carbonation-calcination.

Carbonator: $CaO(s) + CO_2(g) \rightarrow CaCO_3(s)$ ΔH = -178 kJ/mol (1)

Calciner: $CaCO_3(s) \rightarrow CaO(s) + CO_2(g)$ ΔH = 178 kJ/mol (2)

The process model used in this work to simulate CaL integration to the pulp and paper plant, comprising a CaL process, a CO_2 compression unit (CCU) and a steam cycle, was modelled in Aspen Plus®. The CaL model was developed based on the work by Hanak et al. (2015) and validated with data from the 1.7 MW_{th} pilot plant at INCAR-CSIC (Sánchez-Biezma et al., 2013). The calciner, which was modelled as a Gibbs reactor, and the carbonator, which was modelled as a stoichiometric reactor, are the main components of the CaL process (Figure 1). As the conversion of the sorbent decreases over the carbonation-calcination cycles (Fennell et al., 2007; Grasa and Abanades, 2006), fresh limestone is fed, called make-up stream (F_0), to maintain the desired average conversion in the carbonator. In the pulp and paper plant, the make-up stream comprises lime mud from the causticisation process and fresh limestone, and part of the spent CaO is sent back to the causticisation. The maximum average conversion (X_{ave}), which depends on the carbonation (f_{carb}) and calcination extent (f_{calc}), the make-up rate (F_0), the solid looping rate (F_R) and the sorbent characteristics (a_1, a_2, f_1, f_2 and b), is estimated using the model proposed by Rodríguez et al. (2010). Considering the results presented by Sun et al. (2013), which showed at lab scale that lime mud can be employed as CO_2 sorbent, limestone was selected as the sorbent that best represents lime mud behaviour. The sorbent characteristics were selected based on the measurements from 1.7 MW_{th} INCAR-CSIC pilot plant (Sánchez-Biezma et al., 2013).

$$X_{ave} = (F_0 + F_R r_0) f_{calc} \left[\frac{a_1 f_1^2}{F_0 + F_R f_{carb} f_{calc} (1 - f_1)} + \frac{a_2 f_2^2}{F_0 + F_R f_{carb} f_{calc} (1 - f_2)} + \frac{b}{F_0} \right] \quad (3)$$

The gas stream generated in the calciner contains CO_2 and water vapour, which is condensed, and then a high-purity CO_2 stream is available for compression. This stream is initially compressed to 80 bar, above the critical pressure, in a multi-stage compressor. Then, the CO_2-rich stream is cooled to 25°C and compressed to 110 bar, which are the requirements for pipeline transport (Metz et al., 2005).

As a result of the exothermic reaction in the carbonator, a large amount of high-grade heat is available for recovery in a heat recovery steam generator (HRSG) and can be used to generate additional electricity in the steam cycle. The steam cycle, based on a superheated Rankine cycle without reheat, was modelled in Aspen Plus®. It was validated with the CADSIM Plus® model (Table 4), considering fresh water flowrate, steam temperature and pressure and the electricity generated by each turbine. The results were in good agreement between the two models, as the difference between compared parameters was less than 5%. In further analysis, it was assumed that live steam enters the high-pressure turbine at 593°C and 154 bar (Dryden, 1982). Furthermore, a heat exchanger network was introduced to maximise energy recovery (Figure 2). Importantly, the intermediate-pressure steam produced by the boilers is not represented in this simplified diagram.

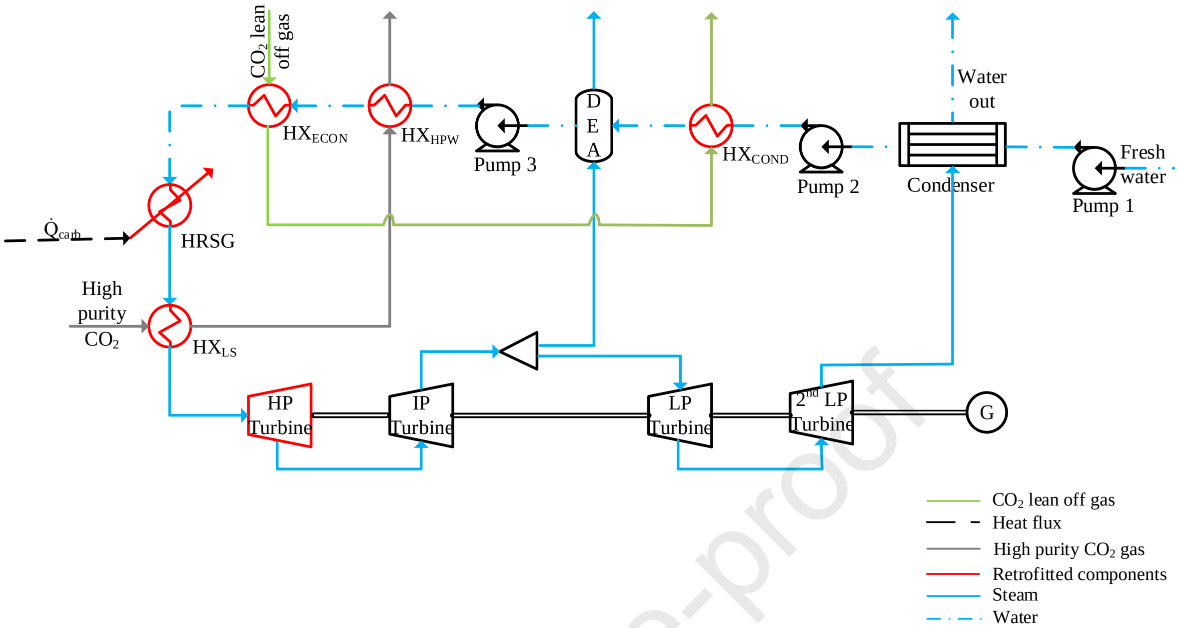


Figure 2. Calcium looping heat network.

Table 4. Steam cycle validation with CADSIM Plus data.

Parameter	Fresh water flowrate [t/d]		Temperature [°C]		Pressure [bar]		Power [kW_{el}]	
	CADSIM Plus	Model	CADSIM Plus	Model	CADSIM Plus	Model	CADSIM Plus	Model
High-pressure turbine	-	-	427.0	436.0	59.6	59.6	-	-
Intermediate-pressure turbine	-	-	244.0	255.0	11.7	11.7	22393.0	23374.0
First low-pressure turbine	-	-	181.0	190.0	4.8	4.8	6262.5	6450.0
Second low-pressure turbine	-	-	90.0	90.0	0.7	0.7	1131.3	1135.4
Condenser	5304.1	5344.6						

A cryogenic air separation unit (ASU) was not modelled in detail in this work, but its energy requirement was considered in the techno-economic assessment. It was assumed that the energy requirement to produce 1 t of O_2 at 95%$_{vol}$ purity is 200 $kW_{el}h$ (Romano, 2013). The main design conditions and thermodynamic assumptions used in modelling the proposed system are summarised in Table 5.

Table 5. Calcium looping model assumptions.

Unit operation	Parameter	Value
Calcium looping		
Carbonator	Temperature [°C]	650.0
	Carbonated sorbent fraction [-]	0.7
	CO_2 capture on carbonator [%]	90.0
Lime kiln (Calciner)	Temperature [°C]	900.0
	Calcined sorbent fraction [-]	0.95
	Excess oxygen [$\%_{vol,dry}$]	2.5
	Relative make-up [-]	0.04
Steam Cycle		
Live Steam	Temperature [°C]	593.0
	Pressure [bar]	154.0
High-pressure turbine	Isentropic efficiency [%]	92.0
	Mechanical efficiency [%]	99.8
Intermediate-pressure turbine	Isentropic efficiency [%]	77.5
	Mechanical efficiency [%]	96.5
Low-pressure turbine	Isentropic efficiency [%]	59.0
	Mechanical efficiency [%]	96.5
Second low-pressure turbine	Isentropic efficiency [%]	60.0
	Mechanical efficiency [%]	98.0
Condenser	Feed water temperature [°C]	10.0
CO_2 compression unit		
Compressors	Polytropic efficiency [%]	80.0
	Mechanical efficiency [%]	99.6
	Intercooler temperature [°C]	40.0
Pump	Isentropic efficiency [%]	80.0
	Mechanical efficiency [%]	99.6
CO_2 final stream	Temperature [°C]	25.0
	Pressure [bar]	110.0
	Purity level [%]	>95.0
Fresh material (Hanak and Manovic, 2018)	Limestone (95.0$\%_{wt}$ $CaCO_3$, 3.5$\%_{wt}$ $MgCO_3$, 0.6$\%_{wt}$ SiO_2, 0.4$\%_{wt}$ Fe_2O_3, 0.5$\%_{wt}$ Al_2O_3)	
Fuel (Hanak and Manovic, 2018)	Natural gas (93.1$\%_{vol}$ CH_4, 3.2$\%_{vol}$ C_2H_6, 0.7$\%_{vol}$ C_3H_8, 0.4$\%_{vol}$ C_4H_{10}, 1.0$\%_{vol}$ CO_2, 1.6$\%_{vol}$ N_2)	

3. Techno-economic feasibility assessment

To assess the effect of the CaL process integration with the reference pulp and paper plant, the process models presented in Section 2 were used to assess the techno-economic performance of the reference pulp and paper plant with and without CO_2 capture.

3.1. Thermodynamic performance indicators

Three parameters were used to evaluate the effect of CaL integration to the pulp and paper plant, including the net power output (P_{net}), the equivalent fuel consumption (q_{eq}) and the specific primary energy consumption for CO_2 avoided (*SPECCA*). The equivalent fuel consumption is defined in Eq. (4) as the sum of direct (q) and indirect fuel consumption of the pulp and paper plant. The latter is defined as the fuel consumption associated with the electricity imported (P_e) and depends on the electric efficiency (η_e) of the power generation.

$$q_{eq} = q + \frac{3600 \cdot P_e}{\eta_e}$$

(4)

The equivalent CO_2 emissions ($e_{CO_2,eq}$), given by Eq. (5), are calculated as the sum of direct (e_{CO_2}) and indirect ($P_e \cdot e_{CO_2,e}$) emissions. The latter is related to the electricity imported from the electric grid, thus depending on the specific CO_2 emissions source of the power plant ($e_{CO_2,e}$).

$$e_{CO_2,eq} = e_{CO_2} + P_e \cdot e_{CO_2,e} \tag{5}$$

If the retrofitted pulp and paper plant becomes a net electricity producer, the second term of Eq. (5) is negative and, therefore, results in negative indirect CO_2 emissions (De Lena et al., 2019). *SPECCA* is defined in Eq. (6), where the subscripts *ref* and *cap* correspond to reference pulp and paper plant without CO_2 capture and pulp and paper plant with CO_2 capture, respectively.

$$SPECCA = \frac{q_{eq,cap} - q_{eq,ref}}{e_{CO_2,eq,ref} - e_{CO_2,eq,cap}} \tag{6}$$

It should be noted that the characteristics of the power generation, electric efficiency (η_e) and the specific CO_2 emissions have an impact on *SPECCA.* For that reason, estimation of the indirect CO_2 emissions is based on the average non-CHP energy mix in the 27 EU Member States and the UK in 2015 ($e_{CO2,e}$=262 $kg_{CO2}/MW_{el}h$ and η_e=45.9%) (De Lena et al., 2019).

3.2. Economic performance indicators

Three economic parameters were selected to evaluate the effect of CaL integration to the pulp and paper plant: the levelised cost of pulp (*LCOP*), the levelised cost of newsprint (*LCON*) and the cost of CO_2 avoided (*AC*). In order to estimate the *LCOP* and *LCON*, the net present value (*NPV*) method was applied, as defined by Eq. (7) (Onarheim et al., 2017a). As a result, the levelised cost of product (pulp, newsprint) is the minimum sale price of that product at which *NPV* is zero. At such point, the present value of revenue from the sales of pulp, newsprint and potentially electricity are equal to the present values of expenditures.

$$NPV = \sum_{t=1}^{n} \frac{CF_t}{(1+r)^t} - TCR \tag{7}$$

The *NPV* method considers the discounted cash flow (CF_t) through the project lifetime (t), the total capital requirement (*TCR*) and the project interest rate (r). The economic assumptions made to estimate *NPV* are presented in Table 6. In order to simplify the calculations, inflation was not taken into account during the project lifetime. Thus, the market price of pulp and newsprint, as well as the price of raw materials and utilities are kept constant.

Table 6. Economic model assumptions.

Parameter	Value
Expected lifetime [y] (Martínez et al., 2014; Yang et al., 2010)	25.0
Project interest rate [%] (Martínez et al., 2014; Yang et al., 2010)	8.8
Capacity factor [%] (Martínez et al., 2014; Yang et al., 2010)	80.0
CO_2 emission allowance price [€/t_{CO2}] (Business Inside, 2020a)	23.74
Average GBP/EUR exchange rate 2017 (Bank of England, 2019)	1.1418

3.3. Cost estimation for pulp and paper plant

Since economic studies on the pulp and paper industry are scarce, estimation of capital and operating costs of the pulp and paper plant is based on research published by Onarheim et al. (2017a). These costs were calculated using Eqs. (8)-(11). TCR_{ref}, Eq (8), is the sum of the total plant cost (TPC_{ref}) and other capital costs ($OCAPEX$) which comprise spare parts, start-up, additional fuel costs, operation and maintenance, chemicals, owner's costs, working capital and interest during construction. These are calculated as a fraction of TPC_{ref}.

$$TCR_{ref} = TPC_{ref} + OCAPEX \tag{8}$$

In order to account for unexpected expenditures, a contingency plan (PC) was considered that, along with the total invested cost (TIC_{ref}), constitutes TPC_{ref}.

$$TPC_{ref} = TIC_{ref} + PC \tag{9}$$

TIC_{ref} was assessed using a scaling law that is the empirical correlation between the cost and the scale of the plant, as shown in Eq. (10).

$$\frac{C}{C_0} = \left(\frac{S}{S_0}\right)^n \tag{10}$$

In this empirical correlation, C represents the actual capital cost and S is the target capacity. The corresponding variables with the subscript 0 refer to the reference value. A cost exponent for the correction of capacity (n) of 0.6 was considered. Furthermore, as the reference capital cost (C_0) was reported for the year 2005, the scaled capital cost was adjusted to the year 2017 by using the Chemical Engineering Plant Cost Index, as shown in Eq. (11) (CEPCI, 2019).

$$C_{2017} = C_{2005}\frac{CEPCI_{2017}}{CEPCI_{2005}} \tag{11}$$

All the assumptions used to estimate the capital and operating costs are presented in Table 7. The operating costs include the fixed and variable components. The former were assumed as a fraction of the TCR_{ref}, while the latter include the costs of raw materials, chemicals, utilities and costs related to waste disposal. Most economic assumptions were based on the data reported by (Onarheim et al., 2017a) The utilities, raw materials and feedstock prices were also obtained from the same study, except for the sorbent price, natural gas price and the price of the electricity imported from the electric grid. The cost of electricity (87.3 €/MW_{el}h) was taken as the mean annual price of electricity to industrial consumers for the year 2017 (BEIS, 2019).

Table 7. Assumptions for capital and operating cost estimation of the reference pulp and paper plant.

Parameter	Value
$CEPCI_{2005}$ (CEPCI, 2019)	468.2
$CEPCI_{2017}$ (CEPCI, 2019)	567.5
Cost exponent for correction of capacity (Onarheim et al., 2017a)	0.6
Project Contingency (PC) [€] (Onarheim et al., 2017a)	10.0% of TIC_{ref}
Other CAPEX	
Spare parts [€] (Onarheim et al., 2017a)	1.0% of TPC_{ref}
Start-up CAPEX [€] (Onarheim et al., 2017a)	2.0% of TPC_{ref}
Additional fuel costs [€] (Onarheim et al., 2017a)	2.1% of TPC_{ref}
Operation and maintenance [€] (Onarheim et al., 2017a)	25.0% of TPC_{ref}
Chemicals and others [€] (Onarheim et al., 2017a)	8.3% of TPC_{ref}
Owner's cost [€] (Onarheim et al., 2017a)	7.0% of TPC_{ref}
Interest during construction, charged annually [€] (Onarheim et al., 2017a)	8.0% of TPC_{ref}
Working capital [€] (Onarheim et al., 2017a)	0.2% of TPC_{ref}
Fixed operating costs [€] (Onarheim et al., 2017a)	5.7% of TCR_{ref}
Waste and disposal [€/a] (Onarheim et al., 2017a)	11.1% of TCR_{ref}
Raw materials and feedstock	
Wood unit cost [€/m^3] (Onarheim et al., 2017a)	40.0
NaOH unit cost [€/tonne] (Onarheim et al., 2017a)	370.0
H_2O_2 unit cost [€/tonne] (Onarheim et al., 2017a)	500.0
$NaClO_3$ unit cost [€/tonne] (Onarheim et al., 2017a)	500.0
H_2SO_4 unit cost [€/tonne] (Onarheim et al., 2017a)	50.0
Methanol unit cost [€/tonne] (Onarheim et al., 2017a)	350.0
Limestone unit cost [€/tonne] (Lisbona et al., 2010; Martínez et al., 2014; Yang et al., 2010)	6.0
Natural gas unit cost [€/GJ] (Perry et al., 2007)	3.0
Hog fuel unit cost [€/m^3] (Onarheim et al., 2017a)	18.8
Unit cost of electricity imported from the grid [€/MW_{el}h] (BEIS, 2019)	87.3
Cooling water unit cost [€/m^3] (Onarheim et al., 2017a)	0.1
Process water unit cost [€/m^3] (Onarheim et al., 2017a)	0.1

3.4. Cost estimation for calcium looping

Besides the costs associated directly with CaL, the cost of the ASU (C_{ASU}), the cost of CCU (C_{CCU}) and the costs related to the steam cycle (C_{SC}) must be also considered. Although the reference plant has an ASU, it is assumed that a new ASU is required to cover the O_2 demand of CaL. The sum of these costs, given by Eq. (12), constitute the total capital requirement of the capture plant (TCR_{cap}).

$$TCR_{cap} = C_{CaL} + C_{ASU} + C_{CCU} + C_{SC} \quad (12)$$

The C_{ASU} and C_{CCU} were estimated based on correlations available in the literature, where the O_2 flowrate and the brake power requirement were used as the scaling factors. The investment cost of the

CaL and the steam cycle are based on the individual costs of their components, presented in Eq. (13) and Eq. (14).

$$C_{CaL} = (1 + i_{P\&C})(C_{calc} + C_{carb} + C_{FP} + C_{Fan}) \quad (13)$$

As shown in Eq. (13), it was assumed that the investment cost of CaL also accounts for the piping and integration cost, which is estimated using the piping and integration cost indicator ($i_{P\&C}$) of 5% (Michalski et al., 2019).

$$C_{SC} = C_{HPW} + C_{ECON} + C_{LS} + C_{COND} + C_{HRSG} + C_{ST} \quad (14)$$

The individual cost of each component, including calciner, carbonator, fuel preparation, fan and heat exchangers was determined from the correlations summarised in Table 8. Furthermore, all assumptions considered in the estimation of the CaL costs are also included in Table 7. It is worth noting that the fixed and variables costs were assumed to be 1 and 2% of TCR_{cap}, respectively. Although the costs associated with sorbent and fuel and the CO_2 transport and storage cost are not included in these fractions, they are considered in the calculations.

Table 8. Assumptions for capital and operating cost estimation of calcium looping

Unit operation	Cost correlation
Air separation unit [O_2 production rate, $\dot{m}_{O2}$ (kg/s)] (Atsonios et al., 2013)	$C_{ASU} = 2.926e5\left(\frac{\dot{m}_{O_2}}{28.9}\right)^{0.7}$
CO_2 compression unit [Brake power requirement, $\dot{W}_{CCU}$ (kW_{el})] (Kreutz et al., 2005)	$C_{CCU} = 1.22914e7\left(\frac{\dot{W}_{CCU,BRK}}{13000}\right)^{0.67}$
Steam turbine [Brake power output, $\dot{W}_{ST,BRK}$, (kW_{el})] (Aminyavari et al., 2016)	$C_{ST} = 3744.3(\dot{W}_{ST,BRK})^{0.7} - 61.3(\dot{W}_{ST,BRK})^{0.95}$
Heat exchanger high-pressure water [Heat exchange area, A_{HPW} (m^2)] (Lee et al., 2014)	$C_{HPW} = 130\left(\frac{A_{HPW}}{0.093}\right)$
Economiser [Heat exchange area, A_{ECON} (m^2)] (Lee et al., 2014)	$C_{ECON} = 130\left(\frac{A_{ECON}}{0.093}\right)$
Heat exchanger live steam [Heat exchange area, A_{LS} (m^2)] (Shirazi et al., 2012)	$C_{LS} = 2290(A_{LS})^{0.6}$
Heat exchanger condensate [Heat exchange area, A_{COND} (m^2)] (Lee et al., 2014)	$C_{COND} = 130\left(\frac{A_{COND}}{0.093}\right)$
Heat recovery steam generator [Heat exchange area, A_{HRSG} (m^2)] (Lee et al., 2014)	$C_{HRSG} = 130\left(\frac{A_{HRSG}}{0.093}\right)$
Calciner [Calciner heat flux, $\dot{Q}_{calc}$ (kW_{th})] (Michalski et al., 2019)	$C_{calc} = 13140\,(\dot{Q}_{cal})^{0.67}$
Carbonator [Carbonator heat flux, $\dot{Q}_{carb}$ (kW_{th})] (Michalski et al., 2019)	$C_{carb} = 16591\,(\dot{Q}_{carb})^{0.67}$
Fuel preparation system [Fuel flowrate $\dot{m}_F$(kg/s)] (Michalski et al., 2019)	$C_{FP} = 14158479\,(\dot{m}_{FP})^{0.24}$
Fan [Brake power requirement, $\dot{W}_{Fan,BRK}$ (kW_{el})] (Michalski et al., 2019)	$C_{Fan} = 103193\left(\frac{\dot{W}_{Fan,BRK}}{445}\right)^{0.67}$
Fixed operating costs [€] (Martínez et al., 2014; Yang et al., 2010)	1.0% of TCR_{Cap}
Variable operating costs [€] (Martínez et al., 2014; Yang et al., 2010)	2.0% of TCR_{Cap}
Limestone price [€/t] (Lisbona et al., 2010; Martínez et al., 2014; Yang et al., 2010)	6.0
Natural gas price [€/GJ] (Perry et al., 2007)	3.0
Electricity exported to the grid [€/MW_{el}h] (Onarheim et al., 2017a)	40.0
CO_2 transport and storage cost [€/t] (Romano et al., 2012)	7.0
Piping and integration costs indicator [%] (Michalski et al., 2019)	5.0

As mentioned above, the third economic parameter used to evaluate the proposed system is the cost of CO_2 avoided (*AC*), defined in Eq. (15). This figure is calculated based on the levelised costs of pulp and newsprint and the equivalent CO_2 emissions ($e_{CO_2,eq}$) of the pulp and paper plant with and without CO_2 capture. It is also dependent on the annual pulp and newsprint production, $\dot{m}_{Pulp}$ and $\dot{m}_{News}$, respectively.

$$AC=\frac{\left[\dot{m}_{Pulp}\frac{LCOP}{(\dot{m}_{Pulp}+\dot{m}_{News})}+\dot{m}_{News}\frac{LCON}{(\dot{m}_{Pulp}+\dot{m}_{News})}\right]_{cap}-\left[\dot{m}_{Pulp}\frac{LCOP}{(\dot{m}_{Pulp}+\dot{m}_{News})}+\dot{m}_{News}\frac{LCON}{(\dot{m}_{Pulp}+\dot{m}_{News})}\right]_{ref}}{e_{CO_2,eq,ref}-e_{CO_2,eq,cap}} \quad (15)$$

4. Results and discussion

The techno-economic feasibility of the proposed system was evaluated using the parameters reported in Section 3 and the design specifications presented in Table 5. To establish a direct comparison basis, the key indicators were estimated for both the reference and the retrofitted pulp and paper plants. To study the impact of CaL design specifications on the thermodynamic and economic performance, a parametric study was carried out.

4.1. Techno-economic performance

The thermodynamic analysis (Table 9) revealed that integration of CaL increases the on-site power requirement from 70.0 MW_{el} to 118.7 MW_{el}. In the base case, 85% of the total power required by CaL was associated with the power requirement of the ASU and CCU. Regardless of the increased power requirement, the amount of electricity generated overcame the energy demand of the retrofitted pulp and paper plant. In contrast to previous studies (Kuparinen et al., 2019; Möllersten et al., 2006; Onarheim et al., 2017a), in which integration of amine scrubbing reduces the net power output, this study showed that integration of CaL led to an increase in the net power output. While the operation of the reference pulp and paper plant relied on electricity import of 27.7 MW_{el}, the retrofitted pulp and paper plant became a net electricity export asset, exporting 9.6 MW_{el} of electricity to the electric grid. Therefore, the additional energy input required in CaL, reflected in 35% increase in the equivalent fuel consumption, is recovered in the steam cycle. Although there are no *SPECCA* data available for the pulp and paper industry, the specific primary energy consumption is more than double (5.7 $MJ_{LHV}/kgCO_2$) compared to the figures reported for the iron and steel industry (2.8 $MJ_{LHV}/kgCO_2$) (Tian et al., 2018) and the cement industry (2.39–3.27 $MJ_{LHV}/kgCO_2$) (De Lena et al., 2019; Rolfe et al., 2018). It should be noted that the latter value, presented by De Lena et al. (2019), corresponds to a total CaL integration; for the tail-end case, this value increased to 4.42 $MJ_{LHV}/kgCO_2$ avoided. The relative make-up of fresh sorbent (F_0/F_R) can explain the difference observed between this work and previous studies. This difference can also be attributed to the fact that the *SPECCA* considers the indirect CO_2 emissions. Therefore, the emissions associated with electricity import or export, the efficiency and emissions of the reference power generation have an impact on the final figure. Nevertheless, this work demonstrated that CaL is superior to post-combustion amine scrubbing that requires a heat duty of 3–5.25 $MJ_{LHV}/kgCO_2$ for the solvent regeneration only (Cormos, 2015; Kuparinen et al., 2019; Nwaoha and Tontiwachwuthikul, 2019).

The economic evaluation showed that the levelised costs of pulp and newsprint are 728.3 and 374.5 €/ADt without CCS and 824.4 and 411.1 €/ADt with CCS, respectively. Thus, the levelised costs of pulp and newsprint increase by 13% and 10%, respectively, on retrofit of CaL. It is noteworthy that the effect of limestone replacement by lime mud was considered in the economics evaluation, as use of lime mud can reduce the cost of fresh sorbent by up to 30%. The estimated CO_2 avoided cost is 39.0 €/tCO_2 (Table 9). This figure is comparable with the initial techno-economic studies considering amine scrubbing (Hektor and Berntsson, 2009; McGrail et al., 2012) and physical absorption (Möllersten et al., 2006). However, when compared with recent studies, which relied on up-to-date costs, the cost of CO_2 avoided for CaL is lower by around 50% and 65% than for post-combustion amine scrubbing (52–131 €/tCO_2) (Nwaoha and Tontiwachwuthikul, 2019; Onarheim et al., 2017a). The superior economic performance of CaL can provide sufficient incentives for the pulp and paper industry to invest in CCS.

Table 9. Summary of techno-economic performance

Parameter	Reference pulp and paper plant	Retrofitted pulp and paper plant
Thermodynamic assessment		
Gross power output [MW_{el}]	42.3	128.3
On-site power requirement [MW_{el}]	70.0	118.7
Net power output [MW_{el}]	27.7	-9.6
Equivalent fuel consumption [MJ_{LHV}/ADt]	25458	35923
SPECCA [MJ_{LHV}/kg CO_2 avoided]	-	5.7
Economic assessment		
Levelised cost of pulp [€/ADt]	728.3	824.4
Levelised cost of newsprint [€/ADt]	374.5	411.1
Cost of CO_2 avoided [€/tCO_2]	-	39.0

It needs to be emphasised that this work aimed to quantify the potential of the CaL integration in the pulp and paper industry. Therefore, the optimisation of the steam turbine island retrofit was considered outside of the boundary defined for the economic assessment. Therefore, the capital costs for modifications of the existing turbines and generator were not accounted for in this study.

4.2. Sensitivity analysis

The techno-economic performance indicators were first evaluated for a range of relative make-up rates, varied between 0.01 and 0.06. As shown in Figure 3, increasing the relative make-up of fresh sorbent resulted in a nearly linear rise in *SPECCA*. The specific exported electricity to the grid reduced on an increase in the make-up rate, which is attributed to the fact that less heat is available in CaL for recovery. This can be explained by the increase of fresh limestone fed to the calciner and consequently, a higher sorbent conversion was achieved in the carbonator and less solids were recirculated. These results are in agreement with the previous study on CaL retrofit (Hanak and Manovic, 2017). Once the retrofitted pulp and paper plant becomes an electricity exporter, the net power output reduction also has a negative impact on the *SPECCA*, increasing the equivalent fuel consumption and the equivalent CO_2 emissions. Namely, an increment of 0.02 from the initial value of F_0/F_R (0.04), led to a 44% reduction in the amount of electricity exported to the grid and a 6% raise in *SPECCA* (Figure 3). Considering the economic performance (Figure 4), an increase of F_0/F_R translates into a rise in the cost of pulp, newsprint and the CO_2 capture. An increment of 0.02 from the initial value of F_0/F_R (0.04), resulted in a 1% increase in the levelised cost of pulp and newsprint (Figure 4a) and a 7% increase in the cost of CO_2 avoided (Figure 4b). However, for values below 0.02, the sorbent activity decay becomes more pronounced and limits the decrease of SPECCA (Figure 3) and the levelised costs (Figure 4). In practice, this is not a desirable situation as very low solid recirculation rate implies the need for larger reactors and, therefore, higher operational costs (Rodríguez et al., 2010)

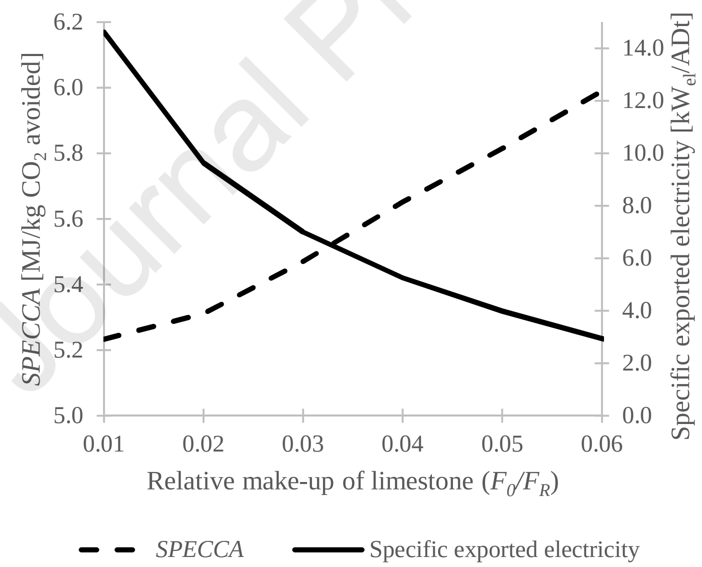
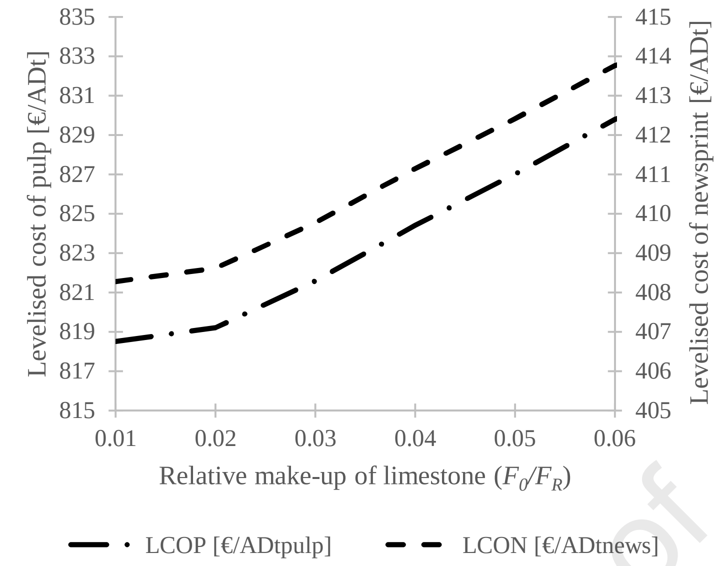
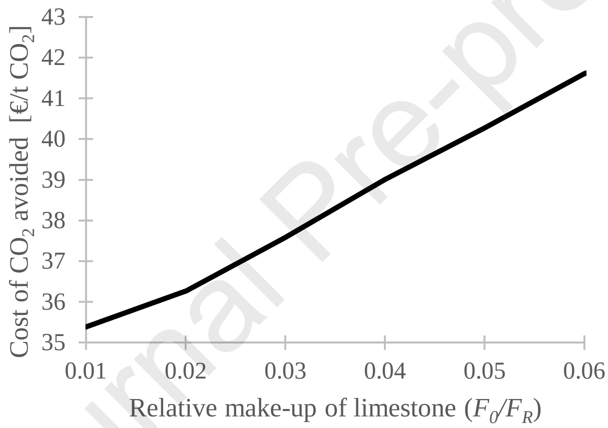


Figure 3. Impact of fresh limestone make-up rate on *SPECCA* and specific exported electricity.



a)



b)

Figure 4. Impact of fresh limestone make-up rate on (a) levelised cost of pulp and newsprint and (b) cost of CO_2 avoided.

Furthermore, as the costs related to CaL are uncertain, a sensitivity analysis was performed by varying the main assumptions in the economic model (Figure 5). The CO_2 avoided cost was estimated by varying (±25%) the initial values of capital, fixed and variable costs of CaL as well as the CO_2 transport and storage, natural gas and limestone prices. It can be observed in Figure 5 that the natural gas price, which has been decreasing in the last year in North America (Business Inside, 2020b), and the CaL capital requirement are the two parameters with the highest impact on the CO_2 avoided cost. Importantly, a 25% reduction in these parameters corresponded to a reduction in the CO_2 avoided cost of 16% and 11%, respectively. It was also found that CaL fixed and variable costs along with the sorbent price have a small impact (<2%) on the CO_2 avoided cost.

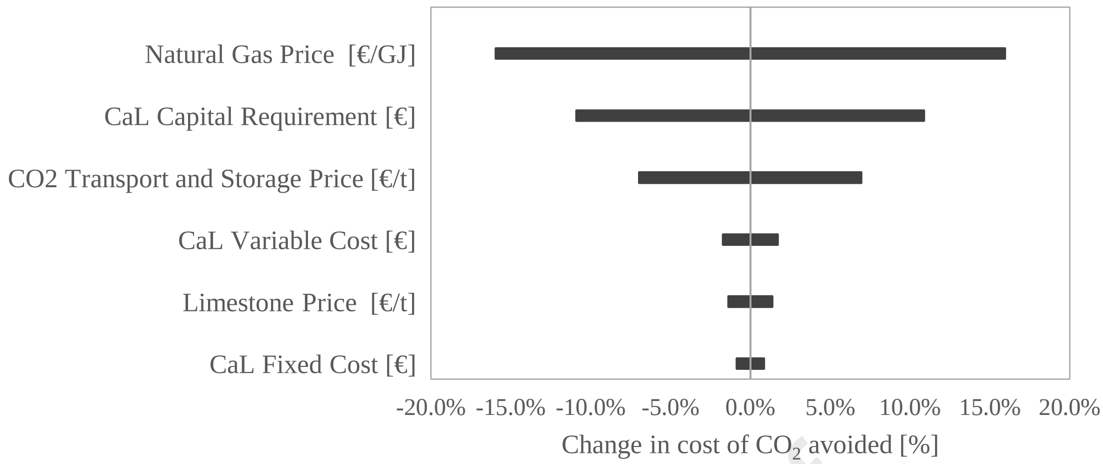


Figure 5. Effect of the key economic parameters on the economic performance of the retrofitted pulp and paper plant.

Finally, the economic indicators were also assessed considering the following scenarios:

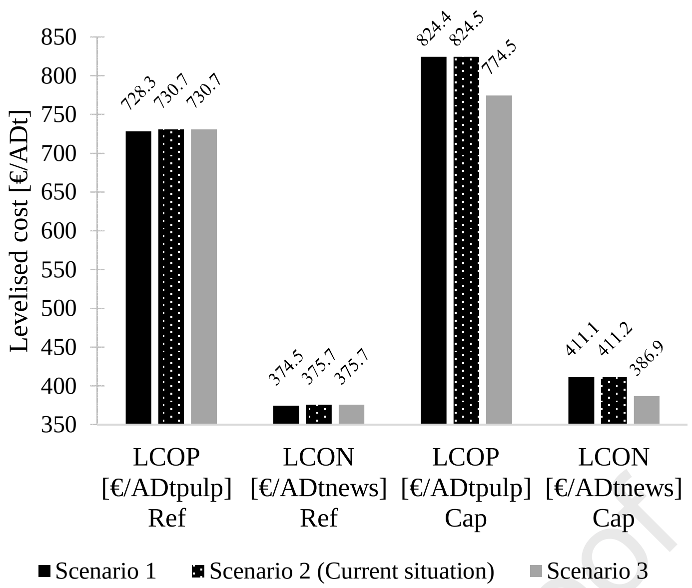
Scenario 1: No CO_2 emissions taxes and no credits for negative emissions (base line scenario)

Scenario 2: Fossil CO_2 emissions tax and no credits for negative emissions (current situation)

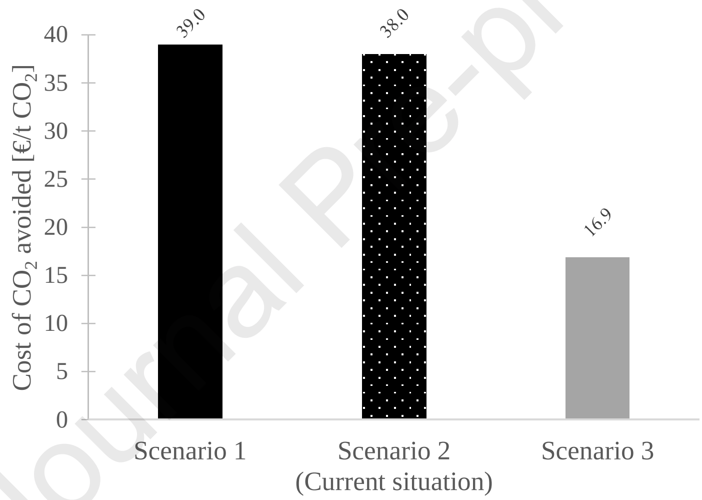
Scenario 3: Fossil CO_2 emissions tax and credits for negative emissions

Before reporting the results obtained, it is worth mentioning that a negative CO_2 emissions credit of 23.74 €/tCO_2 was assumed, which is equal to the current price of CO_2 emission allowance under the EU Emission Trading System (ETS) (Business Inside, 2020a). It should be noted that for a direct comparison, the economic indicators were also estimated for the pulp and paper plant with and without CO_2 capture.

As previously mentioned, CO_2 emissions are considered as carbon neutral under the current EU ETS biogenic. Therefore, only fossil CO_2 emissions are subject to the price of CO_2 emission allowances (Scenario 2). As shown in Figure 6, the CO_2 emission allowance price levied on fossil emissions results in a marginal increase of the levelised costs for the reference plant and no change for the retrofitted plant. The CO_2 emission allowance price is also shown to have a minimal impact on CO_2 capture cost, as the CO_2 avoided cost changed from 39.0 €/tCO_2 (Scenario 1) to 38.0 €/tCO_2 (Scenario 2). This can be explained by the fact that around 96% of the total CO_2 emissions produced by the reference plant were biogenic. Nevertheless, it is clear from these figures that introduction of credits for negative emissions (Scenario 3) has the strongest effect on the levelised cost and CO_2 avoided cost. This can be attributed to the profit obtained from the negative CO_2 emissions. In this study, retrofit of CaL was characterised with an overall CO_2 capture rate of 94%, corresponding to 0.9 MtCO_2/a of negative CO_2 emissions. Consequently, the levelised costs in the retrofitted plant reduced by 6% (Figure 6a) and cost of CO_2 avoided (Figure 6b) decreased from 38.0 €/tCO_2 (Scenario 2) to 16.9 €/tCO_2 (Scenario 3). Thus, the CO_2 capture cost and pulp and newsprint prices are strongly affected by negative CO_2 emissions credits. For that reason, a parametric study was also carried out by varying the CO_2 emission allowance price between 0 and the value at which the cost of CO_2 avoided equals zero. This analysis was performed under Scenario 3, and the results are shown in Figure 7. The sensitivity analysis on electricity imported/exported price is also illustrated in Figure 7. The cost of CO_2 avoided was estimated for three electricity prices, 40 €/MW_{el}h (price electricity exported, base line), 87.3 €/MW_{el}h (price electricity imported, base line) and 120 €/MW_{el}h. Under the initial design assumptions (baseline), it was found that the negative CO_2 emissions credit must be 41.8 €/tCO_2, which means there is no cost associated with CO_2 capture and, therefore, the levelised costs of the retrofitted plant equal the levelised costs of reference plant. The corresponding levelised costs of pulp and newsprint would be 732.6 and 376.6 €/ADt, respectively. Because in this analysis the fossil fuel emissions were taxed, implying an additional cost, these values are slightly higher than the ones presented for the reference pulp and paper plant (Scenario 1). As also shown in Figure 7 the cost of CO_2 avoided depends strongly on the electricity price. As an example, an increase of 25% in the CO_2 emission allowance price initial value presented in Table 6 (23.74 €/tCO_2) results in cost of CO_2 avoided changing between 4.9 and 20.6 €/tCO_2.



a)



b)

Figure 6. Effect of different economic scenarios on (a) levelised cost of pulp newsprint and (b) cost of CO_2 avoided (Ref and Cap correspond to reference pulp and paper plant and retrofitted pulp and paper plant, respectively).

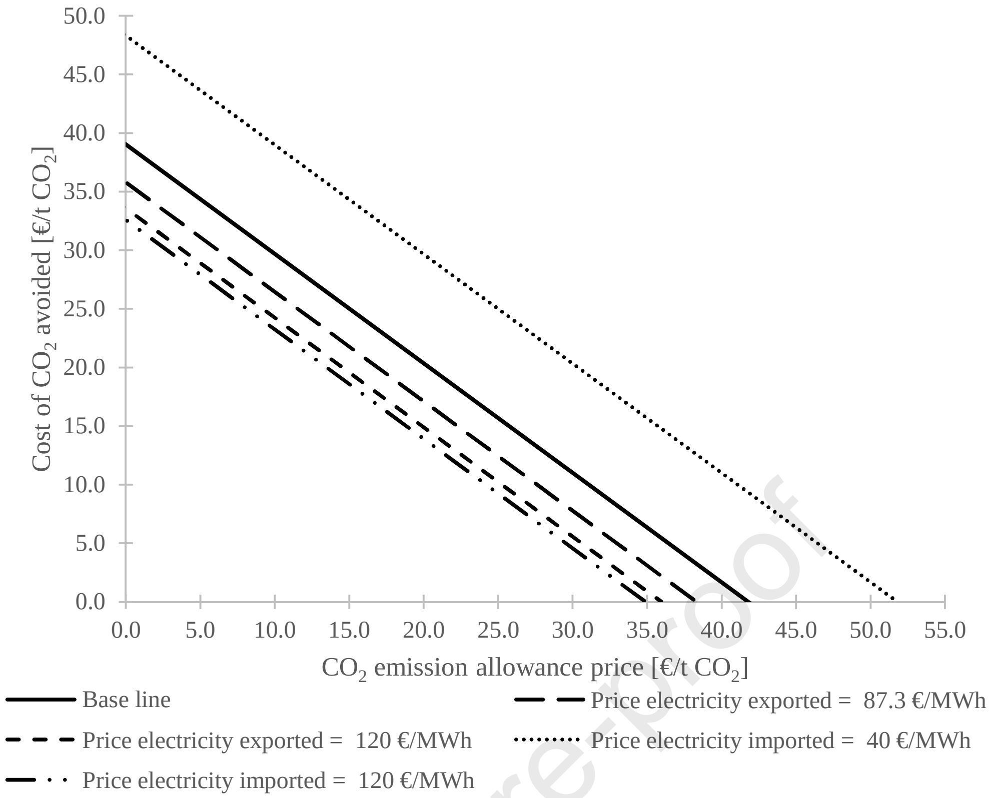


Figure 7. Impact of CO_2 emission allowance price and electricity price on the cost of CO_2 avoided under scenario 3.

5. Conclusions

This study proposed a concept of the Kraft process with inherent CO_2 capture for a pulp and paper plant. Such a concept can be added to existing pulp and paper plants by integrating a CaL process in the existing lime cycle. The techno-economic feasibility of the proposed system was assessed. Under the initial design assumptions, it was found that the reference pulp and paper plant can turn from electricity importer to electricity exporter. Moreover, the cost of CO_2 avoided is estimated to be 39.0 €/tCO_2. This figure is superior to that recently reported for pulp and paper plants retrofitted with amine scrubbing using MEA as a solvent. Such a superior performance can be associated with the fact that the energy input required for sorbent regeneration in CaL is recovered to generate additional electricity in the steam cycle.

A sensitivity analysis on techno-economic performance was also carried out by varying parameters, such as fresh limestone make-up rate, costs associated to CaL, and the sorbent and fuel prices. It was found that an increase in the make-up rate results in a linear rise of *SPECCA* and a reduction in electricity exported to the electric grid, corresponding to an increase in the pulp and newsprint prices as well as in the cost of CO_2 avoided. The latter was strongly affected by the natural gas price and CCS capital requirement. Further work should consider optimisation of the steam cycle integration in the pulp and paper plant with CaL and should account for the cost of modifications to the existing steam cycle.

This study showed that the pulp and paper industry has high potential to become carbon negative, which with a change of policies would make CCS implementation feasible in this industry. Yet, the CCS feasibility depends strongly on the inclusion of biogenic emissions in the EU ETS and/or on the attribution of credits for them. Considering CaL as an emerging technology for CO_2 capture in the pulp and paper industry, its implementation would be viable with the recognition of negative CO_2 emissions and a negative CO_2 emission credit of 41.8 €/tCO_2 applied. Therefore, biogenic emissions should be considered in future policies and incentivised by the implementation of negative CO_2 emissions credits. Therefore, further work is required to develop policies that will incentivise adoption of cleaner production technologies. Such policies should enable carbon-intensive industries to become carbon neutral or, as in case of the pulp and paper industry considered in this study, even carbon negative.

Acknowledgements

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Nomenclature

a_1 , a_2	sorbent maximum average conversion model fitting parameter [-]
A_j	heat exchanger area of equipment j [m^2]
AC	cost of CO_2 avoided [€/$t_{CO2avoided}$]
b	sorbent maximum average conversion model fitting parameter [-]
C_j	capital cost of equipment j [€]
CaL	calcium looping
$CEPCI$	Chemical Engineering Plant Cost Index
CF_t	discounted cash flows through the project lifetime [€]
DEA	deaerator
e_{CO2}	direct CO_2 emissions from the pulp and paper plant [kg_{CO2}/ADt]
$e_{CO2,e}$	specific CO_2 emissions associated with power generation [kg_{CO2}/MW_{el}h]
$e_{CO2,eq}$	equivalent CO_2 emissions [kg_{CO2}/ADt]
f_1, f_2	sorbent maximum average conversion model fitting parameter [-]
f_i	reaction extent [-]
F_0	make-up rate (fresh limestone and lime mud) [kmol/s]
F_R	sorbent looping rate [kmol/s]
G	generator
HX	heat exchanger
$i_{P\&C}$	piping and integration costs indicator [%]
HP	high pressure
IP	intermediate pressure
$LCOP$	levelised cost of market product, pulp [€/ADt]
$LCON$	levelised cost of market product, newsprint [€/ADt]
LP	low pressure
$\dot{m}_F$	fuel flowrate [kg/s]
$\dot{m}_{O2}$	O_2 production rate [kg/s]
n	cost exponent for the correction of capacity [-]
$\dot{m}_{News}$	newsprint production per year [ADt/a]
$\dot{m}_{Pulp}$	pulp production per year [ADt/a]

MEA	monoethanolamine
NPV	net present value [€]
OCAPEX	other capital cost [€]
P_e	specific energy [$MW_{el}h$/ADt]
P_{net}	net power output [MW_{el}]
PC	project contingency [€]
q	direct fuel consumption [MJ_{LHV}/ADt]
q_{eq}	equivalent fuel consumption [MJ_{LHV}/ADt]
$\dot{Q}_j$	heat flux of equipment j [kW_{th}]
r	discount rate [%]
r_0	fraction of never calcined limestone in the system [-]
S	target capacity [ADt/d]
SPECCA	specific primary energy consumption for CO_2 avoided [$MJ_{LHV}/kg_{CO2avoided}$]
t	project lifetime [y]
TCR	total capital requirement [€]
TIC	total installed cost [€]
TPC	total plant cost [€]
$\dot{W}_j$	brake power requirement/output of equipment j [kW_{el}]
X_{ave}	average sorbent conversion [-]

Greek letters

η_e	electric efficiency [-]

Subscripts

0	reference value
ASU	air separation unit
BRKP	brake power
calc	calciner
cap	pulp and paper plant with CO_2 capture
carb	carbonator
CCU	CO_2 compression unit
COND	condensate
e	electric
eq	equivalent
ECON	economiser
FP	fuel preparation

HPW high-pressure water

HRSG heat recovery steam generator

LS live steam

MTPD metric tonne per day

ref reference pulp and paper plant without CO_2 capture

SC steam cycle

ST steam turbine

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Highlights

- Concept of the Kraft process with inherent CO_2 capture
- Calcium looping for CO_2 capture is integrated in the lime cycle
- Reference pulp and paper plant turns from importer to exporter of electricity
- Pulp and paper industry has a high potential to become carbon negative
- Economic feasibility needs the inclusion of biogenic emissions in emission trading

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Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: