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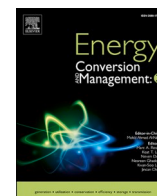
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Techno-economic evaluation of carbon capture via physical absorption from HTL gas phase derived from woody biomass and sewage sludge

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ABSTRACT

Due to its capability to produce negative CO₂ emissions, bioenergy in combination with carbon capture and storage (BECCS) has been identified as a key technology to limit global warming and to support the energy transition in pursue of the climate targets of this century. Among different bioenergy applications, advanced liquid biofuels produced through the hydrothermal liquefaction (HTL) of waste biomass have gained interest as promising drop-in alternatives to fossil fuels. However, there is lack of studies in literature that evaluate the potential of HTL as negative emission technology in the context of BECCS and present detailed process design of such an implementation for different types of biomass. In this paper, we perform carbon capture modeling based on state-of-art experimental data on HTL of waste lignocellulosic and urban biomasses by means of physical absorption via the Selexol™ process. The process model is utilized for developing a techno-economic analysis that highlights key parameters to optimize CO₂ capture cost efficiency. The results indicate that the purity of the CO₂ product fulfils the requirements for geological storage in all cases studied, and is on-spec for pipeline transportation when the composition of C₂₊ hydrocarbons and H₂S in the HTL gas are kept below 4 and 1 mol % respectively, or by the implementation of two absorption steps. For the standard process evaluated, the estimated cost of the captured CO₂ is between 40 and 53 EUR/tonne, which is in the range of the carbon price expected within this decade in the EU emission trading system, given the announced cap reductions until 2030 and the targets set by the European Green Deal.

1. Introduction

The implementation of carbon dioxide removal in the energy sector has been thoroughly recommended by the Intergovernmental Panel on Climate Change (IPCC), supported by the United Nations Environment Programme and World Meteorological Organization (WMO), to secure that the global temperature rise is kept below 1.5 °C within this century. In this context, Negative Emission Technologies (NET's) will be crucial to meet the climate targets, and among these, Bioenergy with carbon capture and storage (BECCS) could play a central role [1]. While in conventional carbon capture and storage (CCS) and bioenergy applications the process can be carbon neutral at its best, the combination of both technologies has potential to be carbon negative, as the anthropogenic carbon in the biomass is used for energy production and the CO₂ obtained as by-product can then be stored, having overall negative emissions.

In the range of BECCS applications, there has been increasing interest in studying the combination of advanced biofuels and CCS. Advanced

biofuels are liquid fuels aimed for the transportation sector produced from certain feedstock that meet sustainability and GHG emission criteria set by the Renewable Energy Directive (REDII) [2]. Examples are lignocellulosic feedstocks (i.e. agricultural and forestry residues), non-food crops (i.e. grasses, miscanthus, algae), or industrial waste and residue streams that demonstrate low CO₂ emission or high GHG emission reduction, and reach zero or low indirect land-use change (ILUC) impact. Due to their compatibility with existing infrastructure, advanced drop-in liquid biofuels can facilitate a faster and smoother energy transition, particularly in the heavy segment of the transport sector where electric solutions are not expected to be commercial in the short-medium term (i.e. maritime, aviation, long-haul road). Furthermore, the REDII in the European Union has set a target of 3.5% advanced biofuels in the transport sector by 2030 [2].

One of the emerging technologies used to effectively produce advanced liquid biofuels is hydrothermal liquefaction (HTL), in which biomass is processed in aqueous medium at temperatures and pressures close to the critical point of water, within a range of 100 to 350 bar and

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temperatures between 250 °C and 450 °C. Under these conditions, the rigid polymeric structure of the biomass disintegrates having as main product a liquid bio-crude. In a secondary step, the bio-crude is typically upgraded through hydrotreatment (e.g. hydrogenation and hydrodeoxygenation) to remove residual oxygen molecules and other heteroatoms transferred from the feedstock such as nitrogen and sulfur, whereafter it can be refined using the existing refinery technologies to meet requirements for transportation fuels [3].

The feedstock flexibility of HTL is high and numerous types of residual biomass have been successfully processed at laboratory/pilot scales including lignocellulosics, algae, sewage sludge, food waste and more. On-going projects at demonstration scale aim to scale up the technology and bring it to commercialization [3]. Still, techno-economic challenges remain regarding product demineralization, upgrading costs (e.g. hydrogen consumption), aqueous phase management and nutrients recovery. The estimated minimum fuel selling price (MFSP) of HTL biofuels remains higher than for their fossil counterparts being in the range of 0.5–3.6 EUR/L [4–8]. Being the bio-crude the main product, most literature available on HTL has focused on different aspects of the fuel production, while there is lack of studies that evaluate its potential to perform as NET taking advantage of the relatively high gas production (15–45%) and high CO₂ concentration in the gas. Only few recent studies in the field discuss the performance of HTL coupled with BECCS in comparison with other biomass-based technologies [9,10], however, these do not provide a detailed evaluation on the technical and economic aspects of the carbon capture implementation. In the study by Cheng et al [9], carbon capture is modelled as a black box via amine-based system with fixed efficiency but the impact of impurities in the final requirements for transport/storage and the economic aspects are not discussed. In the review by Li and Wright [10] carbon sequestration in the biochar from HTL is discussed but the HTL gas phase is not included in the analysis.

Looking at the HTL process with more detail, Fig. 1 shows a typical configuration with heat integration where the gas phase produced in the process is used as fuel to provide the additional heat required to reach the desired reaction temperature in the feed (350–400 °C). Even though direct combustion of the HTL gas phase is a possibility, its high CO₂ concentration (>60% mol.) is not ideal in terms of combustion performance and makes of it a poor quality fuel. In this context, separation of the CO₂ before combustion of the gas is an interesting option due to its high concentration in the gas, its high pressure, the improvement in the combustion efficiency and the possibility to recover H₂ from the HTL gas. In the HTL process with bio-crude hydrotreating, the H₂ cost plays a significant role in the economics of the process and its recovery from the HTL gas in stand-alone HTL configurations in combination with carbon capture can be an interesting alternative.

In our recent study [11], a detailed implementation of HTL in combination with carbon capture for CCS was investigated for forestry residues based on physical absorption with the Selexol™ solvent, showing promising results from the techno-economic and environmental

perspective. Physical absorption is the most mature technology for pre-combustion capture, and is preferred over chemical absorption due to the higher capacity of physical solvents at high CO₂ concentration in the gas, the lower energy requirements for solvent regeneration, and the high solvent stability and its non-corrosive nature [12]. Among different processes that use physical solvents, the Selexol™ process has shown the greatest potential for acid gas removal [13]. The solvent used in Selexol™ consists of a mixture of Dimethyl Ethers of Polyethylene Glycol (DEPG) that absorb CO₂ more effectively compared to other key components present in the HTL gas such as H₂, CO and CH₄ [14]. The results of the previous evaluation indicated that a cost-effective separation of CO₂ from the HTL gas phase is possible using the Selexol™ technology to achieve the purity requirements for pipeline transportation with a low impact in the MFSP. However, the process is sensitive to the presence of C₂₊ hydrocarbons in the inlet gas that decrease the purity of the CO₂ and represent a loss in the energy potential of the combustible gas. Since the purity of the transported CO₂ is essential to maintain the supercritical phase of the fluid, it typically entails a concentrated stream of > 95 mol % despite the lower requirements for storage at 90 mol. % [15,16].

Complementary to that study and with more focus on the carbon capture step, the objective of this paper is to evaluate if the prospective applicability of the Selexol™ process to treat HTL gas derived from lignocellulosics feedstock can be projected onto other types of feedstock, particularly wet biomasses such as sewage sludge and food waste with great potential for HTL. The sensitivity of the final CO₂ purity to the presence of hydrocarbons in the HTL gas is analysed in this study for a wider range of HTL gas compositions. The presence of significant amounts of nitrogen and sulphur containing compounds in urban feedstocks differs from the lignocellulosic case and has been reported in the gas product in the form of N₂, NH₃ or H₂S but their impact in the performance of the carbon capture process has not been evaluated and is undertaken in this study. Furthermore, an alternative configuration is proposed to address the limitations of the process regarding the hydrocarbons co-absorption in the solvent and the consequent reduction of the energy potential in the combustible gas. In this paper, the aim is to investigate the applicability of the Selexol™ process for a broader range of gas composition and to evaluate an alternative configuration by means of detailed techno-economic evaluation to estimate the cost range of captured CO₂ in the HTL process.

The structure of the paper comprises a methodology section followed by results and discussions and finally conclusions. In the first section, composition scenarios are defined for the HTL gas derived from the two types of feedstock considered, namely forestry residues (FR) and sewage sludge (SS), based solely on published experimental data. In the methodology section, the process configurations evaluated are described as well as the methodology for cost estimation. Next, the results are discussed in terms of: 1) the key differences between the types of HTL gas/feedstock evaluated; 2) CO₂ recovery and purity obtained for the two feedstock scenarios in the configurations evaluated compared to the requirements for pipeline transportation and geological storage, 3) impact of minor impurities on purity and recovery, and 4) cost estimation of the process to establish the cost range of the captured CO₂.

2. Process description and methodology

2.1. Gaseous product from the HTL process

The evaluation of the Selexol™ process is based on published experimental data regarding the composition of the gas produced from the HTL process for two different types of feedstock: woody biomass [17–19] and sewage sludge [20,21]. The cited references correspond mainly to data obtained in continuous operation at temperatures and pressures between 350 and 400 °C and 200–300 bar for the woody feedstock, and 260–350 °C and 180–200 bar for sludge, which are within the typical range in the HTL literature. Different composition scenarios are created for the two types of gas by varying the CO₂ content

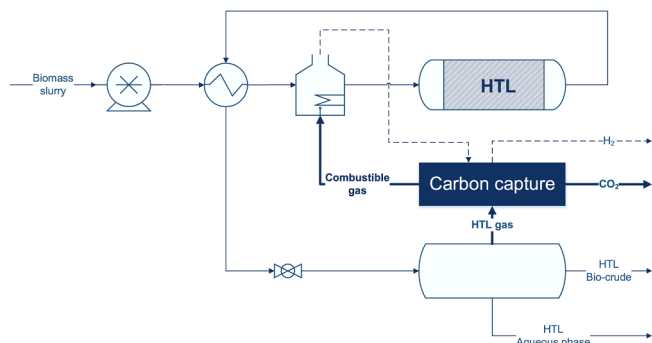


Fig. 1. Role of the HTL gas phase in the process and carbon capture.

in the range reported followed by normalization of the remaining components. These are presented in Fig. 2 along with the CO₂ purity requirements for pipeline transportation and geological storage [16]. The scenarios with the minimum and maximum CO₂ content are indicated for both types of gas. The detailed gas composition can be consulted in the Supplementary information.

It can be observed that despite of the variation in process conditions of the sources consulted, CO₂ is by far the compound with the highest concentration and there is a trend in the relative composition of the remaining compounds. Given the high concentration of CO₂ as a starting point for the carbon capture step, the separation of impurities is first evaluated via gas cooling followed by condensation in order to assess whether it is possible to achieve the purity requirements without additional treatment. Due to its reliability for wide range of temperatures and pressures, the Soave-Redlich-Kwong (SRK) property package is chosen for this step.

2.2. Selexol™ process

The Selexol™ process is designed to perform the carbon capture of the HTL gas based upon the schemes described in the existing literature regarding the removal of CO₂ from high-pressure syngas [22]. The base case scenario is designed for the HTL gas composition of the woody biomass feedstock (FR_{base}) and the gas flow used in the model is set to be 15 tonne/h, in line with the estimates of our previous study for an HTL plant size with an output of 2500 barrels of bio-crude a day [11]. A typical layout of the Selexol™ process fed with the HTL gas is presented in Fig. 3, highlighting the main stages that are described as follows.

Prior absorption, the first step corresponds to removal of condensables from the HTL gas which is necessary due to the significant water content expected in the gas at continuous operation and the high solubility of water and other condensables in the solvent (higher hydrocarbons and ammonia), which is detrimental for the absorption performance and should be avoided [12]. Since the experimental gas composition is reported in dry basis, the gas is saturated with water at the pressure and temperature of the upstream separator. Based on the separation conditions reported at pilot scale in [23], a gas temperature of 150 °C and pressure of 40 bar can be reasonably expected in continuous operation.

After condensates removal, the HTL gas is fed to the absorber tower where it enters in contact with the solvent. The separation occurs by physical absorption due to the higher solubility of the CO₂ in the DEPG solvent relative to H₂, CO, N₂ and CH₄, while remaining condensables and C₂₊ hydrocarbons are co-absorbed in the solvent. This is due to the higher solubility of the C₂ + hydrocarbons in the solvent relative to H₂, CH₄ and CO, which are significantly less soluble at the conditions in the column [11]. The gas and liquid flow in the absorber in counter-current mode, facilitating the transport of CO₂ to the solvent while the non-

soluble gases are separated as a combustible gas. The enriched solvent leaves at the bottom of the absorption column and is regenerated by means of consecutive expansion stages to be recycled back. The gas from the first expansion steps is typically fed back into the absorption column as it contains significant amounts of combustible gasses that are partially dissolved, and CO₂ of higher purity is obtained after the last expansion for further cooling and compression. The process is modelled in Aspen HYSYS V9 using the PC-SAFT property package, specifically developed and validated by Aspen Tech for acid gas cleaning processes, for a broad spectrum of conditions and components [12]. The modelling of the configuration with one absorber tower in Fig. 3 is described with more detail in the next section, and based on this layout, a configuration with two absorption steps is proposed in Section 2.2.2 as an alternative for the scenarios with high hydrocarbons co-absorption.

2.2.1. Single-step absorption

Table 1 shows the main process parameters used in the modeling based on the approach presented in our previous publication for woody biomass-derived HTL gas [11]. This configuration is evaluated for the two types of gas based on the composition in Fig. 2. A sensitivity analysis of the CO₂ purity and recovery with variation in the process conditions (solvent temperature and flash pressure) is presented. The final solvent flash temperature is fixed at 1 bar and therefore the sensitivity analysis is evaluated on the first and second flash pressure. The number of stages in the absorber and the solvent flow (L/V ratio) is established for a high CO₂ recovery in the solvent. For the base case, detailed stream results are provided in the Supplementary material and overall mass and energy balances are presented in the results section for the main process steps in Fig. 4. Heat integration between heat exchangers is evaluated by means of pinch analysis setting a minimum terminal temperature difference of 20 °C.

Particularly in the sewage sludge-derived HTL gas, the impact of minor impurities -H₂S and N₂ - on the CO₂ purity and recovery and the transport/storage purity requirements is evaluated in more detail. The H₂S and N₂ content in the dry HTL gas have been reported in the range of 0 to 1.5% and 0 to 4 mol. % respectively [20,21]. The presence of ammonia is evaluated within the same N₂ limits, as it has been identified in the aqueous HTL effluent from sewage sludge and other N containing feedstock and therefore it is also expected in the gas during continuous operation. The content of these impurities is varied within the established limits followed by normalization of the remaining components.

2.2.2. Two-step absorption

This configuration is proposed to separate the highest soluble compounds from the HTL gas, particularly C₂₊ hydrocarbons, in a first absorption step by using CO₂-saturated solvent. This facilitates the subsequent separation of CO₂ from the non-soluble compounds in the second absorber for further cooling and compression. Due to the high

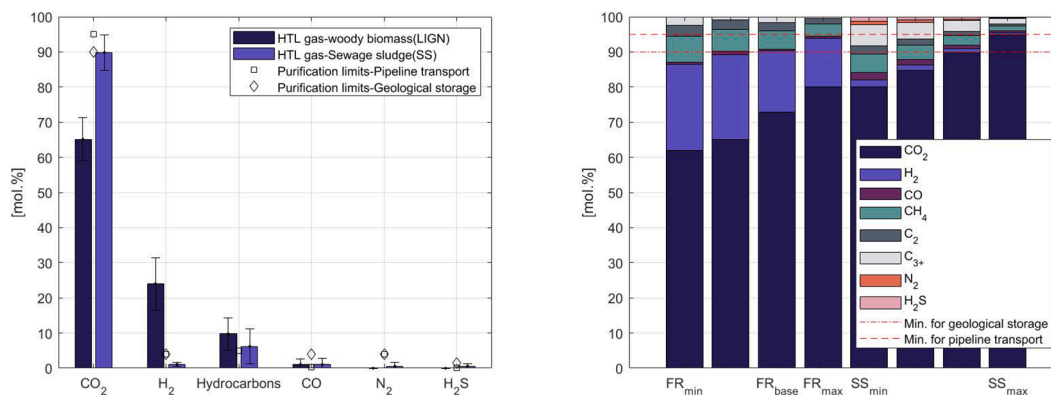


Fig. 2. Composition scenarios of HTL gas (dry basis) derived from woody biomass (FR) and sewage sludge (SS) based on experimental data, and minimum CO₂ concentration for pipeline transportation and geological storage.

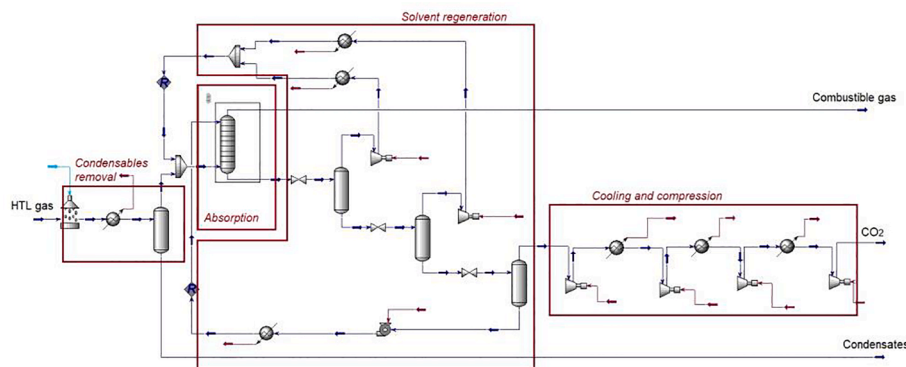


Fig. 3. Carbon capture from HTL gas via physical absorption in typical process configuration of Selexol™ process.

Table 1

Main parameters in the modelling of single stage absorption process in Aspen Hysys.

Section	Variable	Value
Absorption column	Pressure [bar]	40
	Solvent temperature [°C]	10.5
	L/V (inlet gas)	1.95
	Number of separation stages	18
Solvent regeneration	Flash pressure [bar]	18; 9; 1
CO ₂ compression train	Pressure levels [bar]	3.5; 15; 50; 150

solubility of the compounds separated in the first absorber, addition of heat is necessary for solvent regeneration. Oxy-fuel combustion of the gas serves in this scheme as a way to recover the CO₂ for solvent saturation and it suits the ultimate intended use of the C₂₊ hydrocarbons for internal heat production. This two-step absorption configuration is similar to previously reported absorption layouts for syngas desulfurization [24]. This is also an interesting alternative to be further studied

in combination with H₂ production from electrolysis and pressure swing adsorption (PSA) in stand-alone HTL to separate H₂ from the HTL gas phase before combustion (light blue in Fig. 5, not modeled in this study).

The main modeling parameters used are summarized in Table 2. A detailed modelling of the oxy-fuel combustion is out of the scope of this work, so the oxy-fuel burner is modeled as an ideal conversion reactor fed with O₂ in stoichiometric quantities, assuming complete combustion and having as only products CO₂ and H₂O for the woody-derived gas, and additional SO₂ and N₂ (inert) when applicable for the sludge-derived gas. After combustion, H₂O is separated by cooling and the impact of the remaining compounds (H₂O, N₂, SO₂) in the CO₂ recirculation is evaluated. The energy potential of the combustible gas prior combustion is compared with the combustible gas in the previous configuration. As for the single-stage absorption, detailed stream results are provided in the Supplementary material and overall mass and energy balances are presented for the base case for the main process steps in Fig. 5. Heat integration between heat exchangers is evaluated by means of pinch analysis setting a minimum terminal temperature difference of 20 °C.

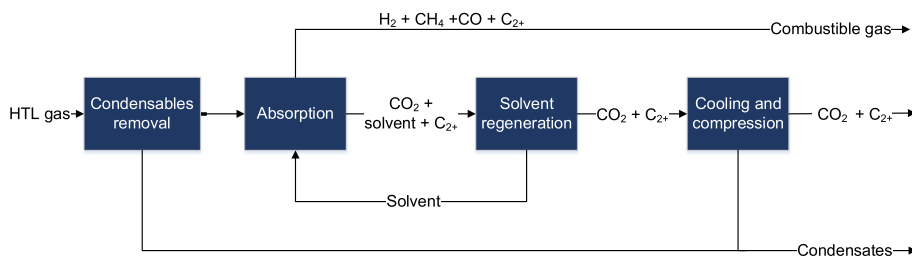


Fig. 4. Process flow diagram of carbon capture from HTL gas via Selexol™ process with single stage absorption.

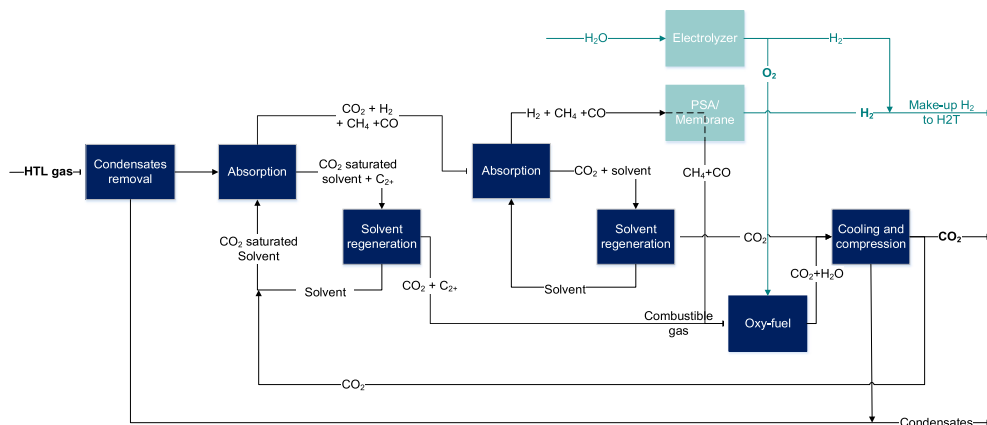


Fig. 5. Process flow diagram of two-step absorption for carbon capture from HTL gas phase.

Table 2

Main parameters in the modelling of two-step absorption process in Aspen Hysys.

Section	Variable	First absorption step	Second absorption step
Absorption column	Pressure [bar]	40	39
	Solvent temperature [°C]	5	3
	L/V (inlet gas)	0.84	1.25
	Number of separation stages	8	8
Solvent regeneration	Flash pressure [bar]	30; 20; 15	25; 1
	Reboiled column stages	10	–
	Reboiled column pressure [bar]	1.2	–
CO ₂ compression train	Pressure levels [bar]	3.5; 15; 50; 150	

2.3. Cost estimation

The capital investment of both configurations is estimated using the Aspen Process Economic Analyzer™ V9, linked to the process simulation results in Aspen HYSYS, with costs reported in 2015 pricing basis. The variable operational costs are estimated based on the price ranges presented in Table 3 for European electricity and heat markets. Operation and maintenance costs (O&M) and fixed operational costs are determined based on fixed factors used by Aspen Process Economic Analyzer and the suggested number of operators for the modelled plant. Revenues from excess heat from the process are included in the analysis given its potential for district heating use.

*Fixed Capital Investment

Based on the capital and operational expenses a net present value (NPV) analysis is performed to estimate the cost of captured CO₂ (Eq. (4)) based on a 10% discount rate (r), 8000 operating hours per year and a project lifetime of 25 years (n). The variation in the cost of captured CO₂ is evaluated by means of Monte Carlo analysis with random variation of the parameters in Table 3.

$$NPV = \sum_{i=1}^n \frac{\text{Cashflow}_i}{(1+r)^i} = 0 \quad (1)$$

$$0 = \sum_{i=1}^n \frac{\text{Income}_i - \text{OPEX}_i}{(1+r)^i} - \text{CAPEX} \quad (2)$$

$$0 = \sum_{i=1}^n \frac{\text{ExcessHeat}_i * \text{Heatprice} + \text{CapturedCO}_{2i} * \text{CaptureCost} - \text{OPEX}_i}{(1+r)^i} - \text{CAPEX} \quad (3)$$

$$\text{CaptureCost} \left[\frac{\text{EUR}}{\text{tonne}} \right] = \frac{\text{CAPEX}[\text{EUR}] + \sum_{i=1}^n \frac{\text{OPEX}_i[\text{EUR}] - \text{ExcessHeat}_i[\text{MWh}] * \text{Heatprice} \left[\frac{\text{EUR}}{\text{MWh}} \right]}{\sum_{i=1}^n \frac{\text{CapturedCO}_{2i}[\text{tonne}]}{(1+r)^i}} \quad (4)$$

3. Results and discussion

3.1. CO₂ purification by condensation

The composition of the HTL gas from woody biomass and sewage sludge presented in Fig. 2 shows the CO₂ as the major component for both types of biomass, being in the range of 60–72 mol % for the lignocellulosic case and in a higher proportion of 85–95 mol % in the

Table 3

Cost parameters for economic evaluation.

	Cost component	Unit	Min	Base and Max
Variable operational costs	Electricity [25]	EUR/MWh	40.00	75.00
	Heating	EUR/MWh	25.00	50.00
	Cooling water	EUR/MWh	0.40	0.75
Operation and maintenance (O&M)	Hourly labor cost[26]	EUR/h	20	30
	Maintenance cost	% of FCI*	1	3
	Operating charges	% of Labor cost	25	–
Fixed operational costs	Plant overhead	% of O&M	50	–
	General and administration costs	% of OPEX	8	–
Revenues	Excess heat [27]	EUR/MWh	18	36

case of sewage sludge. Hydrogen and methane are more abundant in the woody case, while C₂₊ hydrocarbons are present in similar amounts in the order of 5%. The dew curves in the pressure–temperature diagram (Fig. 6a) show that in both cases the temperature of the gas should be decreased below 25–30 °C approximately to initiate the separation of condensables. For the lignocellulosic case, the V-L region covers a wider range of pressures and the bubble curve goes well above 300 bar (not shown), due to the significant higher composition of hydrogen. The individual points represent the critical points of CO₂, methane and ethane as reference.

Fig. 6b shows the CO₂ composition of the gas after removal of condensables at different temperatures. As the gas is cooled down, the composition increases due to water condensation until 73% and 90% for the lignocellulosic and sludge cases respectively at temperatures close to ambient. The drop in composition below 10–20 °C is explained by loss of CO₂ in the liquid phase. Additional results on the VLE indicate that, by condensation, the maximum concentration of CO₂ in the liquid phase is about 90% for both cases, being still below the targeted 95% purity even at significantly low temperatures (–30 °C) as already presented previously for the woody biomass case [11]. Therefore, the Selexol™ process is evaluated for both feedstock scenarios and the temperature for condensates removal is set at 30 °C to avoid CO₂ losses in the liquid.

3.2. Selexol™ process with single-step absorption

The results of the single-step absorption regarding the CO₂ purity and recovery are summarized in Fig. 7 and in Table 4 compared to the requirements for pipeline transportation and storage. The process results in a CO₂ product with a purity of 92–98 mol % and 90–97 mol % for the woody biomass-derived and sludge-derived HTL gas respectively.

Overall, for both cases the purity of the produced CO₂ increases with the CO₂ composition in the inlet gas, however, it is limited by the presence of impurities, being the C₂₊ hydrocarbons the most dominant.

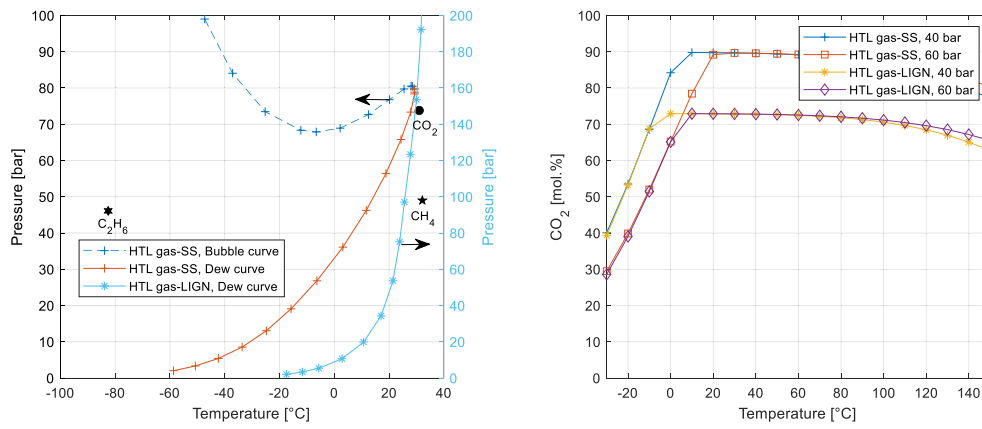


Fig. 6. (a) Pressure-Temperature diagram of HTL gas in lignocellulosic and sludge scenarios, (b) CO₂ composition of saturated HTL gas after removal of condensables at different temperatures.

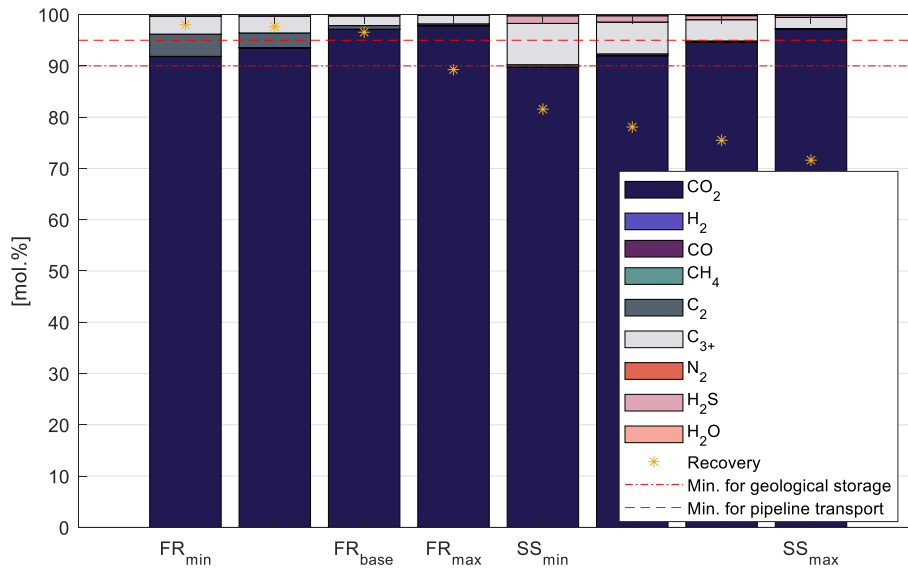


Fig. 7. Modeling results of purity and recovery of CO₂ from HTL gas derived from woody biomass (FR) and sewage sludge (SS) for single-step absorption via Selexol™.

Table 4

Composition of CO₂ product from single-step absorption of HTL gas derived from woody biomass (FR) and sewage sludge (SS) compared to purification limits for transportation and storage.

Component	Pipeline transportation	Geological storage	FR min	FR max	SS min	SS max
CO ₂	95	90	91.83	97.86	89.78	97.19
H ₂	4	4	0.00	0.00	0.00	0.00
Hydrocarbons	5	NR*	7.92	2.09	8.50	2.30
CO	0.2	4	0.00	0.00	0.00	0.00
N ₂	4	4	0.00	0.00	0.00	0.00
H ₂ S	0.02	1.5	0.00	0.00	1.48	0.41
H ₂ O	0.005	0.05	0.25	0.05	0.24	0.10

*NR: Not reported.

Even though in the sludge case the starting CO₂ concentration is higher (average 90 mol %), the composition of C₂ + is comparable to the lignocellulosic scenario due to the relatively lower concentration of H₂ and CH₄. The process fulfils the CO₂ requirement for storage in both types of gas but the minimum purity for transportation is satisfied only in the scenarios where the C₂+ hydrocarbons content in the HTL gas is below 4–4.5 mol % approximately. Even though partial dehydration of the gas is achieved in the process, removal of remaining water to avoid pipeline corrosion during transportation is typically done by the use of

different commercial technologies available (silica gel, molecular sieves, among others [16]), however, is not included in the analysis.

Regarding the recovery, it ranges between 89 and 98 % (96.6 in the design point) for the woody-derived gas, and between 72 and 82 % for the sludge derived gas. Overall, higher CO₂ concentration in the feed gas relative to the design point led to lower recoveries, as more CO₂ is present in the system while the solvent flow remains constant, decreasing the L/V ratio in the column. Even though in the sludge scenarios the L/V ratio can be adjusted to achieve higher recovery, higher

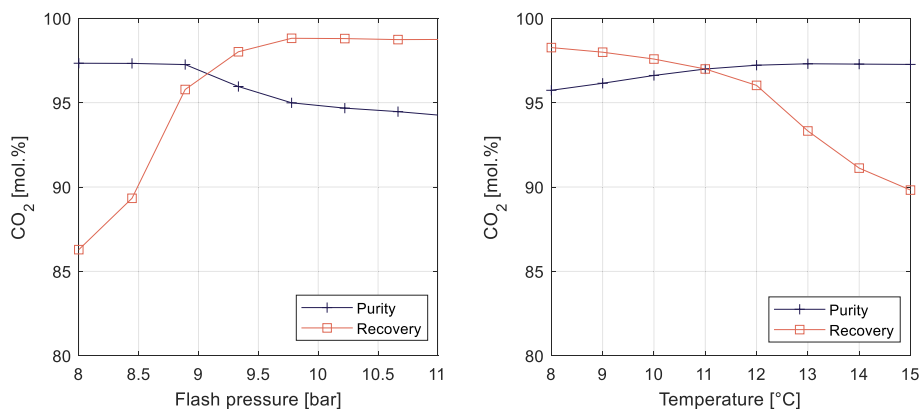


Fig. 8. Impact of varying flash pressure of second expansion and solvent temperature on CO₂ purity and recovery relative to design configuration.

purity would still be limited by co-absorption of C₂ + impurities.

3.2.1. Sensitivity analysis of CO₂ purity and recovery with pressure and solvent temperature

Fig. 8 shows the impact of variations in the flash pressure for solvent regeneration and solvent temperature at the inlet of the absorber tower in the CO₂ purity and recovery relative to the design configuration. Overall, purity decreases with higher flash pressures in the second expansion and lower solvent temperatures due to the increased solubility of the impurities in the solvent that end up with the CO₂ product after the final decompression. The same impact in the CO₂ solubility explains the opposite trend in the recovery, however, since the recovery is already high at the design point (96.6%), a further increase is limited by the solvent saturation.

For the sludge-derived HTL gas, the impact of minor impurities (H₂S, N₂) in purity and recovery is addressed in Fig. 9 for a broader range of composition than the evaluated with the scenarios in Fig. 2. The results show that, among the minor impurities considered, the H₂S has a significant impact, while N₂ and NH₃ showed very little effect at the concentrations evaluated.

For the H₂S, both purity and recovery decrease rapidly below specification at much lower concentrations than the evaluated for C₂ + hydrocarbons, showing the highest detrimental impact. This is explained by the relative solubilities compared to CO₂, as H₂S is almost 9 times more soluble in the solvent, while for the C₂ + hydrocarbons present in the HTL gas the ratio is between 0.4 (ethane) and 2.4 (n-butane). The

limit for H₂S in pipeline transportation is 10 to 200 ppmv, however, the H₂S limit for geological storage is 1.5 vol%. In all cases evaluated, which yielded CO₂ content on specification, the H₂S limit for geological storage was never exceeded. In cases where the H₂S concentration needs to be further lowered, alternative Selexol™ configurations with an additional absorber could be implemented, such as the evaluated in Section 3.3.

In terms of N containing components, the negligible impact of the N₂ on the purity is consistent with the fact that nitrogen has approximately 50 times lower solubility in the solvent compared to CO₂ [14], while the increase in recovery observed is due to less CO₂ entering in the feed gas after normalization. For HTL gas with increased NH₃ content, a negative impact in the results can be expected due to its relatively high solubility in the solvent (4.8 times more soluble than CO₂). However, in the range of composition and conditions used, it is effectively removed from the gas in the condensation step prior to absorption, and therefore the impact observed in purity is minor. In all cases evaluated, the limit for N₂ was not exceeded.

3.2.2. Mass and energy balances

Global mass and energy balances for the base case are shown in Fig. 10, and the detailed stream summary can be consulted in the Supplementary material. Fig. 11 shows an overview of the compounds distribution between the gas products along with the HTL gas feed.

The absorption process results in the complete recovery of H₂, CH₄ and CO in the combustible gas, however, its energy potential decreases by 23% due to the loss of C₂₊ hydrocarbons in the CO₂ stream. The H₂ concentration in the combustible gas is about 65 mol. % being in the range of PSA processes for hydrogen recovery typically between 70 and 85 % [28]. The CO₂ product corresponds to an annual storage of approximately 111,000 tonne based on the estimated size of the HTL-facility evaluated. The carbon capture process requires 2.7 MW of electricity and produces 4.1 MW of heat; out of which about 3 MW are available in a temperature range of 50–170 °C. Since there is no heat requirement, heat integration is not applicable, however, cooling duty of 147 kW at 10 °C requires the use of refrigerant.

3.3. Selexol™ process with two-step absorption

The results of the two-step absorption regarding the CO₂ purity and recovery are summarized in Fig. 12 and in Table 5 in comparison with the requirements for pipeline transportation and storage. The process results in a CO₂ product with a purity between 98 and 99 mol % for the woody biomass-derived HTL gas and 95–99 mol % for the sludge-derived HTL gas, fulfilling the minimum CO₂ concentration for geological storage and pipeline transportation in all scenarios. The increase in purity compared to the single-step absorption is mainly due to the reduction in the hydrocarbons content, which are separated in the combustible gas, as discussed further on in the mass balance.

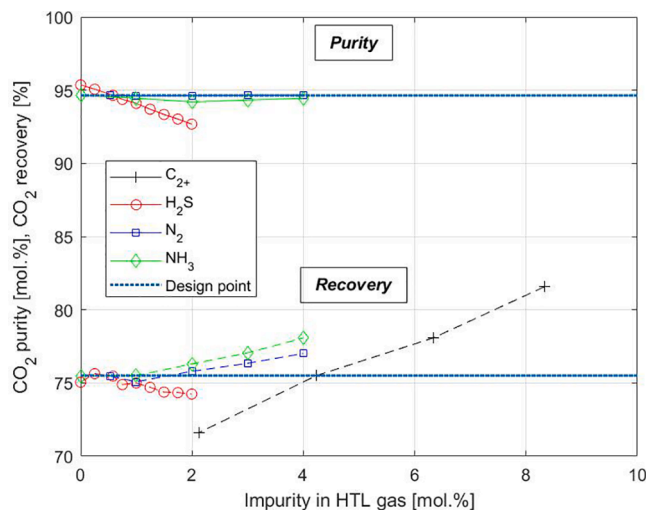


Fig. 9. Impact of varying minor impurities –H₂S, N₂, NH₃– in the HTL gas in purity and recovery of CO₂ product.

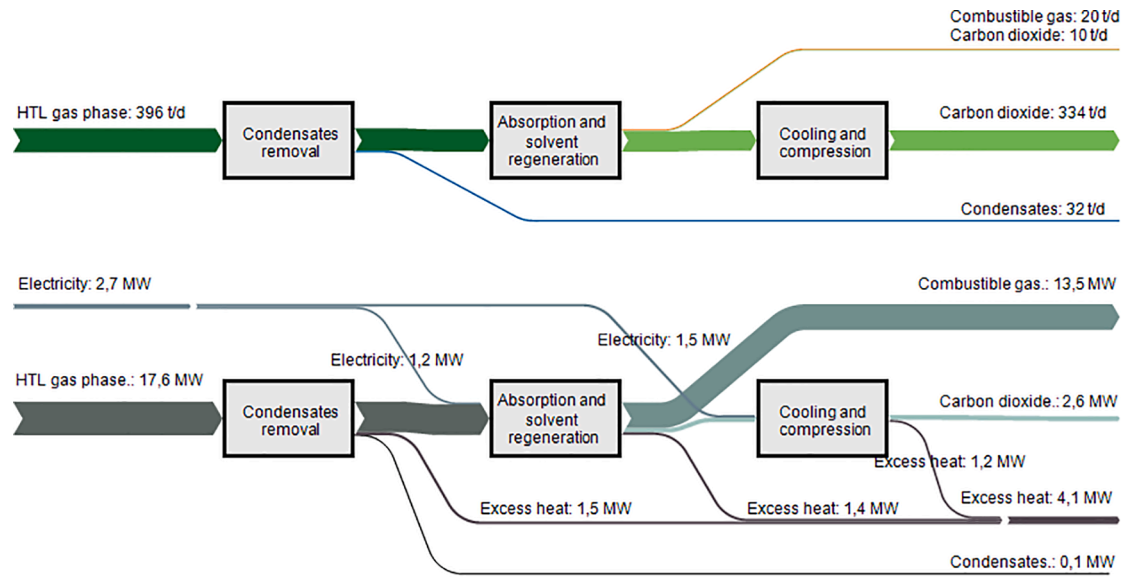


Fig. 10. Mass and energy balance of carbon capture from woody biomass-derived HTL gas in Selexol™ process with single-step absorption configuration.

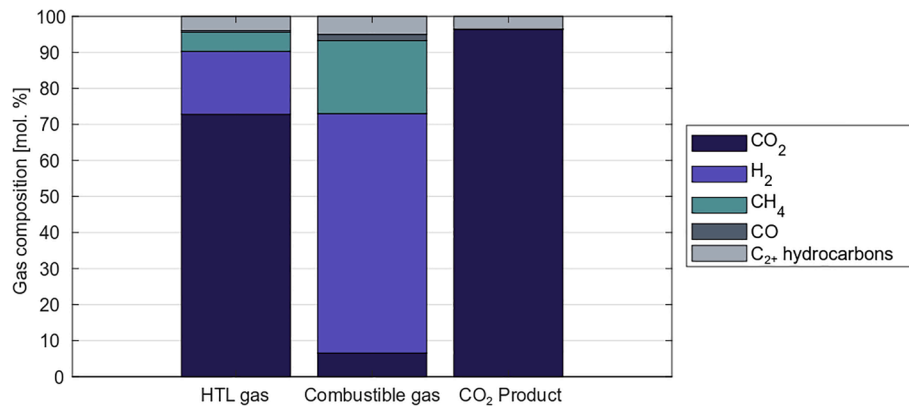


Fig. 11. Composition of gases at inlet (HTL gas) and outlet of the process (Combustible gas and CO₂ product).

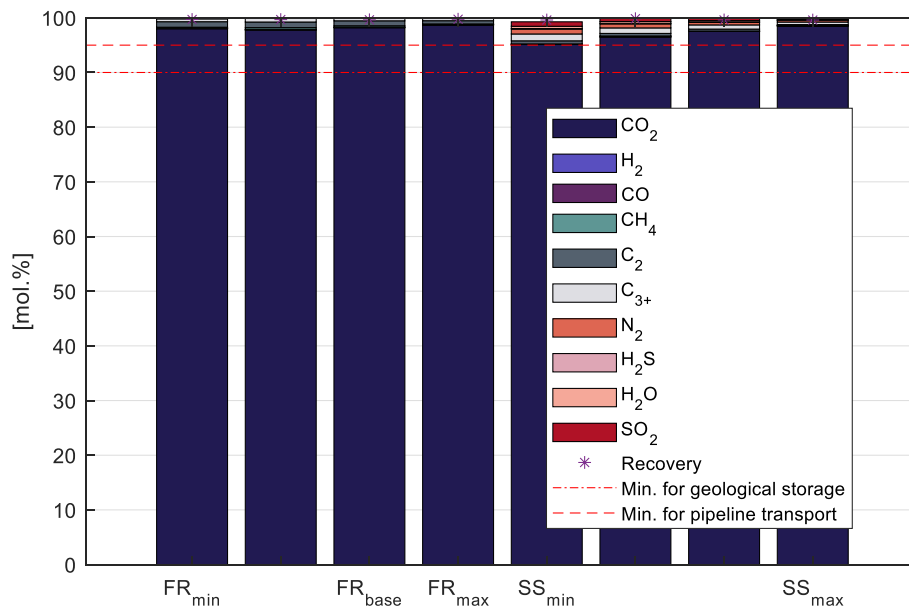


Fig. 12. Modeling results of purity and recovery of CO₂ from HTL gas derived from woody biomass (FR) and sewage sludge (SS) for two-step absorption via Selexol™.

Table 5

Composition of CO₂ product from two-step absorption of HTL gas derived from woody biomass (FR) and sewage sludge (SS) compared to purification limits for transportation and storage.

Component	Pipeline transportation	Geological storage	FR min	FR max	SS min	SS max
CO ₂	95	90	98.00	98.68	95.16	98.54
H ₂	4	4	0.10	0.07	0.00	0.00
Hydrocarbons	5	NR*	1.74	1.16	1.88	0.69
CO	0.2	4	0.00	0.01	0.01	0.00
N ₂	4	4	0.00	0.00	0.88	0.29
H ₂ S	0.02	1.5	0.00	0.00	0.11	0.04
H ₂ O	0.005	0.05	0.16	0.09	0.41	0.15
SO ₂	0.005	0.02	—	—	0.82	0.25

*NR: Not reported.

For the sludge-derived HTL gas a further reduction in the H₂S content compared to the single-step layout is explained by the partial separation of H₂S with the C₂₊ hydrocarbons after solvent regeneration; however, its combustion results in the formation of SO_x (SO₂ assumed in this case) that contaminates the CO₂ product, being a drawback of this implementation. In typical Selexol™ layouts for syngas cleaning, a common approach is processing the separated H₂S for sulphur recovery by means of the Claus process [24,29], which consists in the catalytic reaction of SO₂ and H₂S to produce elemental sulphur. The SO₂ is produced in a first stage from partial H₂S combustion, and modifications of the air-based technology for oxy-fuel operation have been discussed in literature [30] and are commercially available. Alternatively, SO₂ separation by the addition of limestone is used during sewage sludge combustion which results in the formation of CaSO₄ that is removed from the furnace with the fly ash [31]. The evaluation of strategies for sulphur recovery from the HTL gas phase is recommended for future analysis.

3.3.1. Mass and energy balances

Global mass and energy balances are shown in Fig. 13 and the detailed stream summary can be consulted in the [Supplementary material](#). In the first absorption step, 70% of the initial C₂ + hydrocarbons in the HTL gas are separated as combustible gas, and overall, the combustible gas from the two steps accounts for 95% of the initial combustible gases including H₂, CH₄ and CO. Thus, the energy potential of the combustible gas is 84% of the initial HTL gas versus the 77% in the one-step configuration.

Nevertheless, from the mass balance it can be observed that the amount of CO₂ used for solvent saturation is significant and results in a large dilution of the combustible gases prior oxy-fuel combustion, thus not representing an advantage compared to the direct combustion of the HTL gas. The solvent flow and CO₂ loading in the first absorber is set to achieve a large reduction of C₂₊ hydrocarbons in the HTL gas. Fig. 14 shows that for any solvent flow, a higher C₂₊ reduction is achieved at higher CO₂ loading, however, CO₂ migration to the gas occurs at

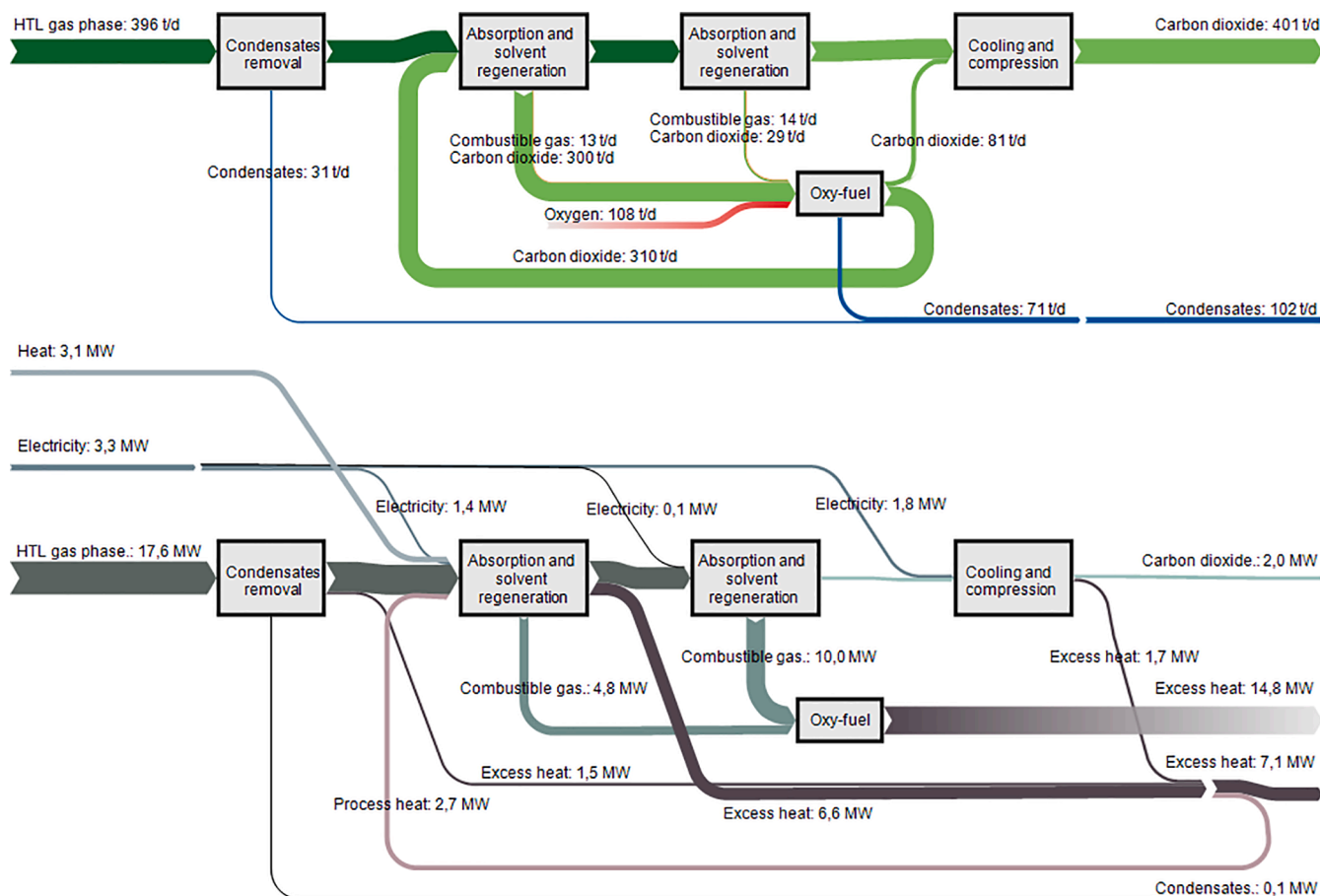


Fig. 13. Mass and energy balance of carbon capture from woody biomass-derived HTL gas in Selexol™ process with two-step absorption configuration.

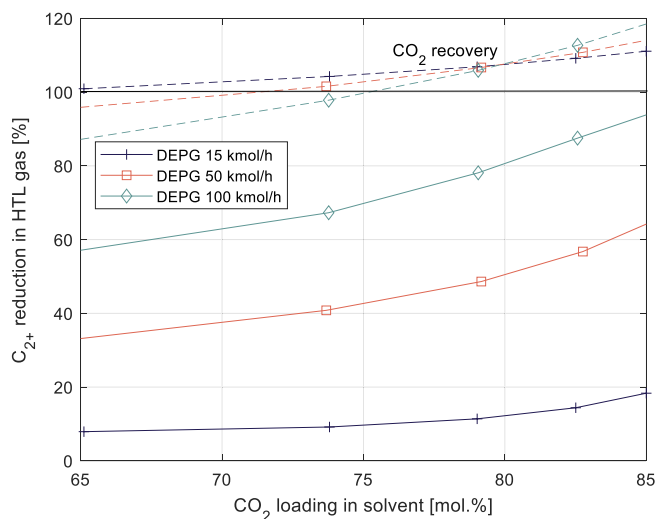


Fig. 14. C_{2+} hydrocarbons reduction in HTL gas after first absorber for different solvent flows and CO_2 loading.

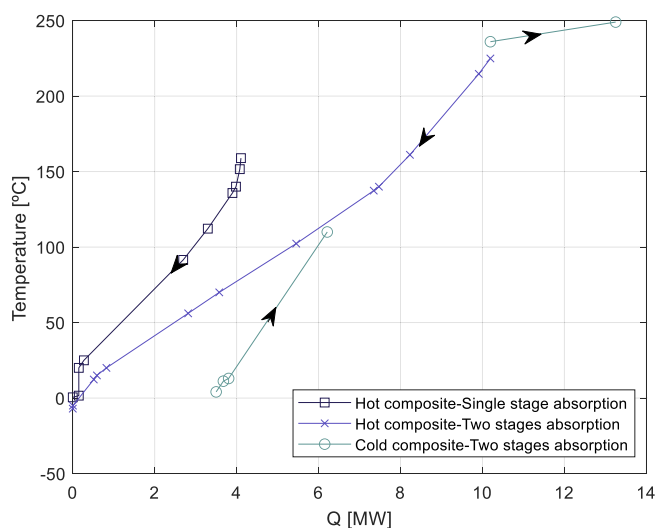


Fig. 15. Composite curves of single-step and two-step absorption configurations.

loadings higher than the saturation. For the design point, a CO_2 loading of 75% and solvent flow of 100 kgmol/h are used to achieve a 70% reduction in the C_{2+} content. Further optimization of this configuration can be done to reduce the recycle of CO_2 or to recycle it directly from the main CO_2 stream, without the oxy-fuel, finding a compromise between purity and recovery.

A CO_2 recovery of 95% is obtained excluding the excess of CO_2 not recycled after combustion, however, if mixed with the main CO_2 stream, the loop is closed yielding a virtual recovery of 100% of the initial CO_2 in the HTL gas. This configuration requires 3.3 MW of electricity, 3.1 MW of external heat, and produces 7.1 MW of excess heat out of which about 4.7 MW are available in a temperature range of 100–225 °C. The composite curves for the two configurations evaluated are presented Fig. 15. For this configuration the requirement of refrigerant is estimated in 580 kW approximately.

3.4. Cost estimation

The results of the cost estimation based on Monte Carlo analysis are shown in Fig. 16. For the single-step Selexol™ configuration, the cost of

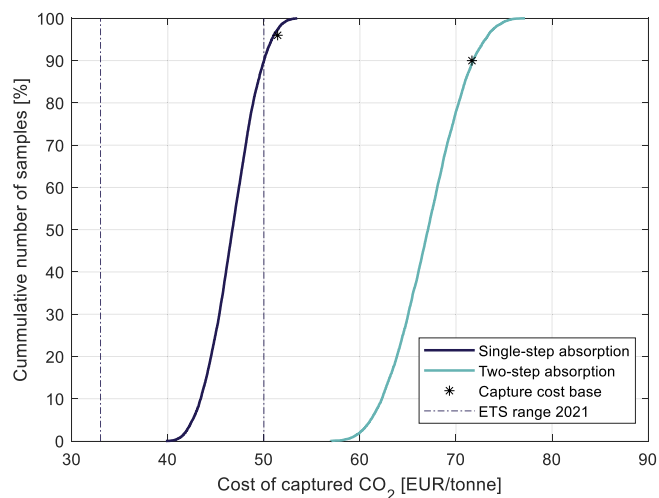


Fig. 16. Cost range of captured CO_2 in single-step and two-step absorption from HTL gas phase.

captured CO_2 is estimated between 40 and 53 EUR/tonne, and for the two-step absorption Selexol™ the result is between 57 and 77 EUR/tonne, representing a substantial increase of 43–45%. For the single-step configuration, the result is in the range of the European carbon market price in 2021 [32]. The different cost contributors are presented in Fig. 16 for the base case and show that the operational expenses are the main item with approximately 2/3 of the total and the remaining 1/3 corresponding to the CAPEX.

Among the operational costs, the electricity and the operation and maintenance costs have the highest shares with approximately 30–35 EUR/tonne in the base case, and the revenues from excess heat can potentially decrease the capture cost by around 10 EUR/tonne. Even though the heat revenues are higher in the two-stage configuration, this does not compensate the additional cost due to the external heat supply and the increase cost in electricity. Thus, the implementation of a second absorption stage is not justified in the base case where the increase in the energy potential of the combustible gas recovered is only 7%.

A more detailed distribution of the CAPEX and OPEX items is shown in Fig. 18 and it shows that the direct costs (equipment purchase and installation) have the major CAPEX contribution with 60% of the total, out of which, the main cost correspond to the compressors (72%).

The result of the single-step absorption is compared in Fig. 19 with the reported for other applications by the Global CCS institute [33], where the bars indicate the lowest and highest values reported depending on the CCS project location, being the lowest in the countries with lower labor and energy costs.

In the present study, it can be expected that relatively high labor and electricity prices used, evidenced in the high shares in the OPEX breakdown (Fig. 17), result in a rather high cost of avoided CO_2 . In fact, if the total operation and maintenance costs are recalculated as 6% of the total plant capital costs, used as a rule of thumb estimate applied in industry that reflect plant utilisation upwards of 85% [33], the cost of avoided CO_2 decreases significantly to 34.6 USD/tonne, below the lowest cost estimated by Monte Carlo in Fig. 15. An approximated cost in the range of 44–60 USD/tonne is in agreement with the reported for BECCS applications between 20 and 175 USD/tonne [34]. The estimated cost is in the range of the current price of the carbon credit in the European market, which registers an increasing trend going from 31 EUR/tonne at the end of 2020 to about 53 EUR/tonne in May 2021; and in the past two years the average price has grown from about 8 to 25 EUR/tonne [35,36]. This increase occurred due to cap reductions in the European Emission Trading Scheme, which are expected to be reduced further by a linear factor of 2.2% between 2020 and 2030, increasing the price of CO_2 -credits and supporting the current trend [37].

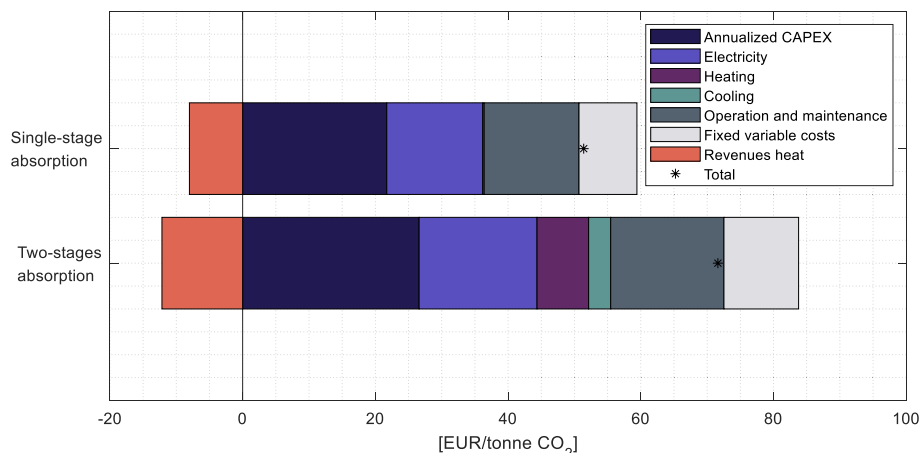


Fig. 17. Cost of captured CO₂ from HTL gas phase for single-stage and two-stage absorption Selexol™.

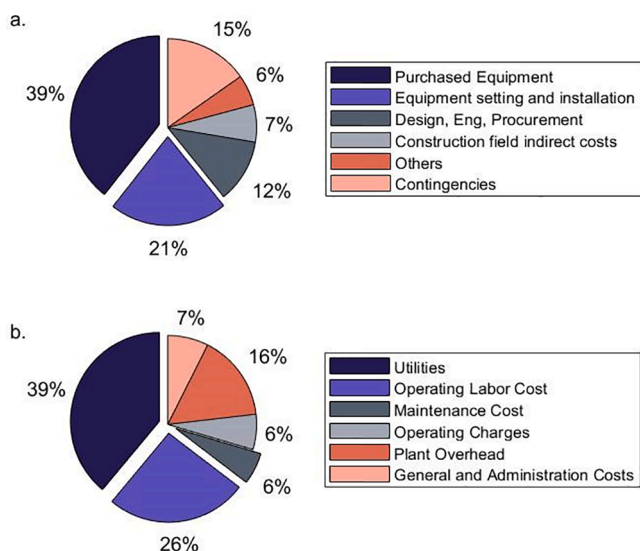


Fig. 18. Breakdown of (a) capital and (b) operational expenses from Aspen Process Economic Analyzer™ results.

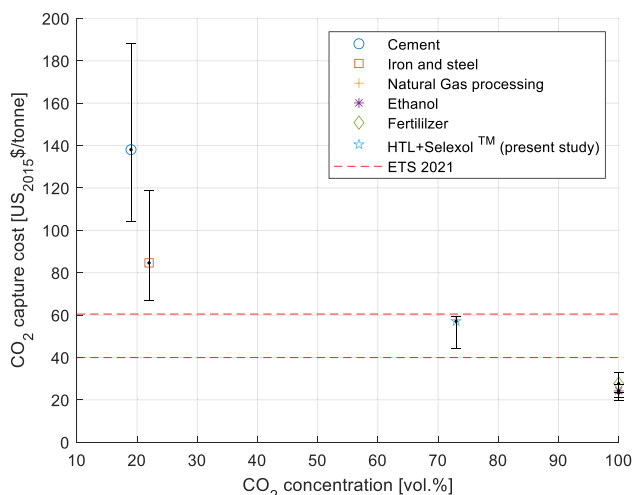


Fig. 19. Estimated cost of captured CO₂ compared to different applications (2015 US\$).

4. Conclusions

The implementation of the Selexol™ process yielded promising results for a cost-effective separation of the carbon dioxide in the HTL gas from lignocellulosic and urban waste biomasses. For the range of compositions evaluated in the feed gas, the purity of the CO₂ product fulfils the requirements for geological storage in all cases (90–98 mol %), and is on-spec for pipeline transportation when the composition of C₂₊ hydrocarbons in the HTL gas is kept below 4–4.5 mol % approximately, depending on the feedstock used. Among minor impurities expected in the urban waste case, H₂S showed the highest negative impact and should be kept below 1 mol % in the feed gas in order to meet the H₂S limit for storage, while N₂ and NH₃ did not have a significant impact in the purity and recovery within the expected concentration evaluated (0–4 mol %). The impact of N₂ is negligible as it is not soluble in the solvent, and NH₃ was effectively separated from the gas by condensation prior to absorption due to its high solubility in water. Still, uncertainty regarding the amount of NH₃ present in the HTL gas can limit the application of the Selexol™ technology if not removed effectively. A modified configuration with two absorption steps was evaluated to circumvent the C₂₊ hydrocarbons co-absorption which results in higher purity product (98–99 mol%), fulfilling the minimum CO₂ requirement for pipeline transportation in all the scenarios considered and reducing the losses in the energy potential of the combustible gas after absorption. This configuration has potential in the scenarios where the hydrocarbons content in the HTL gas phase is above 4.5%; however, a high recirculation of CO₂ is needed in the current scheme requiring further process improvements. As of now, the storage requirements allow more impurities to be present in the CO₂ product compared to the pipeline transportation limits. Thus, matching the storage requirements to future transport requirements can further support the applicability of Selexol™ in combination with the HTL process. However, based on the technical limitations identified, future work should focus in the improvement of the two-step absorption configuration for HTL gas with high hydrocarbons content, or in the evaluation of other capture technologies that harness more effectively the high CO₂ concentration in the HTL gas and are less sensitive to the presence of impurities. In a pre-combustion capture scheme, hydrogen recovery from the HTL gas can be included in the techno-economic analysis for a more comprehensive evaluation of the costs in a stand-alone HTL scheme.

The estimated cost of the captured CO₂ is between 40 and 53 EUR/tonne (44 and 60 USD/tonne) for the standard configuration, being competitive with the prices reported for other applications based on flue gas -between 20 and 100 USD/tonne-. The result is in the range of the carbon price expected within this decade in the EU emission trading system, given the announced cap reductions until 2030 and the targets

set by the European Green Deal. Nevertheless, a more detailed assessment and cost estimation including transportation and storage will depend on the specific location of the plant, and in the long term on the deployment of CCS infrastructure in Europe and expected financial support for the commercialization of NETs, which will be crucial for the realization of this type of applications.

In summary, the results of this study support the technical applicability of the Selexol™ technology for carbon capture in combination with the HTL process when different types of feedstock are used, showing potential for future applications where the utilization of different biomasses is likely to occur for valorization of urban and agricultural residues at a commercial scale. Even though lower availability of urban wastes compared to lignocellulosic biomasses favour a distributed configuration of relatively small HTL plants that would make economically unfeasible to perform carbon capture at small scale, economic incentives on NETs can play in favour of a co-liquefaction scheme. Therefore, the environmental impact of such an application lies not only in the potential for waste valorization that increases the circular economy, but also on the possibility of yielding negative emissions, increasing even further its positive environmental impact.

CRediT authorship contribution statement

E.M. Lozano: Conceptualization, Methodology, Software, Validation, Writing - original draft. **S.B. Petersen:** Conceptualization, Methodology, Software, Validation, Writing - original draft. **M.M. Paulsen:** Conceptualization, Methodology, Software, Validation, Writing - original draft. **L.A. Rosendahl:** Conceptualization, Methodology, Software, Validation, Writing - review & editing, Supervision. **T.H. Pedersen:** Writing - review & editing, Supervision, Project administration, Funding acquisition.

Declaration of Competing Interest

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.ecmx.2021.100089>.

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