Report on

CO_2 Separation, Transportation, and Sequestration

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Abstract

CCS is a well investigated and fairly promising technology for reducing the emission of carbon dioxide (CO_2) to the atmosphere. However, it is rarely implemented in the industry due to its high cost. Therefore, this work proposes a cost optimized CCS chain which can be operated flexibly and safely. For the capture process a post combustion chemical absorption technology is chosen due to its retrofitting possibility to already existing power plants and its low capture cost. In order to find a cost efficient absorption process for different scenarios, the five most promising process configurations from previous work are combined into a superstructure in a rigorous rate based reactive Aspen Plus model. This in turn is optimized by a two-stage stochastic programming approach in Matlab. The optimal supply chain network is identified by a tailor made transshipment model implemented in GAMS, which accounts for the most promising transportation units, storage sites as well as direct utilization locations for one specific power plant in Germany. The optimal capture process contains the following process configurations: absorber intercooling, stripper interheating, cold solvent split (split ratio of 4 %), and vapor recompression. Thereby, the total annualized cost can be decreased from 373 to 361 Mio EUR (3.2 %). The optimal supply chain network suggests to partly utilize the CO_2 (0.85 %) and to store the rest in a depleted gas field, whereas the whole amount of CO_2 is piped to the storage site and then partly transported to the utilization location by trucks. The total annualized cost for transportation and storage are 53 Mio EUR and 4.4 Mio EUR, whereas the utilization generates a revenue of 4.2 Mio EUR per year. This results in an overall CSS cost of 414.2 Mio EUR per year.

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Contents

1 Introduction

Carbon dioxide is a major anthropogenic greenhouse gas that causes severe environmental effects such as global warming. Thus, the global CO_2 emissions need to be reduced by 41-72 % by 2050 [1] with respect to 2010 levels. There are different approaches to achieve this aim, among which carbon capture and storage (CCS) is capable to reduce the CO_2 emissions from large point emission sources, e.g., power plants. These CO_2 sources account for about 70 % of the worldwide large CO_2 emission sources [1].

CCS consists of CO_2 separation, its transportation, and long-term isolation from the atmosphere [1]. In order to develop an economically feasible process and evaluate it in terms of safety and operability, this work addresses the CCS chain in its entirety. Special attention is given to the optimization of the capture process, because it accounts for about 75% of the complete CCS process [2]. There are different capture technologies, i.e. pre-combustion, oxyfuel combustion, chemical looping, and post-combustion. Post-combustion is advantageous in terms of flexibility because it enables the implementation as a retrofit option to existing plants. Additionally, the technology has already been proven at large scale. [3, 4, 5]. In post-combustion, several technologies can be used for the carbon dioxide separation, whereas absorption is a promising and mature technology [6]. However, low solvents' loading capacity, degradation, and in particular high regeneration energy causes high cost [5]. Thus, several process configurations have been published to improve process performance [7, 8]. In most previous research, these configurations were considered separately. Therefore, the influences between different configurations were neglected. In order to account for these influences, superstructure-based optimization is a potential tool capable to cover all the different process configurations simultaneously and to find an optimum [9, 10].

Several publishers worked on superstructure optimization in the carbon capture context recently [9, 11, 12, 13]. Damartzis et al. [9] presented a superstructure optimization considering five solvents, including monoethanolamine, and five process configurations, including a conventional process, absorber intercooling, multi feed absorber, and double section stripper. The objective function is the process capital and operating expense. A rigorous model was claimed, but not described in further

detail, e.g., in terms of model complexity. Javaloyes-Anton et al. [14] used Aspen Plus coupled with Matlab to optimize tray-by-tray based distillation processes with a particles swarm optimization algorithm implemented in Matlab. These works lack either on transparency or on model complexity. Furthermore, they optimize the processes for a certain flue gas specification. Thus, these processes may perform poorly under changing circumstances.
This work combines a two-stage stochastic programming formulation [15] with a superstructure approach [10]. Therefore, the resulting process is able to operate economically feasible under a large CO_2 concentration range. The proposed optimization problem considers rigorous rate based models, a detailed economic objective function, and 14 optimization variables. The process superstructure, including five process configurations, is implemented in Aspen Plus and the resulting optimization problem is solved using a genetic algorithm in Matlab [16, 17, 18]. Besides the optimization of the capture process, the selection of suitable transportation routes, storage locations, and possible utilization poses a challenging problem. In order to find the optimal supply chain for the captured CO_2, a tailor-made transshipment model is proposed, implemented in GAMS and globally solved. The transshipment model includes pipeline and truck transportation as well as different candidates for storage locations and direct utilization. Both, the capture process and the supply chain are optimized for a specific coal-fired power plant located in Lippendorf, Germany. Hence, this work presents a complete CCS process which can be operated economical feasible under realistic circumstances. Ultimately, safety concerns, environmental, and social impacts of the complete process are discussed.
The report is subdivided into six sections. The second section treats the carbon capture process, which is followed by an evaluation of transportation, storage and utilization opportunities in section 3. In Section 4, an economic evaluation of the overall CCS chain is realized. This is followed by a discussion about process safety and environmental impacts in section 5. Ultimately, Section 6 summarizes the report.

2 Capture Process

In this section, existing carbon capture technologies are presented and the most promising technology is selected. Then, different process configurations are included in a superstructure. The superstructure formulation is combined with a two-stage stochastic programming approach and the resulting optimization problem is solved using a genetic algorithm.

2.1 Technologies

The performance of the capture technologies highly depends on the CO_2 concentration of the flue gas [19]. Because over 80 % of the worldwide emitted CO_2 is released by power plants [1], this report focuses on a capture process for power plants which have CO_2 concentrations of 3-14 vol% [19].
In the pre-combustion technology, the fuel is pretreated before combustion. By means of a reforming process natural gas is converted into a mixture of CO and H_2. Afterwards, the resulting syngas undergoes a water gas shift reaction to increase the H_2 concentration while the CO gas is converted into CO_2. The produced H_2 can then be used to fire a gas turbine [20]. Similarly to the water gas shift, coal could be converted in a gasification process. The main advantage of this technology is the high concentration of CO_2 in the flue gas, which leverages the CO_2 separation.
In oxyfuel-combustion, pure O_2 instead of air is used for the combustion. Therefore, the amount of nitrogen in the process is reduced drastically and the formation of thermal NO_x is suppressed. Thus, the remaining flue gas contains 80-98 % CO_2 [6] and can directly be compressed, transported, and stored [6]. The great disadvantage is the energy intensive upstream air separation [20, 21].

In chemical looping, a metal is used as oxygen carrier. The metal oxidizes with oxygen from air. Then, it is reduced to metal while fuel is being oxidized. Finally, the metal is recycled [6].
In post-combustion, the CO_2 separation takes place after the combustion. This technology can easily be retrofitted to existing power plants [5]. Furthermore, post-combustion processes are flexible because they can efficiently treat flue gases from different sources, i.e. coal-fired power plants and natural gas power plants. Therefore, this technology is discussed in more detail subsequently.
In post-combustion, several separation technologies, e.g. adsorption, absorption, or membranes exist [1]. Adsorption is the phenomenon that involves the adhesion of molecules to a solid surface. To capture CO_2, activated carbon, alumina, metallic oxides, and zeolites [22] could be applied as adsorbents. These can be regenerated by pressure swing or temperature swing adsorption [5]. Because current adsorption systems may not be suitable for application in large-scale power plants, this mechanism is not considered in detail. Membrane-based separation uses thin polymeric films as membranes, to separate the CO_2. The driving force is a gradient in the partial pressure of CO_2, whereby this separation is preferred at high CO_2 concentrations in the flue gas [23]. The absorption mechanism can be divided into physical and chemical absorption. While the first one is only based on Henry's law [20], latter also involves reactions between the CO_2 and a solvent [5]. As physical absorption takes place at high CO_2 partial pressures the process has a high energy demand for the flue gas pressurization. Therefore, physical absorption is not economical for flue gas streams with CO_2 partial pressures lower than 15 vol% [24]. The weak bounds of the chemical absorption can be regenerated by introducing heat to the rich solvent [1]. As the selectivity of some solvents is relatively high, this leads to a high purity CO_2 stream. Thus, chemical absorption is well suited for flue gases from power plants. In addition, Hasan et al. [19] have shown, that within typical CO_2 concentrations for applications for power plants, absorption is the most cost effective choice for capture. Thus, the absorption technology is the selected capture technology for this work. The solvent used for chemical absorption has a major influence on the process performance. In recent literature, different solvents such as ammonia and monoethanolamine (MEA) are screened and compared [25]. In this work, MEA is chosen as solvent because it is a mature technology which has already been proven at large scale [5], and this work aims the proposal of a specific technical feasible CCS process.

Conventional CO_2 capture process

In this section, the main characteristics of the conventional absorption-based capture process are presented. Afterwards, possible process configurations are introduced. As shown in Figure 1, the basic absorption process consists of an absorber, a stripper, and several heat exchangers. As the desorption in the stripper is endothermic, a high stripper temperature leverages solvent capacity. However, the temperature is limited due to solvent degradation at 120 C in the bottom of the column [26]. Vice verse, the absorber operating temperature needs to be low in order to keep the working capacity of the solvent high. Both, stripper and absorber are commonly operated at about $1.5 - 2$ atm [1]. On the left hand side, the precooled flue

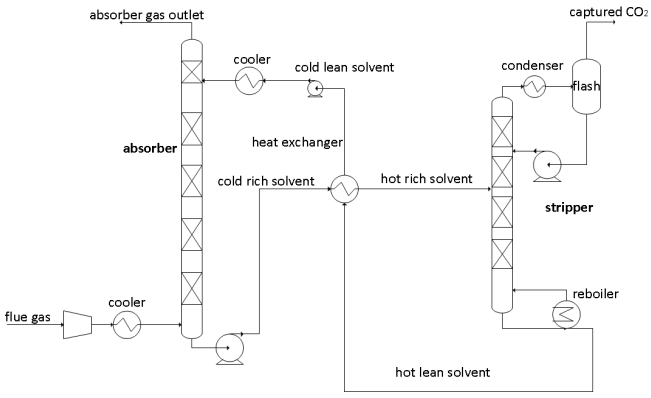

Figure 1: Conventional MEA-based absorption process [7].

gas enters the absorber bottom and is contacted counter currently with the MEA-based solvent that absorbs the CO_2. Then, the scrubbed gas is water washed and vented to the atmosphere. The cold rich solvent leaves the absorber at the bottom. Afterwards, it is heated in a heat exchanger in cross flow with the hot lean solvent coming from the stripper. The cold rich solvent enters the stripper where it is separated in a CO_2 and a lean solvent stream. The high purity CO_2 stream leaves the stripper at the top. The hot lean solvent leaves the stripper at the bottom, is cooled down, and returned to the absorber.

Promising process alternatives

The main disadvantage of the conventional process is the high reboiler duty for solvent regeneration. Furthermore, low solvents' loading capacity and degradation may cause increasing cost. In order to overcome these disadvantages, various process configurations have been published. Based on comprehensive literature reviews [7, 8], we select the most promising configurations: interheating, intercooling, lean vapour recompression, cold solvent split, and stripper overhead compression. These are discussed in detail hereinafter. *Absorber intercooling* is used to increase the solvent working capacity, which leads to a reduction of the amount of circulating solvent [7]. As a result, the reboiler duty can be reduced by 11.6 % compared to the conventional cases [7]. The *stripper interheating* targets to optimize the temperature profile in the stripper. Therefore, a semi-lean solvent stream at the middle of the column is extracted, reheated, and injected into the stripper again [27]. In the *lean vapor recompression*, the lean solvent stream is flashed under low pressure. The gas phase exiting the flash is recompressed and injected back to the stripper. Thereby, the steam requirement can by reduced by 11 % [7]. In the *cold solvent split* configuration, the cold rich solvent stream is split into two parts. One, which is preheated and a second, which is kept cold. The cold stream enters the stripper at the top, while the preheated stream is injected at the middle as in the conventional case. Due to the additional cooling at the top, the reflux ratio and condenser duty of the stripper can be reduced [8]. This leads to a decrease of the reboiler duty of about 10 % [28]. *Stripper overhead compression* uses the latent heat of the water saturated CO_2-rich flue gas, which is released during multi stage compression after the stripper with compression ratios of 2-5 for each stage. Additionally, water is injected between compressors to receive a water saturated stream. The heat resulting from compression can be recovered by generating additional steam for the stripper reboiler, which gives rise to the reduction in the steam requirement from the external steam cycle. Woodhouse et al. [29] claimed that the steam requirement can be reduced by 24 %. Ahn et al. [7] published simulation results around 32 % reduction in reboiler heat duty. All process configurations are included in one superstructure, which is illustrated in Figure 2.
There exist more process configurations than these which have been considered in this work. *Multi pressure stripping* follows the idea to separate the stripper into different sections operated at different pressures. Thus, the vapor flow going up is compressed and the liquid flowing down is expanded and flashed. Therefore, additional steam is supplied and the reboiler duty is reduced [7]. However, simulation studies have shown that the negative effect of additional auxiliary power outweighs the reduction in reboiler duty [30]. Another configuration is the *split-amine flow*. In contrast to the *cold solvent split*, the rich amine flow is split after the preheater and a semi-lean amine stream flows back from the stripper to the absorber. Kun Bae et al. [31] and Oi et al. [32] have found that this configuration has a great impact on the reboiler duty for high CO_2 concentrations of about 40 % but not for the scenarios considered in this work. Also the process configurations *condensate heating* and *condensate evacuation and evaporation* aim a reduction in heat duty [7]. Here, hot distillate from the stripper is fed to the bottom in order to reduce the amount of necessary steam. However, Ahn et. al have found that it does not lead to a significant heat duty reduction. Due to the shortcomings of these process

configurations they are not included in this works' superstructure.

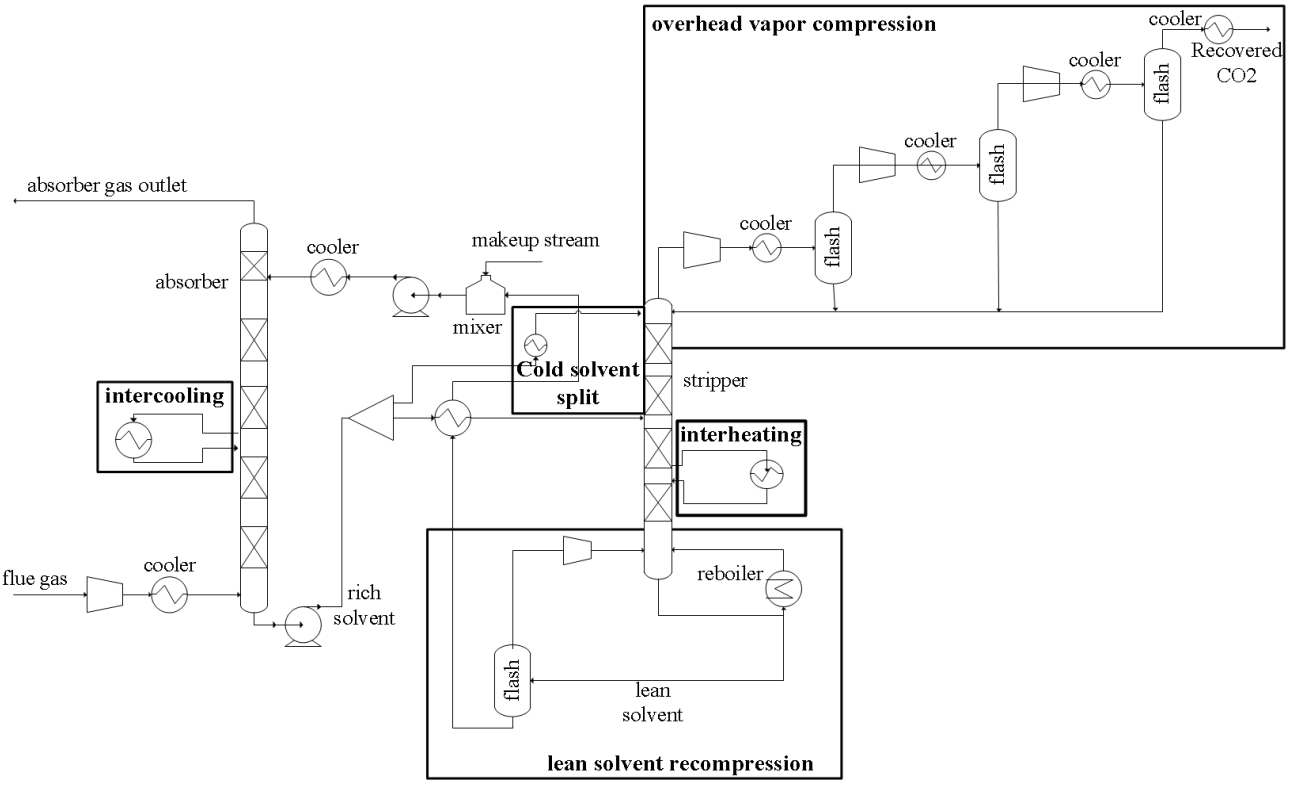

Figure 2: CO_2 capture process flowsheet with all configuration included.

2.2 Superstructure optimization

The process options mentioned in the section above were implemented in one flowsheet in Aspen Plus, where the use of splitters and mixers gives rise to the possibility of by-passing certain configurations. This way the superstructure is realized.

For the simulation of the CO_2 capture process by aqueous MEA, the rate-based reactive distillation model RadFrac of Aspen Plus was used [33]. The model includes N_2, O_2, H_2O, and CO_2 as components of the flue gas and MEA as the solvent entering the process. Corresponding ions are considered in order to account for the complex reaction system described by Austgen et al. [34]. The equilibrium constants for the equilibrium based reactions are adopted from this work and from the Aspen Plus database. The kinetic parameters for the rate controlled reactions are adopted from Pinsent et al. [35] and Hikita et al. [36]. The Electrolyte NRTL activity model and Redlich-Kwong equation of state were used for the calculation of the activity and fugacity coefficients, respectively.

In previous work, the model was fitted to experimental data of a pilot plant by adjustment of the interfacial area factors [8]. Jung et al. showed that the simulation matches the experimental data well. The interfacial area factors of the absorber and the stripper are set to 2.0 and 1.4, respectively.

The superstructure optimization approach [10] yields a process optimized for a certain scenario. This, however, can lead to infeasible or non-flexible performance under different circumstances. To make CCS competitively feasible in todays' volatile markets, the capture process has to offer great flexibility. In order to ensure that, different scenarios are considered simultaneously in this work. Similarly to a two-stage stochastic programming approach [15], we distinguish between design variables, y, and operating variables, x_s. The main idea of this approach is that the design variables are the same for all scenarios while the process can adapt to different scenarios by changing the operating variables. The objective function of the optimization problem minimizes the expected total annualized cost, TAC,

of the process under different scenarios. The TAC are composed of annualized investment cost, AIC, and averaged annualized operating cost, AOC_s, of all scenarios, s.

$$min\ TAC = AIC(y) + \sum_s \frac{1}{n} \cdot AOC_s(y, x_s). \tag{1}$$

Herein, the AIC mainly depend on design variables y. The AOC_s depend on both, the design variables of the process and operating variables of the respective scenario. The cost of the process are treated following the procedure in Hasna et al. [19]. The AIC comprises of the total plant cost (TPC) and the annual maintenance cost (AMC):

$$AIC = \phi TPC + AMC, \tag{2}$$

with the capital recovery factor $\phi = 0.154$ and

$$TPC = TIC + IDC + BPC, \tag{3}$$

whereas TIC is the total installed cost, IDC is the indirect cost, and BPC is the balance of plant cost. The TIC includes the equipment purchase cost (EPC) and the equipment installation cost (EIC). The EPC includes detailed sizing and costing of all process equipment where heat exchangers, compressors, flashes, and pumps are evaluated using methods by Biegler [37]. The absorber and the stripper are evaluated using the method by Hasan et al. [19]. The EIC for mover equipment is 80 % of the EPC and 4% of the EPC for the other equipment (column, exchanger, etc.). The IDC is considered to be 32 % of TIC and the BPC is 20 % of TIC. The AMC is assumed to be 5 % of the TPC.

Table 1: Optimization Variables.

Variable name	**bounds**
Design variables, y	
OHC_{on}	0; 1
LVR_{on}	0; 1
FS_{LVR}	29; 39
FS_{CCS}	4; 17
CR_1	[1.5; 5]
CR_2	[1.5; 5]
SF_{CCS}	[0; 0.2]
Operating variables, x_s	
$\dot{Q}_{intercooling,1}$	[-150 MW; 0 MW]
$\dot{Q}_{intercooling,2}$	[-150 MW; 0 MW]
$\dot{Q}_{intercooling,3}$	[-150 MW; 0 MW]
$\dot{Q}_{interheating,1}$	[0 MW; 150 MW]
$\dot{Q}_{interheating,2}$	[0 MW; 150 MW]

The optimization variables and its respective bounds are summarized in Table 1. The first two design variables, OHC_{on} and LVR_{on}, are binary. They switch the *overhead compression* and the *lean vapor recompression* configurations on and off, respectively. The variables FS_{LVR} and FS_{CSS} set the feed stage of the *lean vapor recompression* reflux stream and the *cold solvent split*. These integer variables are chosen based on literature recommendations [7] and restrictions due to the strippers' column packing. The compressor ratios of the three-stage compressor have a major influence on the heat recovered in the overhead compression and on the electricity demand of the process. As the overall compression ratio is fixed by output specifications, compression ratio one, CR_1, and two, CR_2, remain as design variables. The maximum compression ratio of five is obtained from literature [37]. The split fraction of the *cold solvent split*, SF_{CSS}, is a continuous variable. Previous work states that it usually ranges from 15-20 % [8]. In order to allow the complete by-pass of the configuration, the bounds are set to 0 and 20 %. The operating variables of the process are the interheating and intercooling duties of the stripper and absorber, respectively. In the absorber we allow intercooling on stages 28, 57, and 85. The interheating is included on stages 4 and 17. The stages of the heating/cooling as well as their bounds are selected based on

literature recommendations [7] and restrictions for account of the column packing.

Solution algorithm

The solution of the optimization problem is in particular challenging since the model complexity of the simulation is large and the evaluation of the objective is therefore expensive. This problem is even more challenging due to the stochastic programming formulation which requires s solutions of the process model for one evaluation of the objective function. Furthermore, deterministic solvers cannot be used to solve the problem because of the use of the process simulator. Thus, a stochastic solution algorithm, namely the genetic algorithm of Matlab (GA), is used to solve the problem [16, 17, 18]. Therefore, an interface between Aspen Plus and Matlab was implemented allowing the GA to access Aspen. In Figure 3 the solution algorithm is presented. The algorithm starts with the initialization of the GA by a random population. Then, Matlab passes the variables to Aspen Plus, which in turn runs the simulations for all scenarios sequentially. The results are given back to Matlab, where it is checked whether the model converged. If not, a penalty term is generated. Otherwise, the TAC is calculated according to equation 1. The GA receives the objective value and checks termination criteria. In case the criteria is fulfilled, it terminates. If not, the GA creates a new generation by its three characteristic steps: selection, mutation, and recombination.

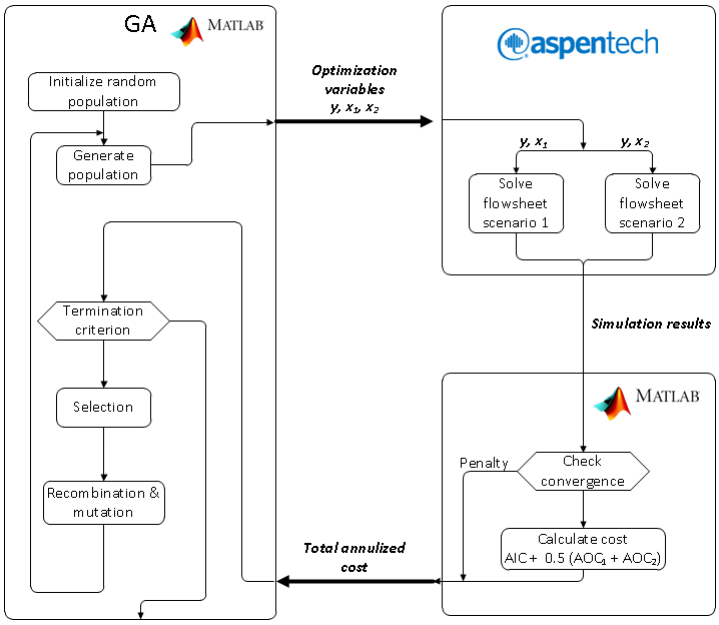

Figure 3: The solution algorithm combines a genetic algorithm in Matlab with the process simulator ASPEN Plus.

2.3 Results

For this work, two different CO_2 scenarios are taken into consideration. Scenario one represents a coal-fired power plant located in Lippendorf, Germany, [38] and scenario two represents a typical flue gas specification of a natural gas power plant [19]. The flue gas specifications of the scenarios are summarized in Table 2. In order to include the two scenarios in a comparable manner, the capture rates are kept constant at 90 % and the purities of the CO_2 product streams are kept at 99.8 mol%. Furthermore, the total flow of CO_2 which is captured is set to 336 kg/sec for both scenarios.

Table 2: Different CO_2 capture scenarios. Flue gas composition in vol% for the two scenarios after Hasan et al. [19].

	scenario 1	**scenario 2**
CO_2	14	8.6
N_2	73	71
O_2	3.7	5.4
H_2O	9.3	15

Having set those specifications in Aspen Plus, the solution algorithm converges within one week to a solution. The proposed solution for a flexible process is presented in Figure 4. The optimal design variables of the process are shown in Table 3. The solution includes *lean vapour recompression*, *inter-*

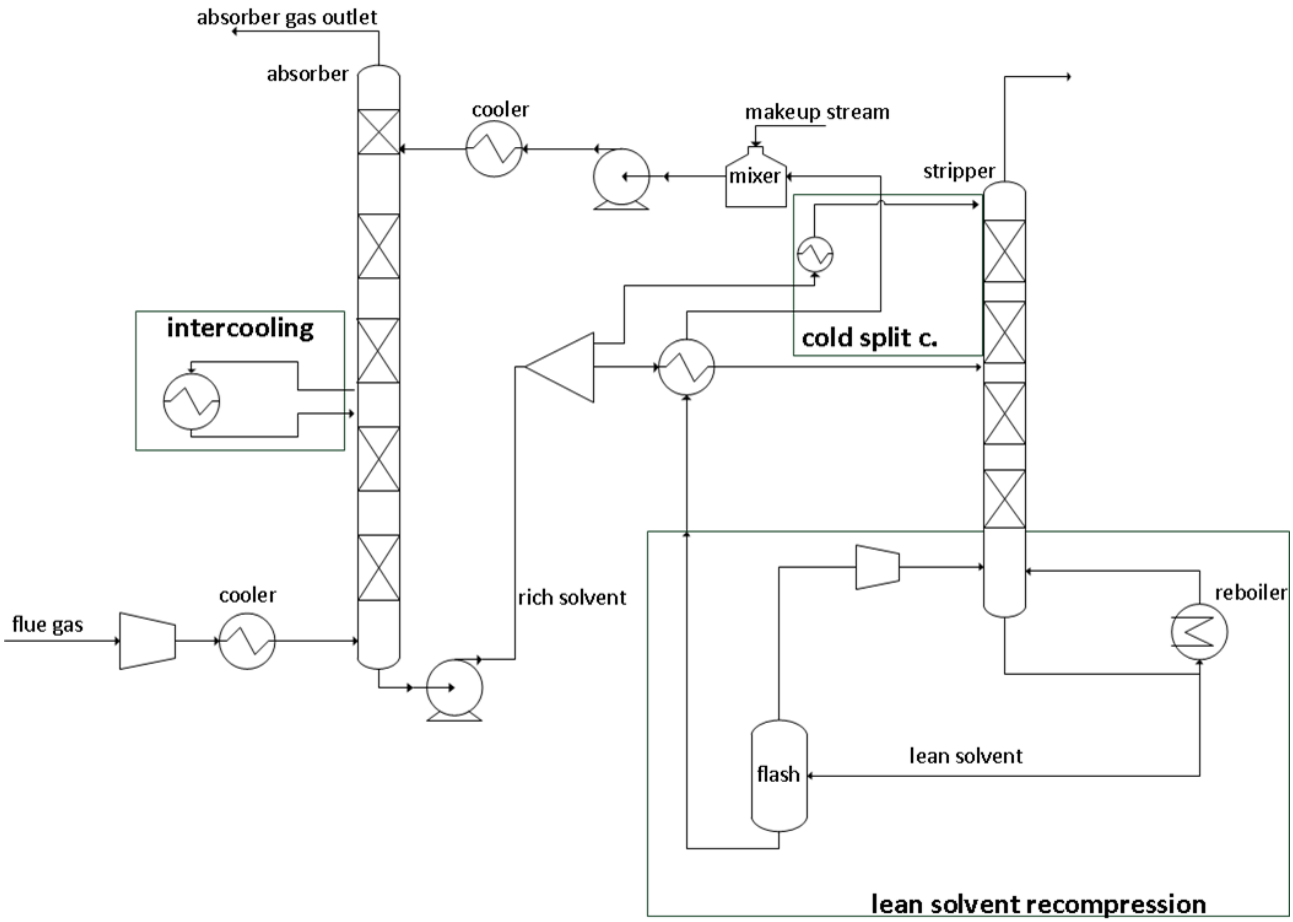

Figure 4: The optimized, flexible process.

Table 3: The optimal design variables of the flexible process.

Variable name	**optimal solution**	
Design variables, y		
OHC_{on}	0	
LVR_{on}	1	
FS_{LVR}	39	
FS_{CCS}	17	
CR_1	2.15	
CR_2	4.20	
SF_{CCS}	4 %	
Operating variables, x_s		
Scenario	**1**	**2**
$\dot{Q}_{intercooling,1}$	-25	-25
$\dot{Q}_{intercooling,2}$	-15	-18
$\dot{Q}_{intercooling,3}$	-23	-90
$\dot{Q}_{interheating,1}$	67	101
$\dot{Q}_{interheating,2}$	18	130

cooling of the absorber, *interheating* of the stripper, and the *cold solvent split*. The split ratio of the lean vapor recompression is 4 %. In the *lean vapor recompression*, the re-injected vapour phase enters the stripper at the bottom on stage 39. The interheating and intercooling duties vary between the given bounds and depend on the scenarios. The compression ratios of the three-stage compressor are 2.15, 4.20, and 3.19 respectively. Besides the optimal variables, the steady state simulation of the flexible process yields the mass and energy balances of both scenarios as well. Most important for further cost evaluation of the process are the energy demands of the process. These are summarized in Table 4 together with the cost of the conventional process optimized for a coal-fired power plant. Evidence is given that the regeneration energy of the flexible process is lower for both scenarios compared to the conventional one. This is in particular due to the *lean vapor recompression* configuration which supports additional steam to the bottom of the stripper and thus, reduces the reboiler requirement. As the regeneration energy decreases, the steam demand of the flexible process decreases because the reboiler accounts most of the total steam requirement. In contrast to the steam, the electricity demand of the flexible process is higher than the one of the conventional configuration for both scenarios. This increase arises primarily due to the additional compressor in the *lean vapor recompression*. However, the saving of steam requirement compensates for the increase of electricity. Ultimately, the amount of cooling water required by the process is lower in the flexible process which is due to the decreased reflux ratio. Considering all utility requirements, it it obvious that the flexible process can be operated under lower operating expenses. However, the additional configurations include extra purchase cost, indirect expenses, and maintenance. Thus, only the assessment of expected economic performance can demonstrate the benefits of the proposed solution in front of the conventional process optimized for a the coal-fired power plant. This economic evaluation is provided in Table 5. Herein, the total annualized operating cost of the processes are calculated for both scenarios using the above mentioned utility consumptions. It is important to note that the TAC include the average of the operating expenses for both scenarios. Thus, it is a valid metric for a fair comparisons of the process flexibility. The economic analysis shows clearly that the flexible process has 4 Mio EUR higher investment cost but 16 or 17 Mio EUR lower operating expenses for scenario 1 and 2, respectively. The lower operating cost compensate for the investment and lead to a total annualized saving of 12 Mio EUR for flexible operation. The total capture cost for capturing one tonne CO_2 is 36.22 EUR for the proposed process. This refers 3 % saving compared to the conventional process.

Table 4: Energy requirements of the flexible capture process.

Indicator	**Conventional**		**Flexible process**	
	scenario 1	**scenario 2**	**scenario 1**	**scenario 2**
reboiler duty [MW]	1,299	1,296	975	1,018
reboiler duty [MJ/kg CO_2]	3.86	3.86	2.90	3.03
steam [Mton/year]	18	18	14	14
electricity [MW]	111	114	129	133
cooling water [Mton/year]	2,203	2,431	1,775	2,068

Table 5: Economic performance of the flexible process compared to the conventional. The AOC are considered for both scenarios (scenario 1//scenario 2). The TAC are the sum of the AIC and the average of both AOCs.

Indicator	**conventional**	**flexible process**
AIC [Mio EUR]	193	197
AOC [Mio EUR]	179//181	163//164
TAC [Mio EUR]	373	361
Cost per CO_2 captured [EUR/ton]	37.43	36.22

3 Transportation, storage, and utilization

After CO_2 has been captured, it needs to be transported to an utilization or storage location where it is further treated. Thus, this section discusses possible transport facilities, utilization options as well as storage sites. A supply chain model is proposed which combines the most promising options. Afterward, the model is solved for the supply chain of the power plant in Lippendorf and the results are stated.

3.1 Technologies

CO_2 transportation

In order to transport CO_2 economical efficiently, it needs to be present at low volume [1]. Thus, converting the gaseous state of CO_2 into a supercritical state is an expensive but necessary treatment. Typical operating pressures at ambient temperature vary from 50 bar for transportation via ship to 150 bar for pipeline transportation [23]. Furthermore, a high CO_2 concentration of more than 95.5 % is favorable, so that corrosion and the formation of hydrate is prevented [39].
Common transportation facilities are pipelines, trucks, and ships. CO_2 transportation by pipeline is the most mature one. There exist more than 6000 km CO_2 pipelines in North America, which proves safe operation of this technology [39]. Since CO_2 emissions of large scale power plants are in the magnitude of 10 Mt per year, the high working capacity of pipeline transportation is advantageous. However, the investment cost of a large pipeline network are commonly high and the installation of pipes in Germany is challenging due to the dense infrastructure [39]. The negative aspect of high investment cost for pipeline transportation can be overcome with transportation by trucks, since infrastructure investment cost can be neglected [40]. Thus, it can be applied flexibly. However, the small working capacity of trucks makes truck transportation only economically feasible for small amounts. This may be worthwhile for the supply of customers with a small demand. In this work, transportation via ships is not considered, because ocean storage does not reflect an appropriate storage facility for the proposed case study located in Germany.

Carbon utilization

There are manifold forms of CO_2 utilization: it can used directly, e.g., in the food and beverage industry or in enhanced oil recovery and indirectly for conversion into chemicals, fuels, and others [41, 42]. Caellar-Franca et al. [41] showed in their life cycle analysis, the convenient CO_2 utilization options have a significant higher global warming potential compared to CO_2 storage. Since CCS aims the mitigation of global warming and since utilization obviously can not fulfill this aim currently, indirect utilization is not considered in this report. CO_2 could be used directly to offset the cost for capturing [42]. However, this requires high purity of CO_2 for most applications, and hence can be doubt to be suitable for many industries [41]. Caellar-Franca et al. [41] mention, that a detailed life cycle assessment (LCA) is necessary for carbon utilization and give several recommendations for future research, including the consideration of a wider range of LCA impacts from carbon capture and storage/utilization (CCS/CCU), and further studies of various utilization possibilities, particularly the production of chemicals and the comparisons of environmental impacts CCS and CCU with other options, such as renewable energy.

CO_2 Storage

In general there are three ways of storing large amounts of CO_2: ocean storage, mineral carbonation, and geological storage. Each of them needs to meet two basic requirements: First, it needs to guarantee long-term isolation from the atmosphere; and second, it needs to be economically feasible. Previous literature has shown that ocean storage cannot guarantee long-term isolation from the atmosphere since the CO_2-isolation via CO_2 seas on the ground of the ocean or the dissolution of CO_2 in depths of 500 m does not guaranty a reliable trapping [1]. Mineral carbonation is not a suitable option as its cost are roughly 100 times higher than those of geological storage [1]. For these reasons, geological storage as the remaining option is discussed in further detail.
Geological storage can mainly be categorized in three technologies: storage in gas/oil fields, saline aquifers, and in coal beds. The differences between these storage sites arise from their different ability to trap the CO_2 reliably, from the availability of data, and from economical aspects. Although the ability of storing CO_2 in rock formations of coal beds is very high, this ability strongly dependents on environmental circumstances [39]. Furthermore, the relatively low permeability of coal beds does not allow a sufficiently high injection rate [43] and due to a lack of specific data about the coal seams, it is difficult to estimate its capacities. For saline formations, the lack of detailed data is even bigger [44]. However, they are known to be the most widespread sites [39]. Issues of the storage in saline aquifers come along with conflicts concerning the alternative usage as geothermal [45], missing opportunities for retrofitting existing infrastructure [45], and possible water replacement within the seams [46]. Nevertheless, the latter problem has not been observed in an existing plant "Sleipner" in Norway and the other shortcomings do not prohibit safe and economic usage [39]. Thus, aquifers are still a promising technology in certain regions. Geological storage in depleted oil/gas fields is a mature and proven technology which ensures long-term isolation of CO_2. It is the only technology, which reliable trapping mechanism for hydrocarbons is proven. This eliminates the strongest doubts of CCS [47]. Besides the availability of detailed data for the characterization of the field, another positive aspect is that the process of injecting the highly concentrated CO_2 is well established and retrofitting is possible. This reduces total cost to 0.5-8 EUR per tonne CO_2 injected and makes the process economically attractive [47, 39]. The only drawback of geological storage is its small capacity compared to saline aquifers.

3.2 Supply chain optimization

The selection of an optimal supply chain for certain locations poses a challenging problem as various candidate storage locations, utilization possibilities, and transportation facilities exist. This problem is intensified by a large number of possible transportation networks. In order to find the optimal supply chain network and storage locations, a non-linear programming problem (NLP) is proposed in this section. The proposed problem accounts not only for candidate storage locations but also for direct utilization locations with associated demands. The objective of the model is to minimize the overall cost of the supply chain which is given by:

$$\begin{aligned} &min\ [overall\ CCUS\ cost] \\ &= [transportation\ cost]\ +\ [storage\ cost]\ -\ [utilization\ revenue] \end{aligned}$$

In the supply chain, CO_2 can either be transported by pipelines or by trucks. The pipeline transportation cost,$TC_{pipeline,j,k}$, is given by its operating cost and levelized investment cost. The cost function

is fitted to published data and given by [48]

$$TC_{pipeline,j,k} = (CCR + OM_{pipe})(C_{base,pipe}(\frac{M_{pipe,j,k}}{M_{base,pipeline}})^{\eta})(L_{j,k} \cdot 10^3 \cdot (\frac{L_{j,k}}{L_{base}})) \tag{4}$$

The pipeline transportation cost are a function of the mass flow of CO_2 transported, $M_{pipe,j,k}$, and the length of the pipe, $L_{j,k}$. The remaining cost parameters are given in the supplementary material. The truck transportation cost,$TC_{truck,j,k}$, is assumed to has zero investment cost and linear operating cost.

$$TC_{truck,j,k} = \alpha \cdot \frac{M_{truck,j,k}}{M_{base,truck}} \cdot L_{j,k} \tag{5}$$

where $M_{truck,j,k}$ represents the mass of CO_2 per time transported by truck and $L_{j,k}$ the transportation distance. The associated cost coefficient, α, is estimated considering the capacity and cost of commonly used LNG trucks. The cost for storage of CO_2 is approximated by the injection cost,SC_j, function by Odgen [49].

$$SC_j = (CCR + OM_{injection})(m_1 \cdot d_{well,j} + m_2) \cdot n_{well,j} \tag{6}$$

The cost is a function of the depth of the well, $d_{well,j}$, and the number of wells needed, $n_{well,j}$. The latter one is necessary because every well has maximum storage capacity, $C_{max,stored,j}$. Ultimately, the revenue from utilization,UR_j, is earned by selling the CO_2 for direct utilization such as in the beverage industry. Each utilization location has therefore a certain maximum demand, $C_{max,utilized,j}$ and purchase cost, $R_{revenue,j}$.

$$UR_j = R_{revenue,j} \cdot C_{utilized,j} \tag{7}$$

In order to formulate the extended transshipment model, the mass balance of CO_2 at each location, j, is required. The balance is given as follows

$$\sum_j (M_{pipe,j,k} + M_{truck,j,k} - M_{pipe,k,j} - M_{truck,k,j}) = C_j \tag{8}$$

where $M_{pipe,j,k}$ and $M_{truck,j,k}$ are the mass flow transported from location j to location k by pipeline and truck respectively. Depending on the location, C_j is either the mass flow of CO_2 stored, sold, or produced. Combining the aforementioned cost functions, a non-linear optimization problem is formulated (NLP) where the cost parameters are given in the supplementary material. A major drawback of the proposed formulation is that the derivative of the concave pipeline cost function at zero mass flow through the pipe is unbounded. This causes failures in the Karush-Kuhn-Tucker conditions of the problem and makes the application of deterministic solvers hard [50]. However, as the size of the proposed problem is small, it is reasonable to use BARON in order to find the global optimum in this case [51]. The problem was implemented and solved in GAMS [52].

$$min \sum_{j,k}(TC_{pipeline,j,k} + TC_{truck,j,k}) + \sum_{j}(SC_j - UR_j)$$

subject to

Cost functions

$$\begin{aligned} TC_{pipeline,j,k} &= f(M_{pipe,j,k}, L_{j,k}) \\ TC_{truck,j,k} &= f(M_{truck,j,k}, L_{j,k}) \\ SC_j &= f(n_{well,j}, d_{well,j}) \\ n_{well,j} &= \frac{C_{stored,j}}{C_{max,j}} \\ UR_j &= f(C_{utilized,j}) \end{aligned}$$

Constrains

$$\sum_{j}(M_{pipe,j,k} + M_{truck,j,k} - M_{pipe,k,j} - M_{truck,k,j}) = C_j$$

$$\begin{aligned} M_{pipe,j,k} &\geq 0 \\ M_{truck,j,k} &\geq 0 \\ C_{stored,j} &\leq C_{max,stored,j} \\ C_{utilized,j} &\leq C_{max,utilized,j} \\ C_{stored,j=6} &= -11,800,000 \end{aligned}$$

3.3 Results

The supply chain model was implemented using the actual geographical location and CO_2 output specification of the power plant located in Lippendorf, Germany. Furthermore, gas fields and saline aquifers, in Solling and Detfurth respectively, were included as candidate storage locations in the model [53]. Moreover, three different locations for CO_2 utilization are included in the model. Here, real data from local industry which has a demand of CO_2 is used.

The solver converged to the global optimum in less than 10 seconds. The optimal supply chain is depicted in Figure 5 where active facilities are depicted in black and inactive ones in gray. In this supply chain, the complete CO_2 stream, 10 Mton per year, is transported by a 230 km pipeline to the nearest available gas field in Solling. Then, the major amount of CO_2, 9.9 Mton per year, is injected in the gas field. The remaining CO_2 is transported by trucks to utilization locations: 0.06 Mton per year to industry located near Frankfurt am Main and 0.04 Mton per year to industry located in Gerolstein. It is important to note that the

Figure 5: The optimal supply chain for the coal-fired power plant in Lippendorf.

amount of CO_2 which is utilized is limited by the maximum demand of the industry. Besides the supply chain infrastructure, the model yields detailed cost. These are shown in Table 6. One can see that the pipeline transportation accounts for about 96 % of the complete transportation and storage cost. These high cost arise from the large distance of the power plant to the depleted gas field. However, the gas field was chosen because the injection cost are in particular low as the existing infrastructure can be retrofitted. In contrast, retrofitting is not possible at considered saline aquifer locations. Therefore, the injection cost account for only 0.8 % of the total cost. Ultimately, the use of trucks allows direct utilization of CO_2. This reduces the cost by 8 %.

4 Economic evaluation of the complete CCS

This section presents the economic performance of the complete CCS are presented. By doing so, economic indicators are stated which allow a fair comparison of the proposed process in front of others. As illustrated in Table 7, the cost of the CCS chain consist of capture, transportation, and storage cost. The cost for capturing one tonne CO_2 are 36.2 EUR. In good agreement with literature, these account for about 87.2 % of the total cost. The transport cost are 5.3 EUR per tonne CO_2 captured. As the CO_2 is stored in a depleted gas field, the cost are only 0.4 EUR per tonne CO_2 captured. Utilization is limited by the demand of the customers. Therefore, the total cost can be reduced by 0.4 EUR per CO_2 captured. In total the CCS chain costs 41.5 EUR per tonne CO_2 captured. Therefore, this technology is only competitively

Table 6: Detailed cost of the optimal CO_2 supply chain.

Name	**annualized cost** million EUR
Pipeline (Lippendorf -> Solling)	51.1
Truck *(Solling -> Frankfurt am Main)*	1.7
Truck *(Solling -> Gerolstein)*	0.2
Injection *(Solling)*	4.4
Revenue from utilization *(Frankfurt am Main)*	-2.4
Revenue from utilization *(Gerolstein)*	-1.8
Total cost	53.2

Table 7: Cost of overall CCS chain in EUR.

Process	**Cost per tonne CO_2 captured**	**Relative to total cost**
Capture	36.2	87.2 %
Transport	5.3	12.8 %
Storage	0.4	1.0 %
Utilization	-0.4	-1.0 %
Total	41.5	100 %

feasible, if CO_2 taxes are at least 41.5 EUR per tonne CO_2. The current political situation in Germany does not permit this conditions, but e.g., Sweden has launched a legal CO_2 regulation in 1991. The tax rate was increased in 2014 to 1076 Skr/ton CO_2, which corresponds to 115 EUR/ton CO_2. Nevertheless, Sweden was able to increase the gross national product as well, which shows, that Carbon Capture and Storage is economically feasible.

5 Safety and environmental impact

A key component for the design of a generic CCS process is the inherent risk for humans and environment and the resulting acceptance for CCS in the society [54]. Therefore, this paragraph states and evaluates possible hazards and thereby show that the chosen design is safe. CO_2 capture and compression are common industrial operations which are regarded to fulfill industrial safety standards [54]. The major risk of CO_2 piping is the release of CO_2 caused by damaged pipes [54]. In surroundings where the gas can accumulate, it can seriously harm the whole environment including humans due to anoxia [55]. A concentration of 20 % can lead to death in less than 30 minutes [55]. Therefore, a monitoring system for the pipeline is necessary. Nevertheless, the risk of CO_2 piping is regarded as low and smaller than the risk of natural gas piping [54]. Thereby, the main attention needs to be directed to the storage process. In this project, the captured CO_2 is supposed to be stored in a depleted natural gas field. These fields have proved their ability of trapping natural gas for millions of years without mentionable leakage [54]. On top of that, abandoned gas fields were studied in detail so that various information are accessible. This helps to predict the sites' reaction on CO_2 injection, which might be in the worst case earth tremors, ground movement, and displacement of brine [54]. The major risk regarding CO_2 storage in depleted gas fields is CO_2 leakage through damaged wells [54]. Therefore, detailed information about „the number, type, and age of wells, completion technique and type of material used“ [54] are essential . This in turn provides the possibility to choose an appropriate storage site and to arrange maintenance work if necessary. Additionally, monitoring systems need to be installed to detect possible leaks [54]. Regarding these procedures a safe CO_2 storage can be realized [54], which allows to conclude that the proposed CCS chain is safe.

6 Conclusion

Carbon capture and storage is one possible solution for the mitigation of the global climate change. Yet industrial applications of CCS are still very limited due to its high cost. The goal of this project was to propose a safe, effective, and especially cost competitive CCS process which is able to be operated flexibly under different circumstances. This report proposes a specific process design for the whole CCS chain consisting of a post combustion carbon capture process, a transportation network, storage, and direct utilization facilities. The optimal capture process was found using superstructure optimization which includes five process configurations. The process was simulated using rigorous models and optimized with respect to total annualized cost for different CO_2 sources: a coal-fired and a gas-fired power plant. The transport and storage facilities were identified by optimizing an extended transshipment model. A case study for a specific coal-fired power plant located in Lippendorf, Germany, shows that the overall CCS cost are 41.5 EUR per tonne CO_2. The optimal capture process includes *lean vapor recompression*, *split amine flow*, *absorber intercooling*, and *stripper interheating*. This way, the capture cost could be reduced by 12 Mio EUR per year compared to the conventional process. The transportation and storage process design yields a supply chain in which the 10 Mton CO_2 are transported through a 230 km pipeline to a depleted gas field. There 99 % of the CO_2 are stored. The remaining CO_2 is transported by trucks to industry for direct utilization. The utilization revenue reduces the cost by 0.4 EUR per tonne CO_2. In addition, the report shows that the proposed CCS process is safe. There are still some things future work should focus on. Since the solvent strongly influences the process performance, different solvents should be considered in the superstructure optimization. Additionally, a more detailed cost calculation for transportation and storage could be realized in order to account for more detailed sizing of the injection and transportation.

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