Technical, policy and stakeholder analysis of direct air capture

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ABSTRACT

Direct air capture (DAC) can remove CO_2 directly from the atmosphere and therefore can be used to reduce atmospheric concentrations of CO_2. Despite this, there is little research which carries out a detailed comparison of these systems. This is also true for research which analyses the effect policies regarding climate change are having on DAC development. Furthermore, there is no available analysis on potential stakeholders for DAC companies. These factors have contributed to a lack of widespread implementation of DAC. By answering the following research question the negative effect these elements are having on DAC development can be minimised.

"How do the most promising available DAC systems described in literature compare, where does DAC fit in current policy and how can DAC companies best position themselves to maximise the potential of their technology?"

To answer this question a set of criteria are used and categorised as contributing to different objectives which can help DAC development. This can be used to identify technology and policy combinations which are likely to be successful. Following this a stakeholder analysis is carried out from the point of view of DAC companies. This is used to identify where there is dependency between DAC companies and other stakeholders to achieve their objective.

Through this analysis it was additionally found that combining an amine carbon capture system which captures CO_2 from flue gas with a humidity swing DAC system which captures CO_2 from the atmosphere that an extra 8% of CO_2 can be captured than with the amine carbon captures system alone. In this system the humidity swing DAC system uses the waste heat from the amine carbon capture system as a heat source. This means that there are no extra thermal energy requirements to capture this extra CO_2. This type of system can be used in the energy production sector where energy is produced from natural gas or coal and therefore reduce emissions from this sector.

By using a policy which passes the cost of this system onto the consumer by promising a minimum price for electricity it, generally called a contract for differences, it reduces burden on public finances and therefore be an attractive option for governments.

The implications for this study are that DAC companies and energy suppliers from natural gas or coal with an amine carbon capture system should explore potential collaboration. This is because it has been shown in this research that technically these systems can be combined to reduce CO_2 emissions with minimal increase in operating cost.

Caskie, A.B.R. (Alexander)
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Chapter 1: Introduction

This chapter consists of the background information related to climate change, Direct Air Capture (DAC), and the socio-technological background in which DAC can fit in. Further to this an outline of the knowledge gap and research question is given.

1.1 Anthropological climate change

It has been shown that limiting the increase in the Earth's average temperature to 1.5 degrees Celsius or less, this would minimise the effect of climate change on human and biological ecosystems (Hoegh-Guldberg, Jacob, & Taylor, 2018). These changes include rises in sea level, reduction in Arctic ice levels and an increase in extreme weather events, and would result in a reduction in food and water security (Hoegh-Guldberg, Jacob, & Taylor, 2018). Achieving the goals outlined in the Paris agreement, an undertaking to pursue measures which limit temperature rise to well below 2 degrees Celsius from industrial levels (UNFCCC 2016), this will limit the risks posed by climate change.

The Intergovernmental Panel on Climate Change (IPCC) supplies scientific information regarding human-caused climate change. To limit the temperature rise to well below 2 degrees Celsius a number of scenarios have been produced by the IPCC (IPCC 2018). Ideally the IPCC has advocated to limiting a temperature of no more than 1.5 degrees Celsius. All of the scenarios which achieve this limit employ Carbon Dioxide Removal technologies (CDR). This is where more CO_2 is removed from the atmosphere than added plus traditional mitigation technologies; this is depicted in figure 1: Pathway to negative emissions (Hunt, 2018). Negative emissions have occurred if the following criteria have been met: physical greenhouse gases are removed from the atmosphere; the removed gases are stored out of the atmosphere in a manner intended to be permanent; upstream and downstream greenhouse gas emissions associated with the removal and storage process, such as biomass origin, energy use, gas fate, and co-product fate, are comprehensively estimated and included in the emission balance; and the total quantity of atmospheric greenhouse gases removed and permanently stored is greater than the total quantity of greenhouse gases emitted to the atmosphere (Ramirez and Tanzer 2019). More CO_2 will be required to be removed from the atmosphere than is emitted is because it is expected that the 1.5 degree target is going to be exceeded.

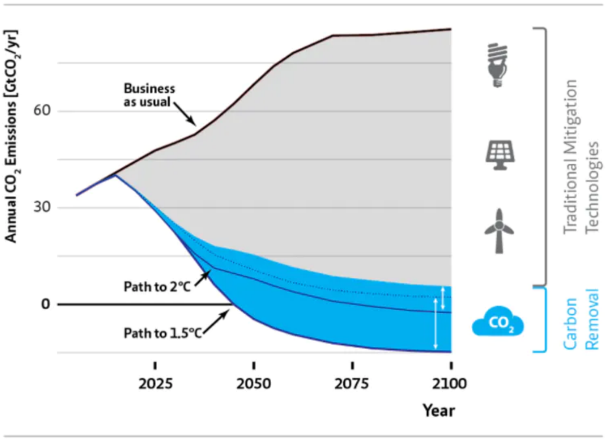


Figure 1: Pathway to negative emissions (Hunt 2018).

CDR options remove CO_2 from the atmosphere and if they meet the criteria described by Ramirez and Tanzer (2019), they can be deemed to be a negative emission technology. Examples of CDR technologies are afforestation, carbon fixing to soils, bioenergy production with carbon capture, enhanced weathering, direct air capture and storage (DACS) and ocean fertilisation (European Academies Science Advisory Council 2018). For each option there is a differing amount of available literature and analysis (IPCC-Summary for policy makers 2018).

A comparison of these CDR processes is outlined in table 1: CDR processes comparison (European Academies Science Advisory Council 2018).

Technology	Afforestation	Land management	Bio-energy with carbon capture and storage (BECCS)	Enhanced weathering	DACS	Ocean fertilisation
$\Delta \boldsymbol{CO_{2,process}}$ $[\boldsymbol{Gt-CO2/year}]$	1.1-3.3	2-3	3.3	1	3.3+	<1
Cost* *[$/t-CO$_2$]	<100	<100	100-400	100-400	100-400+	0-400

Table 1: CDR processes comparison (European Academies Science Advisory Council 2018).

Box 1: Description of DAC and DACS:

DAC, an umbrella term for processes which remove CO_2 chemically and directly from the atmosphere. DACS is where this CO_2 is permanently stored underground, this is marked in green in figure 2: CO_2 captured via DAC supply chain (Olfe-Kräutlein, et al. 2017). The captured CO_2 can be used as a feedstock for other processes and this is marked as the utilisation pathway in figure 2: CO_2 captured via DAC supply chain (Olfe-Kräutlein, et al. 2017). DAC has a number of technical uncertainties (Hills, et al. 2017) and those that have been deployed have a high cost. This has blocked widespread implementation of the technology. CO_2 utilisation options, apart from direct use, also have a number of technical uncertainties (Olfe-Kräutlein, et al. 2017), though utilisation is not considered in this thesis. CO_2 underground storage has been identified as well-developed and deployable (Royal society of chemistry 2018).

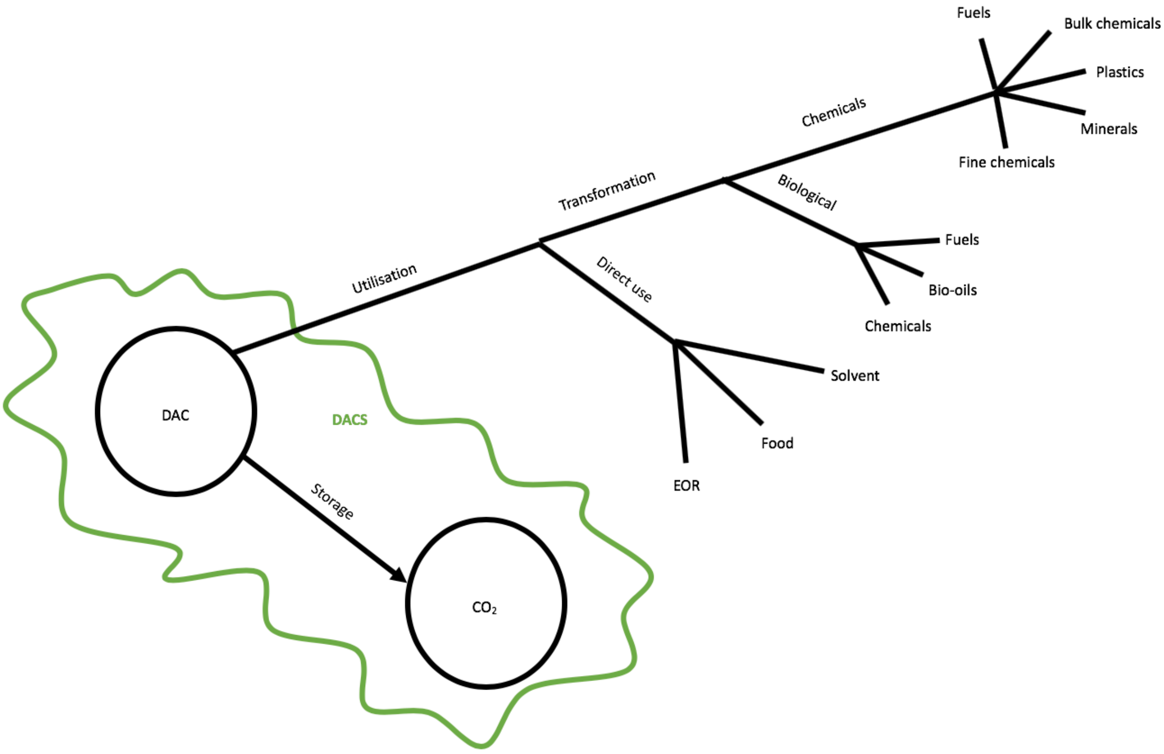


Figure 2: CO_2 captured via DAC supply chain (Olfe-Kräutlein, et al. 2017)

1.2 Why hasn't DACS been implemented?

There is technical uncertainty which results in the large cost range seen in table 1: CDR processes comparison (European Academies Science Advisory Council, 2018). There are also issues regarding climate change solutions in a wider context which are described here.

Governments' collaboration in the IPCC context has been described as pluralistic (Biermann 2004). This is due to the authority that each government has, which comes from groups with differing rules, norms and belief systems. This type of interaction has been identified as a contributing factor to a concept coined a "wicked problem" (Rittel en Webber 1973). This is defined as situations where stakeholders cannot come up with a problem definition and/or solution which they can agree upon (Rittel en Webber 1973).

In the context of climate change the IPCC has defined the problem in a scientific, evidence-based manner and has developed a number of technical solutions. The support shown by almost all countries for the Paris Agreement to limit global temperatures to well below 2 degrees Celsius shows that there is a consensus as regards to the IPCC findings. Despite this, there is a lack of inertia in translating the solutions outlined by the IPCC into action. Examples include the United Kingdom which is set to miss its targets for 2023 (Committee on Climate Change 2019). This can be seen across the whole of Europe with all member states being unable to meet targets given to them by the EU (Climate Action Network Europe 2018). As such, a lack of meaningful implementation on climate change policies, and therefore CDR processes, comes from countries' inabilities to agree on a solution. This means that this can be described as a "wicked problem" (Rittel en Webber 1973), which stems from these groups having differing rules, norms and belief systems.

This disconnect between problem and implementation of suitable solutions can be further explained with the Multi-Level Perspective (MLP). MLP was first introduced to describe lock-in to social technological systems (Rip 1995). This details the development of such systems in terms of external factors, preferences, alternatives, pressures and interaction with new novelties (Loorbach, Frantzeskaki en Avelino 2017). Development can be described as an increase in the organisation of the socio-technical structure over time. This starts outside the current socio-technical construct with ideas based on visions of niche groups. Technical development is created through linkages of these niches. This state is maintained until a window of opportunity is created through interaction between socio-technical regime spawning cohesion of the system. This creates pressure on the current sociotechnical system which creates a new regime; figure 3: The multiphase transition of socio-technical systems (Loorbach, Frantzeskaki, & Avelino, 2017), describes this phenomenon.

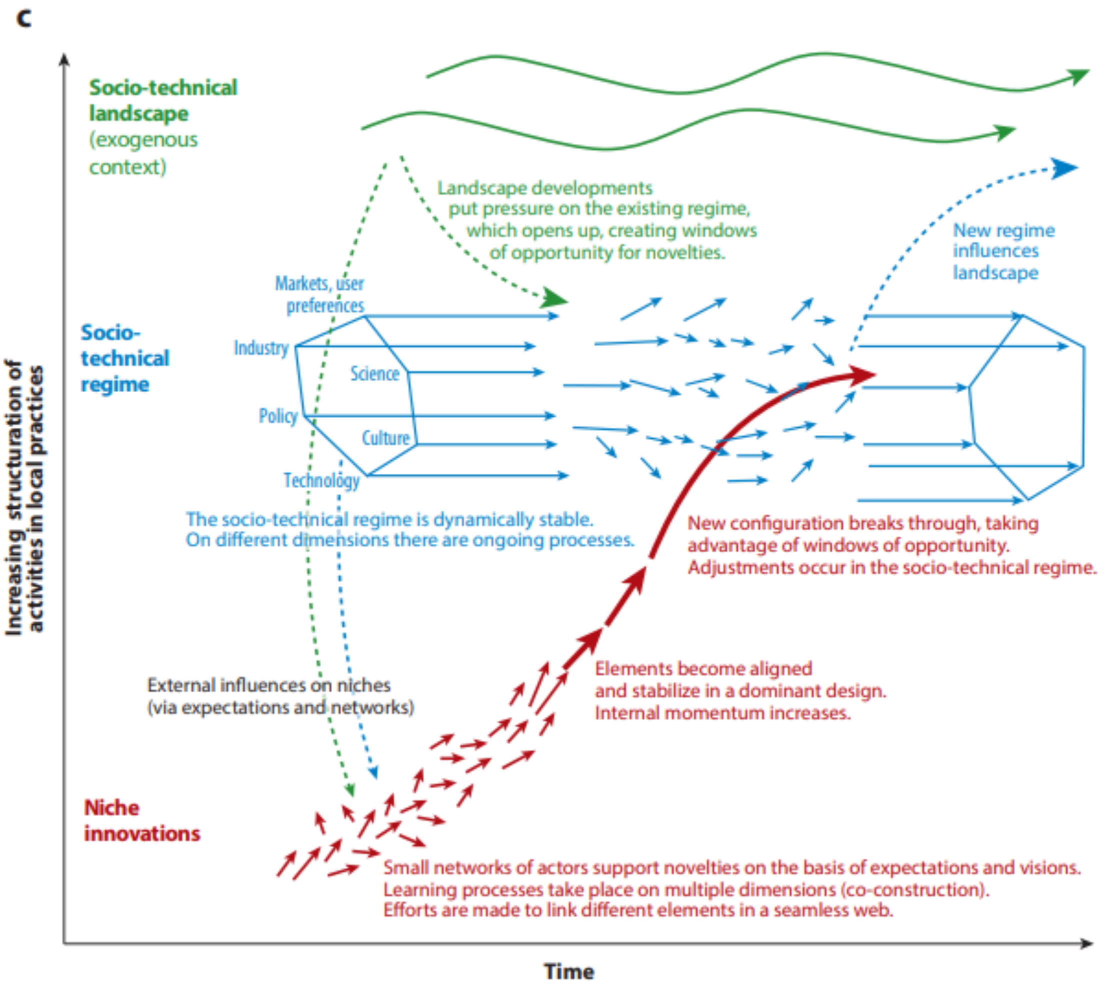


Figure 3: The multiphase transition of socio-technical systems (Loorbach, Frantzeskaki en Avelino 2017).

Separate groups are working on developing the technical aspects of DAC. Prominent groups in DAC include companies such as Climeworks (Climeworks 2017) and Carbon Engineering (Carbon Engineering 2019). A number of technical niches exist for DAC. None of these options have been able to achieve to widespread implementation into the socio-technical system at a level which can have a meaningful effect on climate change. Further development requires disturbance by internal tensions within the current system, an external crisis or a better alternative (Loorbach, Frantzeskaki en Avelino 2017). This better alternative can be developed by reducing the uncertainty that exists in DAC technology. This reduction in uncertainty can be compounded by analysing the effect policy can have on DAC development and the stakeholder interaction.

1.2.1 Scientific & technological component of socio-technical regime

A workshop involving different stakeholders and experts related to DAC took place to explore DAC's potential and its limitations. This was organised by the National Academies of Sciences, Engineering: Committee on Developing a Research Agenda for Carbon Dioxide Removal and Reliable Sequestration, it took place in Washington, DC in 2018 (The National Academies of Science, Engineering and Medicine 2018). At the workshop, G. Holmes from the DAC company Carbon Engineering identified DAC technologies as technically possible though they require validation to be able to attract investment. Furthermore, there is little literature which compares different DAC systems against a set of standardised criteria which investors can use. Considering figure 3: The multiphase transition of socio-technical systems (Loorbach, Frantzeskaki en Avelino 2017), this limitation relates to the scientific and technological components of the socio-technical regime to examine the current technical descriptions of DAC to validate, or compare, these systems. This can then contribute to the required internal tension of the regime to transition to a new socio-technical.

There are three main technological DAC systems used to separate CO_2 from air: High Temperature (HT) solvent, Humidity Swing, and Temperature Swing. The HT solvent process uses a solvent to capture the CO_2 directly from the air. In figure 4: HT solvent process diagram, air is passed through the CO_2 capture unit where it is contacted with a solvent which removes the CO_2 from the air. The solvent is then regenerated in the pellet bed reactor so it can be re-used. The CO_2 is then transferred to solvent loop 2 via the reaction taking place in the pellet reactor. The CO_2 is then removed via heating to temperatures in excess of 900 degrees Celsius. Once the CO_2 has been liberated it is then compressed for use in downstream processes. This is the approach taken by Carbon Engineering (Keith, Holmes, et al. 2018) and is discussed in more detail in section 7.1 Chemistry and mass balance.

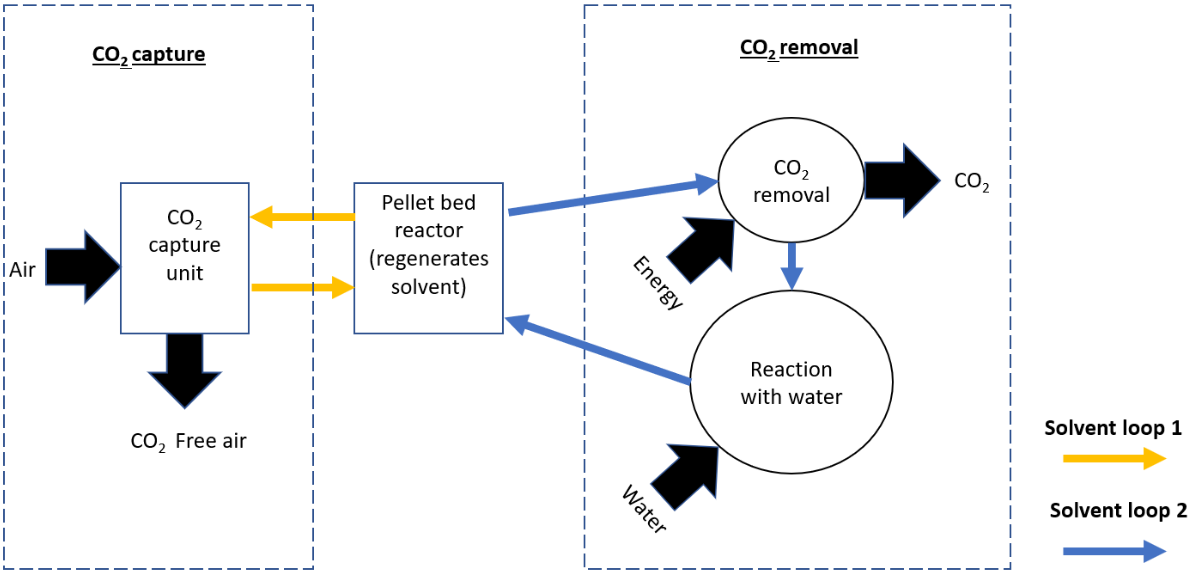


Figure 4: HT solvent process diagram

The temperature swing and humidity swing processes have many similarities. Both capture CO_2 onto a solid surface called a sorbent and both involve a two-step process. This solid sorbent is generally based on an amine, which is a common chemical used to remove contaminates from gases. An important aspect of these systems is the surface area of the sorbent which should be maximised so it comes into as much contact with the air as possible, and therefore increasing the effectiveness of CO_2 removal. The sorbent sits inside a chamber through which air passes through.

For the temperature swing process, when the sorbent is no longer able to capture CO_2 from the air passing over it, it is exposed to hot air which releases CO_2 from the sorbent. This means that the sorbent can then be reused. This method is carried out by the company Climeworks (Climeworks 2017).

The humidity swing process differs in that once the sorbent cannot remove any more CO_2 from the atmosphere, it is exposed to wet air which removes the CO_2 from the sorbent. The atmosphere within the chamber is then dried by exposing it to hot air. This method is being researched by Klaus Lackner (Wang, Lackner et al. 2011). Figure 5: Humidity and temperature swing processes diagram, describes these processes.

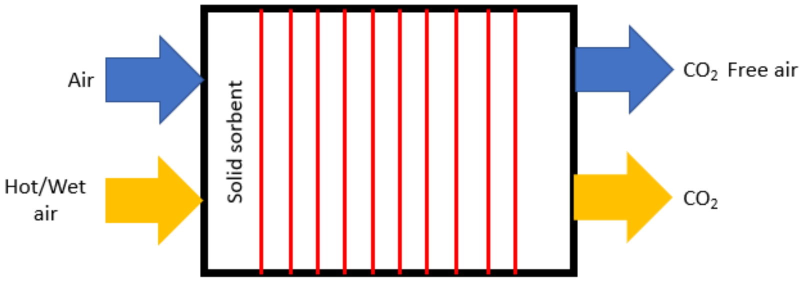


Figure 5: Humidity and temperature swing processes diagram.

1.2.1.1 Technological readiness level of DAC technologies

TRL is a method used by NASA and the European Space Agency to determine the technological development of systems (Héder 2017) and to compare the maturity of different technologies. It was developed by NASA during the 1970s. It consists of nine levels with level 1 being the least mature, where only a basic technological description of the system has been given, to level 9 which has been successfully and commercially deployed. Initially TRL only considered the technical dimension though it has now expanded to integration readiness, hardware, system readiness, manufacturing readiness, software readiness and programmatic readiness (US department of energy 2012). This analysis only considers the technical TRL levels as there is significant uncertainty regarding these aspects that need to be reduced before others can be considered. Table 2: TRL for HT solvent, temperature, humidity swing systems, details each level.

All DAC processes have been identified as being at an early technological readiness level (TRL) of 1-3 and a high mitigation potential of 4[Gt-CO_2/year] (Hills, et al. 2017), while others put it at TRL 2-4 (Mclaren 2011). DAC is commercially available and currently costs 600[\$/t-CO_2] (Climeworks 2020), though it is expected that this can be reduced to 100[\$/t-CO_2] (Evans 2017).

Carbon Engineering have released a paper describing a full-scale design of a HT solvent process with a description of their prototype system (Keith, Holmes, et al. 2018). This places their system at a TRL of 5-7.

The humidity/temperature swing process has been identified as at TRL 5-6 (Dowling and Ramakrishnan 2018). The temperature swing process is commercially available from the company Climeworks (Climeworks 2020) and was laboratory tested (Wurzbacher, et al. 2012) and therefore has been shown at TRL 9 and 4 respectively.

For the humidity swing system, laboratory testing (Wang, Liu, et al. 2013) (Wang and Qie 2018) has been carried out together with a rough potential process design (Lackner 2009). As a result this system has been demonstrated at TRL 4 and 5.

Table 2: TRL for HT solvent, temperature, humidity swing systems, outlines the TRL for each system with associated reference. Due to the differing levels of TRL for each system the starting point for developing a full system to compare it with others is different.

Classification	Details	HT Solvent	Temperature swing	Humidity swing
Basic research				
1	Basic principles observed and described, potential application conceivable	✓	✓	✓
Reference		(Hills, et al. 2017)	(Hills, et al. 2017)	(Hills, et al. 2017)
Technological development				
2	Description of a technological concept and/or application	✓	✓	✓
Reference		(Hills, et al. 2017) (Mclaren 2011)	(Hills, et al. 2017) (Mclaren 2011)	(Hills, et al. 2017) (Mclaren 2011)
3	Basic functional verification of individual elements of an application/technology	✓	✓	✓
Reference		(Hills, et al. 2017) (Mclaren 2011)	(Hills, et al. 2017) (Mclaren 2011)	(Hills, et al. 2017) (Mclaren 2011)
4	Basic proof of function, technology/application in the laboratory	✓	✓	✓
Reference		(Keith, Holmes, et al. 2018)	(Wurzbacher, et al. 2012)	(Wurzbacher, et al. 2012) (Wang, Liu, et al. 2013) (Wang and Qie 2018)
Basic research				
5	Functional verification in an application-relevant environment	✓	✓	✓
Reference		(Keith, Holmes, et al. 2018)	(Dowling and Ramakrishnan 2018)	(Dowling and Ramakrishnan 2018) (Lackner 2009)
6	Verification using a demonstrator in an application-relevant environment	✓	✓	✓

Reference		(Keith, Holmes, et al. 2018)	(Dowling and Ramakrishnan 2018)	(Dowling and Ramakrishnan 2018)
7	Prototype test in an operating environment	✓		
Reference		(Keith, Holmes, et al. 2018)		
8	Quality system with proof of functionality in an operating environment			
Commercialisation				
9	Successful commercial system deployment		✓	
Reference			(Climeworks 2020)	
Box is ticked if process has been reported at that TRL				

Table 2: TRL for HT solvent, temperature, humidity swing systems.

1.2.2 Policy component of socio technical regime

J. Wurzbacher from the DAC company Climeworks and A. Chanaewa from Skytree have identified that there needs to be an increase in policy support for the development of DAC (The National Academies of Science, Engineering and Medicine 2018). These policy support mechanisms include carbon pricing and funding methods for DAC.

The most significant carbon pricing mechanism is the European Trading Scheme (ETS), which is a carbon pricing policy (European commission 2019). The ETS is a system where large point source emitters pay a penalty for their emissions. There is a cap on the total emissions and a market mechanism is used whereby emitters can buy and sell carbon credits which allow them to emit. Over time the cap on emissions reduces which increases the cost of the credits.

There has been some policy development in terms of DACS with the development of the 45Q legislation. This is a funding mechanism for DACS which applies a tax rebate for CO_2 captured via DAC and geologically stored (US Congress 2017). Another funding process is Contract for Differences (CfD), which is a system where the additional cost of a product or service due to making the process more sustainable is passed onto the consumer, despite the market value of that product or service. An example of CfD use has been the conversion of coal power, run by the company Drax, to Bio-energy with carbon capture and storage (BECCS) plants within the UK (European Commision 2016). It has not been used in the context of DACS though DACS has been used in combination with a geothermal power plant in Iceland (Climeworks 2017). It could therefore be used as a funding mechanism for the incorporation of DACS into the electricity production sector.

The Clean Development Mechanism (CDM) is used by industrialised countries to meet their climate change targets by financially supporting the development of projects designed to reduce emissions in developing countries (UNFCC 2019). These projects include afforestation or supporting the development of energy efficiencies in developing countries. The CDM is designed to give industrialised countries flexibility in meeting their targets and continuing the economic development of developing countries. There are currently no projects which are used to fund DACS. Despite this, projects such as afforestation, which is also a CDR alternative as outlined in table 1: CDR processes comparison (European Academies Science Advisory Council 2018), demonstrate that the CDM can be used to fund CDR systems. This means that the CDM could be used a funding mechanism for DACS since DACS is a CRD.

Only the 45Q legislation specifically addresses DAC. Therefore, it is not known how the other methods described here can be used to attract investment into DAC and how they will affect DAC development.

1.2.3 Cultural component of socio-technical regime

Culture in the context of DAC relates to the relationship and interdependencies between DAC companies and other actors. It has been identified by R. Munson from the Global CCS Institute that an outreach plan for stakeholders is required for future development of DAC (The National Academies of Science, Engineering and Medicine 2018). This can help DAC companies leverage their position by identifying potential allies and threats from other actors. This relates to the culture component of the socio-technical regime. Therefore, this sub-section briefly outlines the stakeholder environment for DAC which is used later in this analysis to determine where DAC companies should best position themselves.

Stakeholders include energy providers such as oil/gas suppliers. Interaction between these actors and DAC companies is already taking place with a joint agreement signed between Exxon Mobil and the DAC company Global Thermostat (Exxon Mobil 2019).

Other stakeholders include those in potential CO_2 value chains. This includes those in the chemical industry where CO_2 can be used as a feedstock material for chemical processes. Stakeholders are required for geological storage of CO_2. An example of collaboration between CO_2 storage stakeholders and DAC companies is the DAC company Climeworks collaborating with the CarbFix project who store CO_2. This is done via injecting CO_2 which chemically reacts with carbonate rock underground (CarbFix 2020).

The potential allies will have similar objectives to DAC companies. By allying themselves with these stakeholders and identifying threats, DAC companies can increase the likelihood of their success through information sharing, identifying new opportunities for DAC and creating a road map for successful DAC implementation (Enserink, et al. 2010). There is currently no description in literature of the stakeholder environment in which DAC companies sit.

1.3 CO_2 supply chain

The purpose of this sub-section is to show where DAC sits in potential CO_2 supply chains. There are a number of supply chains available for CO_2 such as for synthetic fuel production or use in food and drinks industry though the technical and policy analysis of this thesis solely focus on Direct Air Capture Storage (DACS), while the stakeholder analysis also analyses interaction already taking place between stakeholders who are utilising CO_2 from DAC.

CO_2 can be stored via in situ mineralisation, where it is pumped underground and reacts with alkaline rock mixture. CO_2 storage locations require porous geological formations such as aquifers and empty oil and gas formations (Carbon Capture and Storage Association 2019). When this reaction is done above ground in a separate process it is called ex situ mineralisation (Romanov, et al. 2015).

CO_2 concentrations vary little around the world (NASA 2013). DACS systems would not necessarily require transportation as they could be located next to storage sites. This is a common challenge for carbon capture from industrial processes as these processes are generally not located next to suitable geological locations. Since the CO_2 does not need to be transported if the DAC system is located next to the storage system thus reducing the technological complexity and therefore increasing safety when compared to point source capture as the latter requires pipelines or shipping for transport of the CO_2 which can fail (UK Parliamentary Office of Science and Technology 2009). This would not be the case for offshore storage locations, such as in the North Sea, where it would not be economical or the safe option to have DAC system next to the storage location. This would mean the DAC system would need to be located away from the storage system and the CO_2 transported to it.

With regards to storage, there exists a legislative framework within the UK which outlines the licencing framework for CO_2 storage (UK Government 2008) and the EU have created a directive on the geological storage of CO_2 (EU 2009). Furthermore, this reduces institutional change requirements as legislation related to CO_2 transportation would not apply to DACS systems without transportation requirements.

1.4 Research direction

1.4.1 Research statement

DAC is an interesting area for research as it can remove CO_2 directly and if this undergoes long term storage it can reduce CO_2 emissions; though it is hindered by technical uncertainties. Furthermore, the effect policies are having on DAC development is not fully known. This is compounded by the fact that no description exists of the stakeholder environment in which DAC companies sit.

Different DAC processes are at different levels of TRL. The HT solvent process has a number of uncertainties related to a full system design regarding the heat and mass balance. While the humidity swing requires further exploration of potential system designs before a preferred alternative can be selected for development. For the temperature swing system further R&D is required to bring the cost down and make the system more economical.

The effects, both present and future, of different policies on DACS development are mainly unsolved. The only policy which directly addressed DACS was the 45Q legislation. While with other policies it may be possible to incorporate DACS though they have not been specifically designed with DACS in mind. Therefore, the effect these policies can have on DACS development should be analysed.

Stakeholders related to DAC companies have a variety of objectives. These objectives may aid or hinder the development of DAC. Furthermore, they each have different resources in which to achieve these objectives. This means that stakeholders can be categorised into allies or threats. Despite this, which stakeholders are allies and which are threats is unknown.

From answering the technical questions, a better comparison between the technologies can be made. This can then mean that a better DAC system can be selected to be coupled with other processes. For the question raised by the effect different polices will have on DACS this can accomplish how different factors may affect DACS development and what other co-effects maybe. Regarding questions related to how stakeholders will interact with each other, this can identify potential allies and threats.

To answer technical related questions the most appropriate method is to develop an energy and mass model which is measured against a set of criteria. A more detailed description of this is given in chapter 2. This was done in collaboration with the research institute TNO. The policy analysis is done through a system diagram where means on which to achieve objectives are analysed against a set of criteria. The stakeholder analysis is carried out in terms stakeholder's interdependencies and resources. These methods are discussed in more detail in chapter 2.

This research bodes to the benefit of society through the danger climate change poses which DAC has been identified as a contributing to the tackling of. This is due to the potential that DAC has to contribute to the reduction of CO_2 emissions. It has also been identified that CDR technologies, which DAC is a member of, as a requirement to achieve the Paris Agreement targets (IPCC 2018).

From this research, a comparison between the DAC process analysed can be made. It also contributes to policy insight on the effect policies are having on DACS development and allows DAC companies to understand potential allies and threats.

1.4.2 Research gap

The first knowledge gap relates to the technical aspects of DAC while the second is in relation to how different policies will affect DAC. The third knowledge gap is related to how stakeholders relate to each other in terms allies or threats.

- **Of the DAC technologies currently available which the better alternative is unknown.**
- **What the affects policies regarding climate change are having on DACS is unknown.**
- **How DAC companies might ally themselves with other stakeholders and who are the potential threats.**

By closing these gaps, it couples the social and technological aspects of DAC. This increases the understanding of how to develop the socio-technical structure of a low-carbon future and the effect this will have on the implementation of DAC. As outlined in Figure 4: The multiphase of transition of socio-technical systems (Loorbach et al. 2017), dual development in these areas results in a new techno-social landscape. As outlined by the IPCC the current system around greenhouse gas emissions is not conducive to continued human development (IPCC 2018). This therefore motivates change in the socio-technological landscape.

1.4.3 Problem owner

For stakeholder analysis, the results will be different for different stakeholders. Therefore, it is important to define your problem owner before carrying out the analysis. Here, the problem owner has been selected as the DAC company.

DAC companies such as Climework, Carbon Engineering and Global thermostat have identified issues relating to the high cost of the process, policy issues and stakeholder educations of the potential for DAC as factors blocking uptake of the process (The National Academies of Science, Engineering and Medicine 2018). From this, it can be identified that DAC companies have recognised that there is a problem which requires a multi-dimensional solution spanning technical, policy and stakeholder dimensions. This thesis is, therefore, aimed at DAC companies and considers solutions across these dimensions. This means that this analysis identifies DAC companies as the problem owner.

1.4.4 Research question

From this the following research question is composed which is designed to encapsulate the limiting factor of DAC(S) from technical, policy and culture regime of the socio-technical regime described in this chapter.

Research question:

"How do the most promising available DAC systems described in literature compare, where does DAC fit in current policy and how can DAC companies best position themselves to maximise the potential of their technology?"

Sub questions:

1. *Of the representative DAC technologies currently outlined in literature, how do these technologies compare with each other?*
2. *Of the policies that already exist regarding the tackling of climate change, what potential affect will they have on development of DACS?*
3. *What are the resources and interdependencies between the main actors which are related to DAC and can DAC companies leverage their position as stakeholders reduce their CO_2 emissions?*

Chapter 2: Methodology

This chapter outlines the research approach and method used to answer the research questions. Furthermore, it describes how information flows through this research.

2.1 Research approach

In chapter 1, a brief outline of the problem was given. Here, it was outlined that DACS has not been widely implemented due to technical, policy and stakeholder knowledge gaps. This section argues: that sub-question 1 and 2 requires a “bottom-up” (Lodico, Spaulding and Voegtle 2010) inductive approach, and that sub-question 3 requires an exploratory approach. This chapter will also outline the method in which each sub-question will be answered and give a description of the flow of information through each chapter which is shown in Figure 6 Research structure.

2.1.1 Main research question

“How do the most promising available DAC systems described in literature compare, where does DAC fit in current policy and how can DAC companies best position themselves to maximise the potential of their technology?”

In relation to the comparison aspect of the research question a number of criteria are developed to measure DAC technologies performance against. For this it is assumed that all the CO_2 is geological and long terms stored. For this the objective of climate change policies outlined in chapter 4 are used to develop a means-end diagram. The means are then operationalised into measurable criteria in an objectives tree against which policy and DACS systems can be measured against. These KPIs are then categorised as contributing to different criteria in which can make that DAC technology and policy combination an attractive option. This method to develop criteria comes from (Enserink, et al. 2010).

2.1.2 Sub-question 1

Of the representative DAC technologies currently outlined in literature, how do these technologies compare with each other?

Inductive reasoning is often described as “bottom-up”. Where the researcher builds from previous work to describe a picture (Lodico, Spaulding and Voegtle 2010). To make a fair comparison between these systems a full mass balance will be developed with sized and energy rated units. This will use the highest TRL available for that system and develop in a “bottom-up” manner from these a full mass balance, with sized and energy rated units. This means that an inductive reasoning process will be used to answer this sub-research question.

This method starts with a literature review on DAC as a whole. For this the search term “capturing CO_2 from air” and “techno-economic assessment of DAC” was applied to the search engines Google Scholar and Scopus. This yielded an approximately 92,230 results and by limiting this to “economics” resulted in the paper (House, et al. 2011). This was used as a starting point for the analysis and demonstrated that DAC is technically possible but that there are a number of limitations which makes the process expensive. While (Fasihi, Efimova and Breyer 2019) gave a more up to date description of the different state of the art DAC systems. In (Fasihi, Efimova and Breyer 2019) there were 6 descriptions of the HT solvent system, which was the most out of all systems available and therefore was selected for further study. While the humidity swing system was described to have current cost of 144 [$/t-$CO_2$] with a future price of 23 [$/t-$CO_2$] (Fasihi, Efimova and Breyer 2019), which is the lowest out of the three main types of DAC system though has low concertation for the CO_2 that it is produced. This was therefore selected for further study due to this price development.

A literature review on the humidity swing was then carried out. This used the search term “Direct air capture humidity swing” and “DAC humidity swing” using Google Scholar and Scopus. This resulted in an extensive list and systems which use humidity and temperature/vacuum were removed. The paper (Wang, Liu, et al. 2013) demonstrated experimentally that the humidity swing system can capture CO_2 at concentrations found in the atmosphere.

(Wang, Lackner, & Wright 2011) was used to understand the underlying chemistry of the system. The paper (Lackner 2009) was used to understand what a potential humidity swing DAC system would look like on a system level. From this paper a new system diagram was developed to be studied further. The paper (Wang, Lackner, & Wright 2011) was used for the capacity of sorbent, adsorption, de-adsorption time, inlet/outlet concentrations.

For the HT solvent system, a number of systems are proposed in the paper (Fasihi, Efimova and Breyer 2019). The system produced by the company Carbon Engineering is the most detailed (Keith, Holmes, et al. 2018) and studied further. The system design here had an unusual air contactor so a more detailed examination of the development of this system was made. The paper (Baciocchi, Stori and Mazzotti 2006) outlined a system which used off the shelf units. The paper (Keith, Heidel and Cherry, Capturing CO2 from the atmosphere: rationale and process design considerations 2010) describes the novel air contactor used in (Keith, Holmes, et al. 2018). From (Keith, Holmes, et al. 2018) a process flow diagram (PFD) was developed for further analysis. The units required for further analysis were also identified from (Keith, Holmes, et al. 2018).

With all the relevant data now collected regarding the humidity swing and HT solvent system it is now possible to model these systems. The humidity swing system was designed to match the outlet flow rate and concertation of the HT solvent system. This system has three sub-systems these are described in more detail in chapter 3. The first of these is the humidity swing sub-system. The volume requirements of the sorbent were first determined. Following this a sensitivity analysis was carried out on the factors which contribute to the cost of this sub-system. This sub-system was sized and costed. The second sub-system is an amine carbon capture system. For this an Aspen plus model of this type of system was used. The model used was on a different scale than required. Therefore, a sensitivity analysis was done on parameters related to the other sub-systems to determine if the system can be scaled. It was found that there was a linear relationship between thermal energy requirements and inlet flow rate therefore the system can be scaled linearly. The base case was then costed using Aspen Plus economic analyser. This was costed at the appropriate scale by scaling using the 6th 10th rule. For the 3rd sub-system, the solar thermal system, the area which was based on the solar thermal system of the other two systems was determined using the method outlined in (Zhai, et al. 2018). The levelised cost of solar thermal energy was determined using in (Zhai, et al. 2018) paper. This results from this were supplied to Aspen plus as hotutility. The solar thermal system was then costed using method outlined in (Zhai, et al. 2018). Following this the safety concerns was considered. The final technical analysis analysed the use of the re-boiler in the amine carbon capture sub-system as a heat source for the humidity swing sub-system.

The PFD developed from the literature review was used as a starting point. A full mass balance was developed from this. The identified units for further analysis were analysed for their energy requirements and size. For the air contactor this was done using Aspen Plus which was used to determine flow rates and composition changes. For the pellet reactor, steam slacker and calciner the duty was determined under isothermal conditions. The calculated values were then compared against the reported values.

The criteria developed through the methodology described in section 2.1.1 is used to measure to measure each technology.

2.1.3 Sub-question 2

Of the policies that already exist regarding the tackling of climate change, what affect will they have on development of DACS?

To answer this question a desk method approach was used, with an internet search on "significant climate change policies" to understand different climate change policies. The focus of research was narrowed to contract for differences (CfD), European Trading System (ETS), Clean Development Mechanism (CDM) and the 45Q legislation. These policies were selected as they are all significantly different. The CfD is a funding mechanism where consumers of electricity pay for green technology, the ETS is a system where large emitters pay a penalty for emissions, the CDM allows emissions to be offset to account for climate change targets and the 45Q legislation applies a tax credit for DACS. A full description of these policies is given in chapter 4.

The research approach used here is also an inductive reasoning approach. This is based on knowledge that CfD can be used as a funding mechanism in electricity production sector where the cost of the low-carbon electricity production is met by the consumer (Department for Buisness, Energy & Industrial Strategy 2019). Also, that the ETS is used to cap emissions where offsetting is not included (European commission 2019). Furthermore, that the CDM is used to reduce emissions by offsetting emissions in one location by removing emissions in another location (UNFCC 2019). While the 45Q legislation applies a tax credit for atmospheric CO_2 captured via DACS (Bennet 2018). From this it is possible to categorise these policies as supportive or not for DACS development.

Using these policies, a system diagram is developed using the method outlined in (Enserink, et al. 2010). This analyses the affect these policies have on DACS development and the co-affects carbon dioxide emissions and public finance requirements.

2.1.4 Sub-question 3

What are the resources and interdependencies between the main actors which are related to DAC and can DAC companies leverage their position as stakeholders reduce their CO_2 emissions?

The resources for an actor are the formal or informal means in which they can achieve their objective. An interdependency is the importance for one actor on another's resource to achieve their objective (Enserink, et al. 2010). These resources and interdependencies result in interaction by stakeholders to achieve their own objective. The analysis of this is generally called actor or stakeholder analysis. A meaningful study is yet to be completed regarding a stakeholder analysis of actors related to DAC. Exploratory research approach has been identified as appropriate approach for such problems (Brown 2006). This is used to identify priorities and definitions for a particular problem that future work can be built upon (Shields and Rangarjan 2013).

This used a desk research approach with a focus on internet searches on "direct air companies' joint development". From this it was found that there is a developing relationship between aviation sector and DAC companies (Virardi 2019). It was also identified that there is a developing relationship between oil/gas companies and DAC companies (Exxon Mobil 2019).

The technical analysis showed that the humidity swing system can capture CO_2 from the atmosphere which uses waste heat from an amine system which captures CO_2 from flue gas, and this resulted in an increase of 8% increase in CO_2 captured than would be captured by the amine system alone from the flue gas. This is described in further in section 6.6 Using stripper re-boiler as heat source for de-humidification. If the flue gas results from NG or coal combustion for electricity production, then a relationship between DAC companies and electricity producers shows potential. Therefore, a further desk research was carried out on the stakeholders in the energy production environment.

2.2 Information flow through research

The initial sections following this chapter are used to collect the information used to develop criteria on which to analyse the technology and policy on. Furthermore, it is the starting point for the development of technical description of the DAC technologies which are measured against these criteria. This is followed by the analyses on how policies will affect DAC. The penultimate section analyses the stakeholder environment in which DAC companies sit. This is concluded with a set of recommendation for DAC companies which are designed to give technical direction for DAC development, understand which policies they should be supportive of and where they best fit in relation to other stakeholders. Figure 6: Information flow through thesis, depicts this, along with the flow of information through this research.

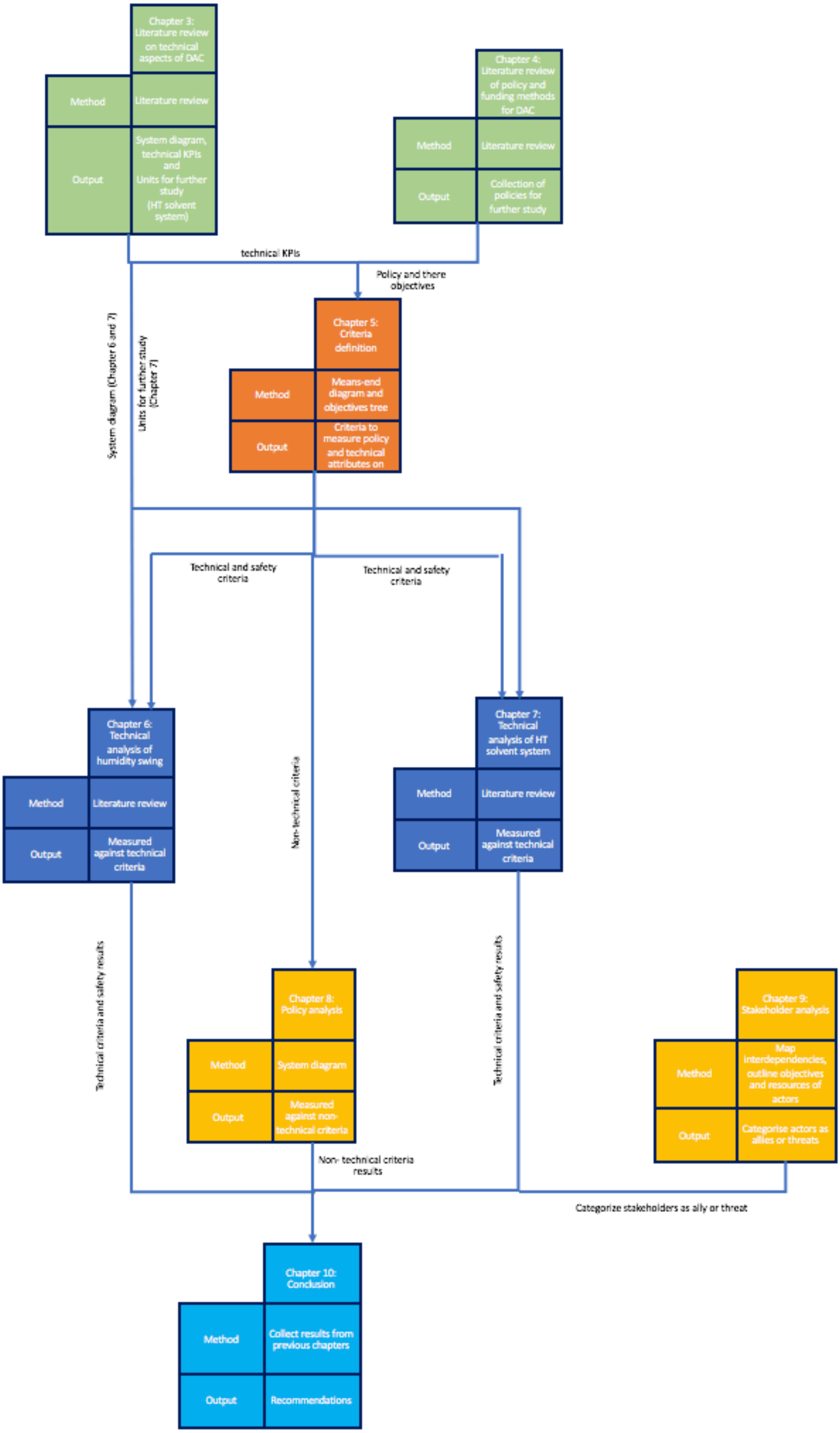


Figure 6: Information flow through thesis.

2.3 Scope of work

Sub-question 1 relate to the technical aspects of Direct Air Capture (DAC). This is because inclusion of utilisation options would increase the complexity of the work beyond the scope of Engineering and Policy Analysis (EPA) Master's thesis due to the technical uncertainties of such options. Storage options are comparatively well understood (Kelemen, et al. 2019) to DAC and therefore in comparison to DAC little knowledge can be gained by further study. Despite this, it is assumed that all of the captured CO_2 is geologically stored on a long-term basis. The cost of this is not included in the reported cost. This has been done as ff this thesis is to be used for future research with regard to CO_2 utilisation, which has shown promise (Royal Society 2019), the latter can be easily coupled with the DAC processes described here without having to remove the CO_2 storage element.

In terms of the policy aspect, this research focuses on Direct Air Capture and Storage (DACS). This is because policies require the CO_2 to be dealt with in some way, either by utilisation or by storage. For utilisation there are number of options available and each would need to be considered for a meaningful analysis. This would also be beyond the scope of a EPA Master's thesis. Therefore, policies are only analysed in terms of the affect it can be expected to have on DACS.

For the stakeholder analysis DAC is considered with regard to both storage and utilisation is considered. This is because they are key components in understanding the stakeholder environment, and also this requires no technical or policy implication analysis of utilisation or storage. It is therefore appropriate, to include in this analysis.

Chapter 3: Review of literature on technical aspects of DAC

This chapter contains a technical literature review for the potential of DAC in addition to a review of the literature related to the humidity swing and the HT solvent system. The main outputs for this chapter are a high-level system design for humidity swing and HT solvent system. This is followed by a list of the units to be further analysed from (Keith, Holmes, et al. 2018) and a list of relevant KPIs.

3.1 Potential for DAC

A thorough examination has been made of the limitations of atmospheric capture of CO_2 (House, et al. 2011). The starting point for this considered the use of the Sherwood plot. The Sherwood plot demonstrates the inverse relationship between metal concertation during extraction of the target material and market value of that metal (Sherwood 1959). This was then updated to include a range of other biological and pollutant materials (Grubler 1998) and indicates a high cost for capturing atmospheric CO_2. (House, et al. 2011) used this to estimate the cost of capturing atmospheric CO_2. House *et al* used the equation $P=A/C$ where P is the price in [\$/kg], C is the initial concertation and A is a separation constant with units [\$/kg]. Using a separation constant of 0.001[\$/kg] yielded them cost of 2,500[\$/kg-CO_2]. Looking at the cluster of acid gases it was recognised that this has a shallower slope than other groups. The researchers therefore re-estimated this function to determine the price for removing the gas, this new function is $P = 0.0208(1/C)^{0.5434}$. With a concertation of 400ppm for atmospheric CO_2 the cost is estimated to be approximately 1100[\$/t-CO_2]. The Sherwood plot can be seen in Figure 7: Extended Sherwood plot (Grubler 1998), with the concertation of CO_2 marked in red.

(House, et al. 2011) consider contributing factors of this value as the limiting factor of the thermodynamic efficiency, cost attributed to energy generation, high cost of similar systems and the process plant design.

Despite these findings, since publication of this work the cost of DAC has come down dramatically and it has becomen commercially available at 600[\$/t-CO_2] (Climeworks 2020) . Analysis suggests the price could come down to 100 [\$/t-CO_2] (Keith, Holmes, et al. 2018). From this it can be seen that recent work deviates from earlier work and demonstrates the potential for DAC.

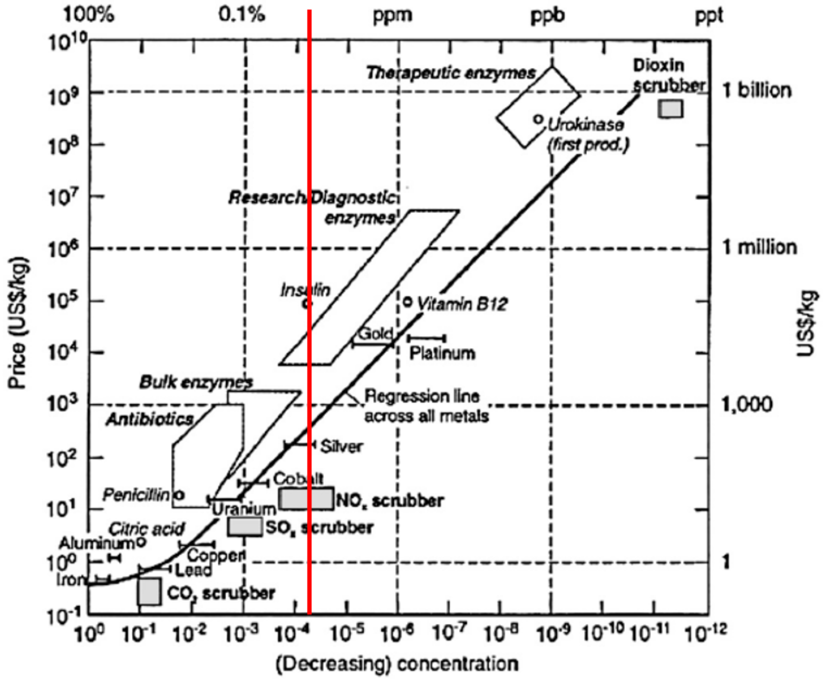


Figure 7: Extended Sherwood plot (Grubler 1998).

3.2 Selection of DAC systems for further study

As outlined in chapter 1 there are three main types of DAC systems available: humidity swing, temperature swing and HT solvent system. In (Fasihi, Efimova and Breyer 2019) a techno-economic analysis is carried out of each system. Out of the three the humidity swing system is the least understood process with only one system used in (Fasihi, Efimova and Breyer 2019). Also, searching “DAC humidity swing”, “DAC temperature swing” and “DAC high temperature solvent system” into google scholar returns 1,820; 16,300; 21,000 results respectively. Due to the lack of research, though it is a promising alternative (Fasihi, Efimova and Breyer 2019), the humidity swing is selected for further study.

For the HT solvent system, a description of a full-scale system is given with the results from a prototype plant for the system (Keith, Holmes, et al. 2018). This system gave very promising results in terms of CO_2 production rate and cost. Despite this, there are a number of uncertainties relating to the system design. These include recycle rates, unit sizing and energy ratings. To fully interrogate these uncertainties and (in)validate their findings through full process design, designing and operating a protoplanet goes beyond the scope of this thesis. Despite this, a PFD is developed which is created from there description with associated mass balance, with sized and energy rating given to the most significant units.

Due to the description given in the last two paragraphs the humidity swing and HT solvent system were selected for further study. This study compares these two systems.

3.3 Humidity swing DAC system

A description of the humidity swing system is given in chapter 1: Anthropological climate change. This system has been selected for further analysis as a complete process design of such a system is missing.

This process has been pioneered by K. S. Lackner who first published the potential for such a system in 2009 (Lackner 2009). This type of process was motivated as previous research had focused on strong sorbents such sodium hydroxide and potassium hydroxide which requires high temperatures, of 900 degrees Celsius, to release the CO_2 from the solvent. Further there was concern that the solvent would be entrained in the CO_2 free air and would exceed legal limits therefore the need for a demisting unit as, incorporated into the APS design of a HT Solvent system (Socolow, et al. 2011). This drew the researcher to a solid sorbent finally concluding that the ion-exchange resin Marathon A Dow Chemicals company (Dow 2002) to be the most appropriate due to its high uptake rate of CO_2 even from low concentrations. From experiments it was found that by introducing the sorbent to water it rapidly released the CO_2 with a partial pressure of 5[kPa].

3.3.1 Early proposed design

This research also proposed the design of a prototype. The capture unit would be exposed to atmospheric air and capture CO_2. During the regeneration, the capture unit would be placed inside a chamber and exposed to a wet atmosphere. After the equilibrium is reached the and no more CO_2 can be expunged from the sorbent, the air capture unit would move to another chamber where it will again be exposed to a wet atmosphere. This would be done in a cascade manner with the partial pressure of CO_2 increasing in each stage. Figure 8: proposed system for humidity swing system (Lackner 2009) depicts this. There are no detailed costings calculations attributed to this system.

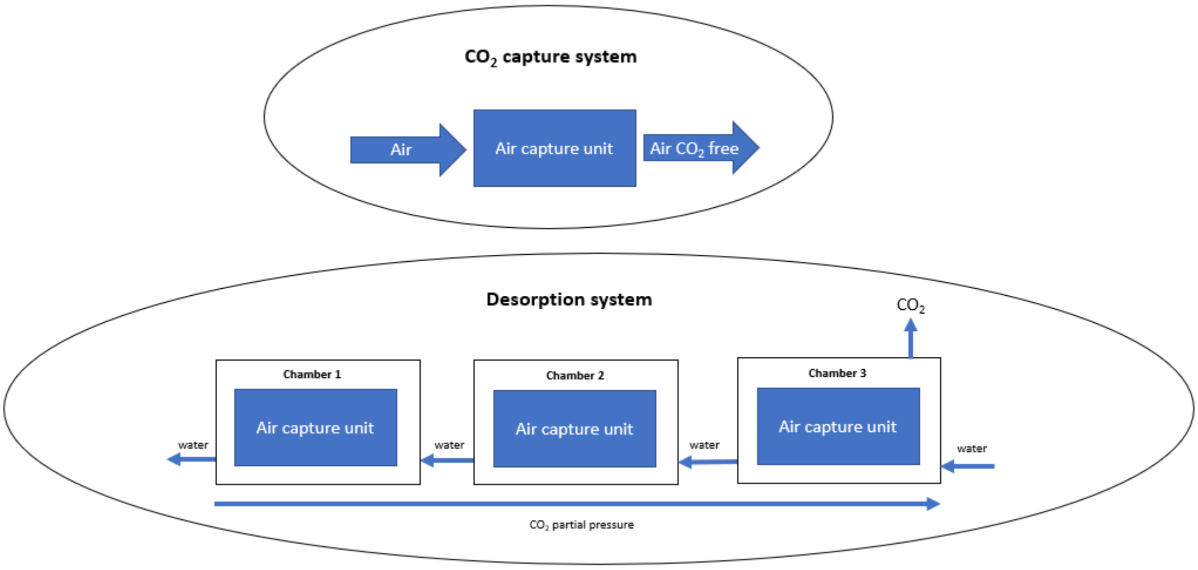


Figure 8: proposed system for humidity swing system (Lackner 2009).

3.3.2 Reaction kinetics

A more detailed examination of the reaction mechanism has been given in (Wang, Lackner and Wright, 2011). Experimentation was employed and it was found that atmospheric concentrations of CO_2 could be increased from atmospheric concentrations of CO_2 at 0.04[kPa] to 5[kPa]. In this experiment the resin Marathon A (Dow 2002) was exposed to air with a CO_2 concertation of 430[ppm]. This was done over a 24-hour period by injecting the gas into the chamber every time the system came to equilibrium. This meant that the sorbent was saturated with CO_2. The sorbent was then exposed to liquid water. This was done by filling the chamber with water. The water was then removed leaving no air in the chamber. The water was left in the chamber for two minutes. Some of the CO_2 will have absorbed into the water but the water was not left in the chamber long enough for the CO_2 on the sorbent to come to equilibrium with the water. The resulting gas within the chamber had a concentration of 5% CO_2. These results were corroborated through other experimental research (Wang, Liu, et al. 2013).

Further research on the humidity swing system focussed in the reaction kinetics (Shi, Xiao, et al. 2016). This is because the mechanism goes against the mass action law. The reaction that takes place can be seen in equation 1.

$$OH^- + CO_2 \leftrightarrow HCO_3^- \; eqn. (1) \; carbon \; dioxide \; adsorption \; (dry)$$

$$CO_3^{2-} + CO_2 + H_2O \;\leftrightarrow HCO_3^- + HCO_3^- \; eqn. (2) \; water \; adsorption \; (dry)$$

$$CO_3^{2-} + CO_2 + H_2O \;\leftrightarrow HCO_3^- + HCO_3^- \; eqn. (3) \; carbon \; dioxide \; de - adsorption \; (wet)$$

When water is added to the system it would be expected that CO_2 is adsorbed and then released when removed water is removed in accordance with mass action law. In reality, the opposite is true. This peculiarity is explained through Gibbs free energy of CO_3^{2-}. Gibbs free energy is the thermodynamic potential energy available. When water is exposed to CO_3^{2-} the available energy to react with CO_2 reduces, ehich promotes the backwards reaction in *eqn. (3)*. This means that when CO_3^{2-} has this water removed it has an increased amount of energy to react with CO_2 which promotes the forward reaction in *eqn. (2)*. This reaction set is also depicted in Figure 9: Humidity swing mechanism (Wang, Lackner and Wright, 2011).

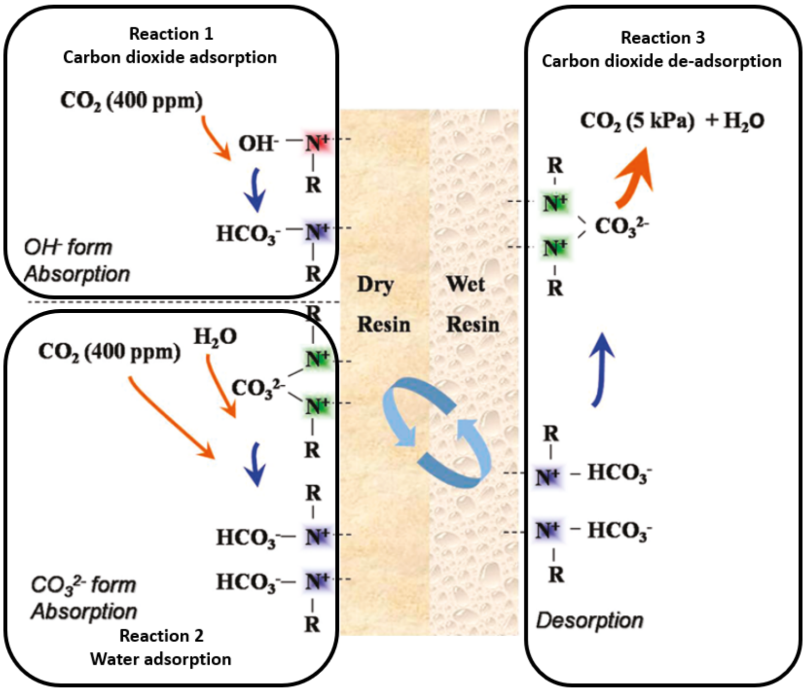


Figure 9: Humidity swing mechanism (Wang, Lackner and Wright, 2011) .

3.3.3 Location specific

Experiments have been based on the humidity of a dry location (Baciocchi, Stori en Mazzotti 2006) (Wang, Liu, et al. 2013). If a process plant is based in a dry location it would be possible to exploit the dry air when removing water from the system, therefore reducing operating costs. This limits where the plant could be located. Furthermore, if the energy source is solar thermal energy then the system should be located where this is maximised.

3.3.4 System design

The proposed system design to be studied further in this thesis, deviates from previous system design depicted in figure 8: proposed system for humidity swing system (Lackner 2009). This is shown in figure 10: high level system design for humidity swing. The system only consists of one air capture unit which captures atmospheric CO_2. This then releases the CO_2 at concentration of 5% which is then further concentrated to a high concentration in conventional carbon capture amine system. The humidity swing unit is novel while the amine unit is well understood. Reducing the number of novel units therefore simplifies the system. This system is colour coded in terms of the two steps. Step 1 is in blue when the system is dry and step 2 is in red when system is wet. For this it was assumed that all the CO_2 is removed from the air that contacts with the sorbent. In reality not all the CO_2 will be captured. Though there is no information regarding the rate the CO_2 is absorbed onto the sorbent. The experimental work that was carried which this analysis is based on (Wang, Lackner and Wright, 2011) exposed the sorbent over a 24-hour period to atmospheric concentrations of CO_2 to remove the maximum amount of CO_2.

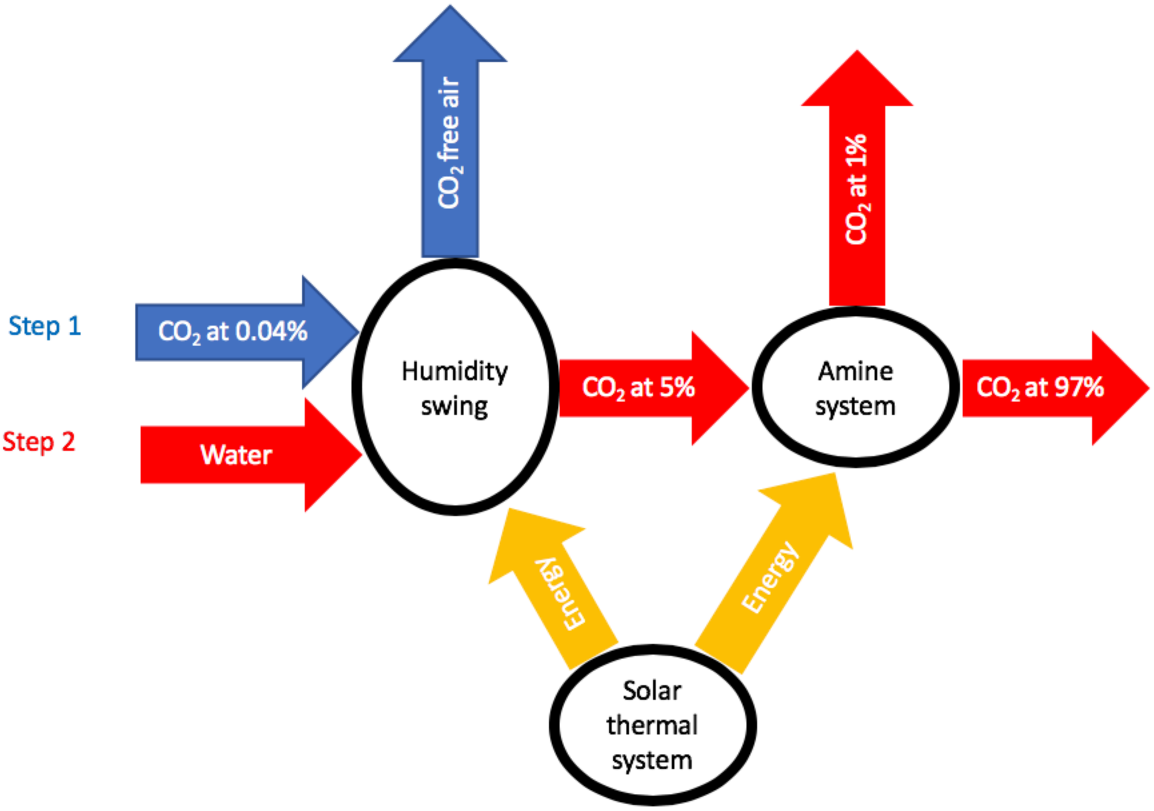


Figure 10: *high level system design for humidity swing.*

Despite CO_2 capture from air there are a number of limitations which have been outlined in (House, et al. 2011). These centred around the difficulty caused by the low concentrations of CO_2 that exist. This means that a large amount of air needs to be processed to capture useful amounts CO_2 which requires pumping energy. The material that needs to be processed can be reduced by using a solvent which binds strongly to the CO_2 though this will require large energy input to regenerate the solvent. If the energy used to process the material and regenerate the solvent comes from a source which is not CO_2 free this limits the effectiveness of the process. Further similar processes such as SO_2 removal are limited by contamination of the solvent.

3.4 HT solvent DAC system

A simplified process flow diagram of the HT solvent system can be found in figure 4: HT solvent process diagram. This is a three-step process consisting of the air capture unit, the energy intensive removal of CO_2 from the solvent and finally the regeneration of the solvent which requires water. It was first proposed by Lackner et al. (Lackner, Grimes en Ziock 1999) and it was demonstrated that using a solvent such as potassium hydroxide or sodium hydroxide was feasible at capturing atmospheric CO_2 further they identified that main cost of such a system would be attributed to the regeneration of the solvent.

3.4.1 Early process designs

The first process design of such a system was proposed by Baciocchi et al. (Baciocchi, Stori en Mazzotti 2006). This design focused on the capture and regeneration units. This process uses commercially available units and components. The air contactor has similarities to a conventional carbon capture process from flue gas. Its main deviation comes from the fact that since atmospheric concentration is significantly lower than that of flue gas this means that the solvent used to capture the CO_2 must come into contact with a significant amount of CO_2 to make the capture rate effective. To overcome this, the researchers used an internal packing specifically designed for a high gas through put and pressure drop. Furthermore, they use the solvent calcium hydroxide to remove CO_2 from the atmosphere due to its high selectivity for CO_2. The solvents are limited to those which are strongly basic and therefore have a high selectivity for CO_2.

The next unit process is similar to the process used in the production of cement. This is where the CO_2 is released via through calcination. This is done by heating the solvent which has the CO_2 captured in it to high temperature, greater than 900 degrees Celsius. This unit consist of a steel rotating drum which is heated releasing the CO_2. To complete the regeneration of the solvent the product from the calcinator needs to react with water. This is done by a unit called a steam slacker which operates at 90 degrees Celsius. This is depicted in Figure 11: HT solvent process depiction (Baciocchi, Stori, & Mazzotti, 2006).

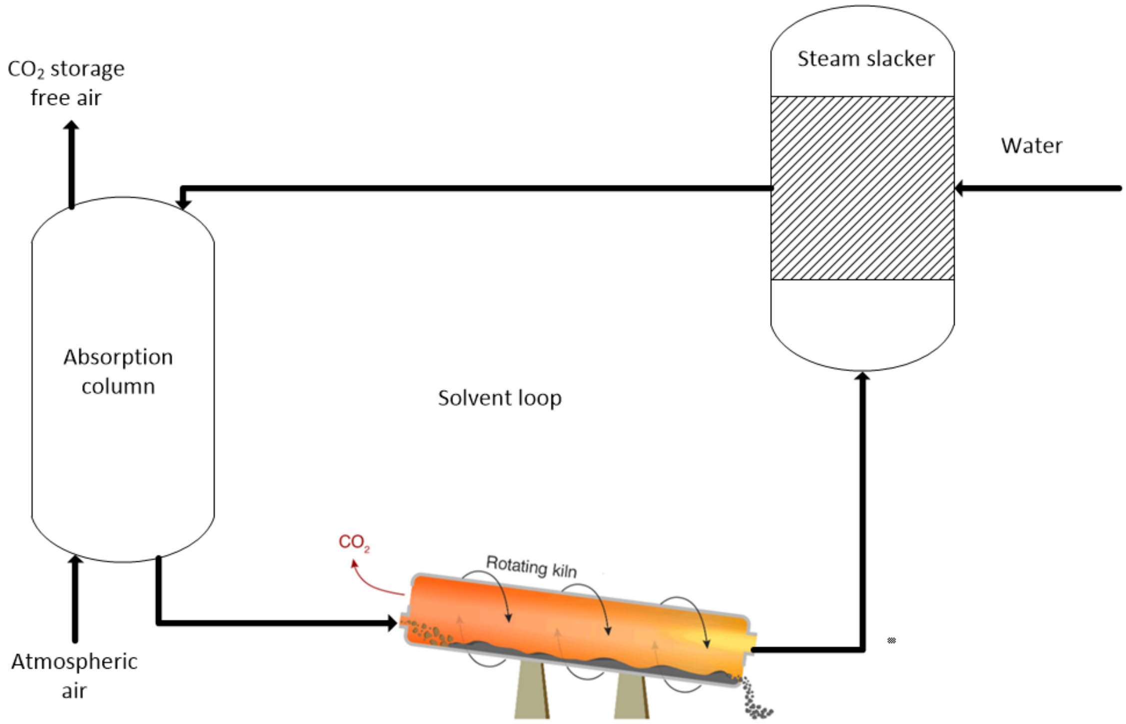


Figure 11: HT solvent process depiction (Baciocchi, Stori en Mazzotti 2006).

This type of system was analysed further by Zeman (Zeman 2007). This recommended that future research should focus on the air contactor as the throughput of air is proportional to the capture rate.

3.4.2 The American Physical Society system

The American Physical Society (APS) explored the process design of DAC system (Socolow, et al. 2011). This type of system used solvent sodium hydroxide with a standard air-liquid air contactor. Due to the high contact area requirement between air and the solvent there the proposed design would use a number of air contactors in parallel which would have a footprint area of 50,000[km^2] to capture 1[Mt-CO_2/yr] using steal packing. This system also had a demister unit to capture any solvent entrained in the gas exit of the air contactor. The reasoning for this is that there are legal limits on concentrations that sodium hydroxide can have in air which the APS believe could be exceeded in the gas exit of the absorption column without a demister unit.

3.4.3 Novel air contactor

Following work by (House, et al. 2011) outlining some of the main constraints of a DAC system (Holmes and Keith 2012) developed the air contactor so to increase contact area while minimising pressure drop which was identified by (House, et al. 2011) as a significant contributor to the total costs. The proposed system has a slab geometry which has similarities to cooling towers and gas scrubbing systems. The solvent travels vertically driven by gravity while gas travels across this flow horizontally. This also has the effect that there is an increased flexibility in the type of packing that can be used in the contactor (Holmes and Keith 2012). Packing is a used in a range of chemical processes where there is a requirement for a high contact area between the processed materials. An example of a packing material used in contactors can be seen in Figure 12: Example of structured packing used in absorption and distillation (AMACS Process Tower Internals, 2018). This type of system is found to have much lower solvent to gas ratio flow rate requirements. This means that lower pumping requirements for the liquid solvent It is a modular system where each contactor is arranged in a rectangular cross-sectional geometry. This makes it more flexible in terms of the amount of gas that can be processed. This research also indicated that there is a requirement for air pumping as winds cannot always be assumed to have the required air velocity which would have increased costs attributed to this system. Despite this, since this uses equipment already widely used in cooling towers and gas scrubbing there is little need for finance for R&D. The largest liquid-gas contactors that are in existence are 100[m^2]. (Holmes and Keith 2012) calculate that the required size to capture 1[Mt-CO_2/yr] is 38,000[m^2]. This would therefore require a significant number of units set up in parallel which would increase the initial capital cost but could exploit economies of scale. Figure 13: Carbon engineering slab air contactor (Holmes and Keith 2012) depicts how this system would be set up.

Figure 12: Example of structured packing used in absorption and distillation (AMACS Process Tower Internals 2018)

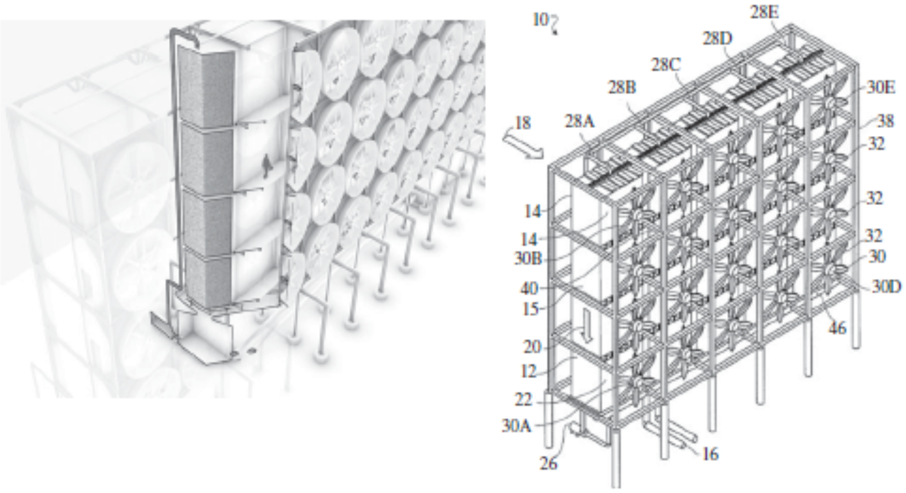


Figure 13: Carbon engineering slab air contactor (Holmes and Keith 2012).

3.4.4 Complete process plant design

The HT solvent process is the process used by the company Carbon Engineering, which has released a paper describing their process (Keith, Holmes, et al. 2018). A process flow diagram for this can be found in Figure 14: Original PFD of the HT solvent process (Keith, Holmes, et al. 2018). This is the most detailed description of a DAC plant available.

This system uses potassium hydroxide to capture CO_2 and uses the same air capture unit as outlined by (Holmes and Keith 2012). It uses the combustion of CH_4 in the energy generation system. The CO_2 in the exhaust to the energy generation system is captured in a conventional carbon capture system which also uses potassium hydroxide as a solvent.

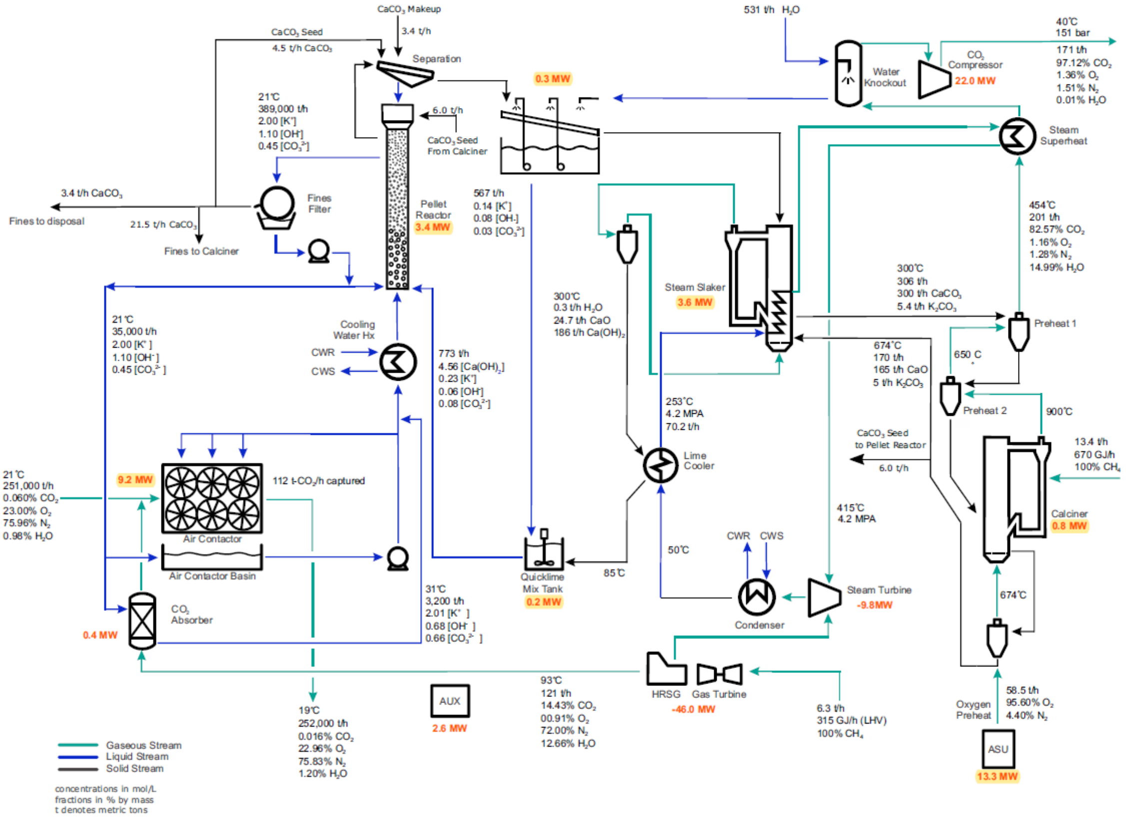


Figure 14: Original PFD of the HT solvent process (Keith, Holmes, et al. 2018).

Table: 3 Input output variables (Keith, Holmes, et al. 2018), outlines the most significant criteria on which this system was measured from this paper by Keith *et al.*

Fan energy cost [kWHr/ t-CO_2]	Fluid pumping energy cost [kWHr/ t-CO_2]	Cost of electricity [$/MWHr]	Energy cost (not electricity) [$/MWHr]	Inlet conc. of CO_2 [PPM]	Outlet conc. of CO_2 [Mass%]	Outlet flow rate [t/hr]	Levelized cost of CO_2 [$/ t-$CO_2$]	CAPEX [$Mill]	Inlet flow rate [t/hr]
Input	Input	Input	Input	Input	Input	Input	Output	Output	Output
61	21	60	3.5	400	97.12	171	232	1,126.8	251,000

Table 3: Input output variables for (Keith, Holmes, et al. 2018).

The levelised cost of CO_2 is determined through the sum of capital costs, non-fuel operating maintenance cost and energy costs. The CAPEX is calculated from the cost of building the plant plus the labour and land costs. This was based on cost of these in Canada in 2018.

Despite the system described by Keith *et al* been the most detailed a number of uncertainties remain. These includes the characteristics of the reactors, the concentration around the air contactor, the recycle rates and many of the stream splits which are unknown. Furthermore, using fossil fuel as energy source may negatively affect LCA assessment for downstream CO_2 utilisation methods; though this is not further analysed in this research.

3.4.5 Future of HT solvent DAC systems

The temperature requirements for the regeneration process are 900 degrees Celsius (Socolow, et al. 2011). This means that the use of waste heat or a low temperature source such as renewable power generation are not suitable

Borrowing research from the cement industry, which uses the same process as the calciner where the CO_2 is released using high temperature, it has been demonstrated that this process can be done electrochemically (Ellis , et al. 2019). This means that there is no need for high temperature and since the energy source is electricity it can be generated without CO_2 production via wind, solar or other renewable method. Furthermore, this means that if this system was used in a context where it was utilised which releases CO_2 it has the potential to be net-zero CO_2 emitting. This is of relevance as CO_2 utilisation methods have been identified as contributing to CO_2 emission mitigation (Bujnicki, et al. 2018).

3.4.6 Timeline of development of HT solvent DAC system

Figure 15: Development of HT solvent system, outlines the development of the HT solvent system through a timeline of developments of this system. This research does not focus on use of electrochemistry to release CO_2 from the solvent as outlined in (Ellis , et al. 2019) as there is no detailed design description of such a system available in literature.

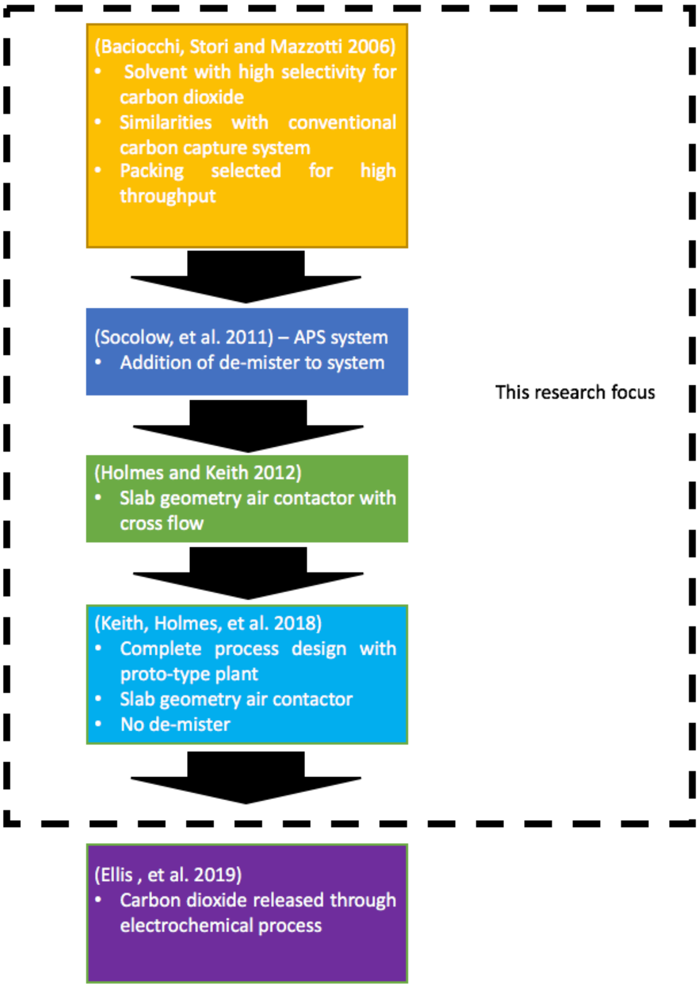


Figure 15: Development of HT solvent system

3.4.7 System design for HT solvent process

The units that will analysed further in this thesis are outlined in table 4. These are discussed in chapter 6. These units are selected for further study as they contribute the most to the energy requirements of the system.

Unit	Air contactor	Pellet reactor	Calciner	Steam slacker

table 4: HT solvent system uncertainties

Using figure 14: Original PFD of the HT solvent process (Keith, Holmes, et al. 2018), this has been simplified to a high-level process design. Figure 16 PFD for high temperature solvent process, can then be used in chapter 7 during the technical analysis of this system.

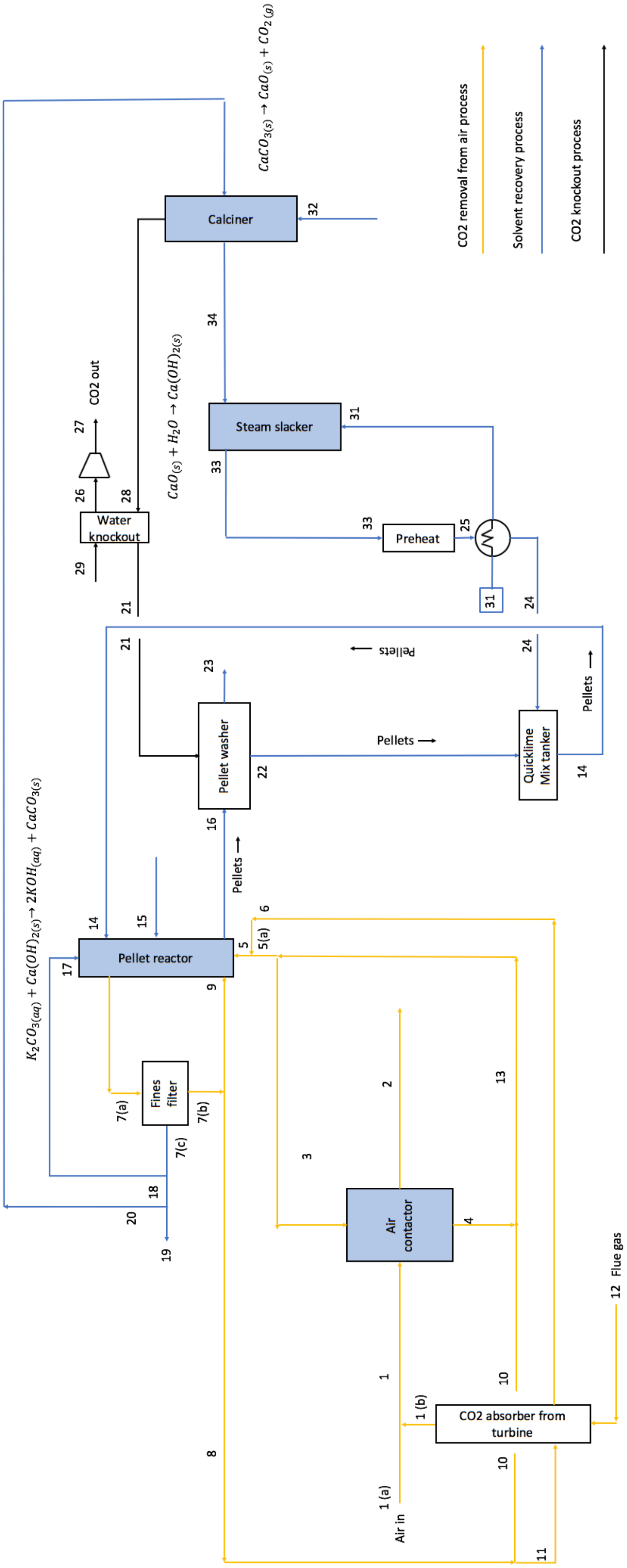


Figure 16: PFD for HT Solvent process.

3.5 Technical KPIs

Considering Table 5: Input output variables for (Keith, Holmes, et al. 2018), levelised cost of CO_2 and CAPEX were identified as relevant KPIs to measure for the HT solvent system. Further to this the OPEX is relevant factor. The OPEX is defined as the cost per unit time. This is relevant to operators as allows them to easily calculate total cost per year. These relate to the cost of system and are analysed further in this thesis. Since both systems are energy intensive then the direct energy requirements is a relevant criterion. If these systems result in direct emissions, then it reduces the effectiveness of the system. This can be seen in the HT solvent system as they capture the CO_2 from the combustion of methane which is use as an energy source. Therefore, direct CO_2 emissions from the plant is relevant criterion. These criteria are outlined in table 5: Technical KPIs.

Technical KPIs	levelised cost of CO_2	CAPEX	OPEX	Direct energy requirements	Direct CO_2 emissions from the plant

table 5: Technical KPIs

3.6 Summary

3.6.1 Limiting factors for DAC

From the paper (House, et al. 2011) it is identified that there needs to be a large amount of air that needs processed to capture a meaningful amount of CO_2. This can be reduced by using a strong solvent to capture the CO_2 though this will require increase energy to liberate the CO_2.

3.6.2 Humidity swing DAC system

Development of the humidity swing process was motivated by concerns regarding the HT solvent process related to energy requirements of regeneration and solvent which has been identified as a limiting factor by (House, et al. 2011). Most researched has focussed on the kinetics of the reaction which takes place (Wang, Lackner & Wright 2011). Experimentation has demonstrated that CO_2 concertation can be increased from 0.04% to 5% using the sorbent Marathon A Dow Chemicals company (Dow 2002).

3.6.3 HT solvent DAC system

This was the first type of process outlined after capturing atmospheric CO_2 was proposed by (Lackner, Grimes en Ziock 1999). The greater contact area required by the absorber was identified and packing was selected with this in mind.

The APS then proposed a design which was able to capture 1[Mt-CO_2/yr] which had significant costs attributed to the number of absorber columns required to capture this amount of CO_2. Further, in their proposed system there was a demister unit to capture sodium hydroxide entrained in the air leaving the air contactor.

With much of the research identifying that the majority of the cost been attributed to the air contactor (Holmes and Keith 2012) researched efficiencies that could be found in this unit. It was found that by having an air-contactor which has similarities to a cooling tower gas scrubbing system where solvent flow vertically and gas horizontally was able to increase efficiency of capturing CO_2 which means that the solvent to gas ratio can be reduced. This means that pumping energy requirements can be reduced. Despite this development, this unit it will still require a significant cross-sectional area to capture relevant amount of CO_2.

The air contactor described in (Holmes and Keith 2012) is incorporated into the design produced in (Keith, Holmes, et al. 2018). In this design potassium hydroxide, which binds strongly to CO_2, is used as the solvent. This system uses coal/gas as the energy source and captures the CO_2 from this. It regenerates the solvent via high temperature. It is the most developed system, though a number of uncertainties still exist. It has been studied further as it has the most information available and the authors have demonstrated its feasibility regarding price per ton of CO_2.

There has been developments from the cement industry which use a similar process to the regeneration of the solvent in the high temperature DAC process where electrolysis is used (Ellis , et al. 2019). This means that there is no need for the high temperature step and this type of system is more suitable for CO_2 free energy source such as wind or solar.

Chapter 4: Review of policy and funding methods for DAC

This chapter will first describe how funding responsibilities are distributed between different countries via the Common but Differentiated Responsibility (CBDR) method. It will then examine how industrialised countries fund systems to tackle climate change in developing countries using the common but Clean Development Mechanism (CDM). It will then examine EU policy objectives which are used to aid design of policies within the EU. This will be followed by a description of the European Trading System (ETS) which is designed to reduce emissions within the EU. The mechanism used in the UK to fund sustainable technologies in the electricity production sector, Contracts for differences (CfDs), will then be considered. This chapter will then describe the 45Q legislation taking place in the USA. A comparison will then be made with other technologies, designed to tackle climate change, from the private investment sphere. The hierarchical relationship between these is shown in Figure 17: Hierarchical relationship between means to invest into climate change technologies. The arrows represent the hierarchical relationship.

The output for this chapter is a list of methods to tackle climate change with their associated objectives which is used to help define the criteria developed in chapter 5.

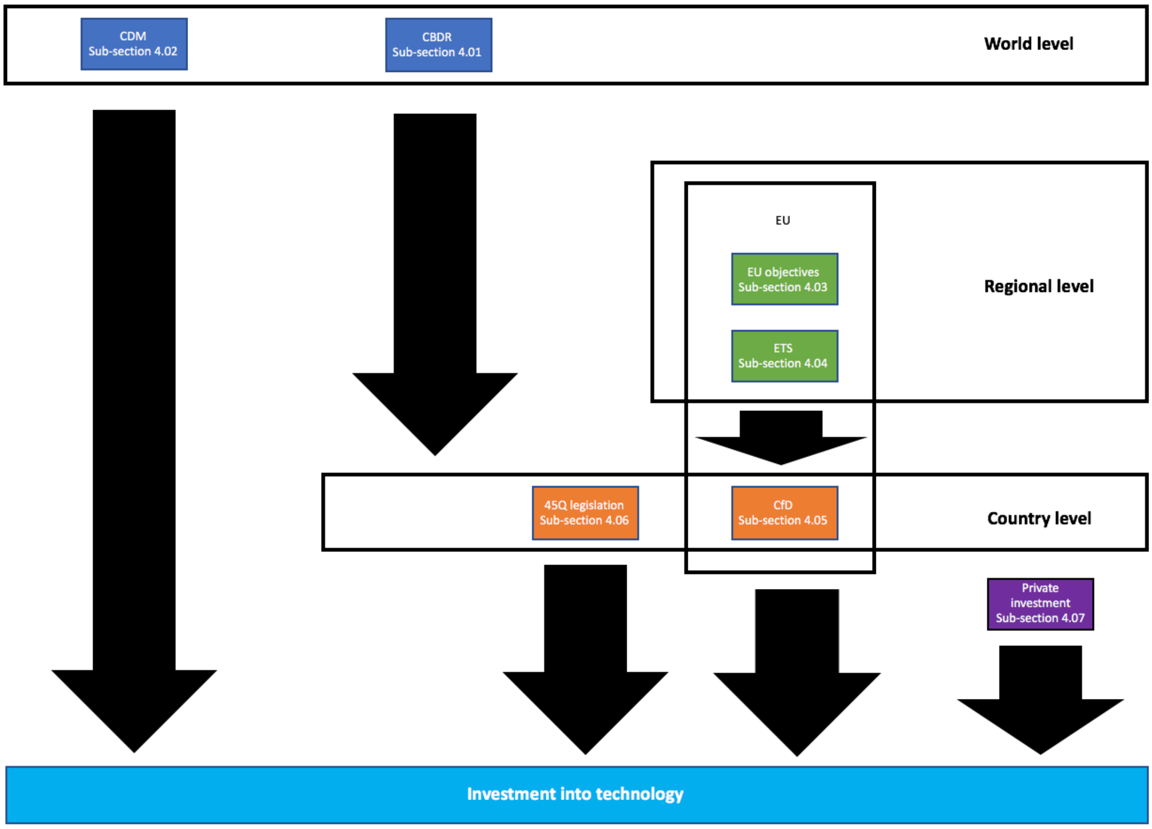


Figure 17: Hierarchical relationship between means to invest into climate change technologies.

Following this is policy recommendations that relate to DACS from (IOGP 2019), which gave policy recommendations regarding CO_2 storage, are outlined. The output for this section is a list of the policies that is used to define criteria in chapter 5 and the objectives of these that relate to DAC.

4.1 Common but differentiated responsibilities

The EU has identified that that many emerging economies should not have the same commitment as developed economies (Dimas 2005).The United Nations Framework on Climate Change (UNFCCC) use the Common but Differentiated Responsibilities (CBDR) method to attribute financial responsibility to different countries. The CBDR method accepts different countries cannot contribute equally to combating climate change. It is a philosophy used to design frameworks relating to tackling climate change which takes into account this in-equality between nation states (Cullet 1999).

It differentiates countries into two groups. Group one, Annex II, countries are those that are industrialised and those that are in industrialisation transition. While non-Annex I countries are developing countries and are at particular risk from climate change (United Nations Climate Change 2019). Annex II groups are industrialised countries and are required to supply finance to enable developing countries to develop emission reduction measures (United Nations Climate Change 2019).

In principle the CBDR method is of sound reasoning though in reality there is scope for disagreement on the effect of and solutions to climate change (Bortscheller 2010). This is due to the range of solutions to climate change and with a large range of alternatives there is uncertainty in which is the optimum path to take. Another criticism of CBDR is that it effectively excludes countries where there is the greatest growth in emissions and therefore cannot effectively contribute to climate change solutions (Bortscheller 2010). The reasoning for excluding these countries is because it is expected that by reducing emissions it would negatively and unfairly affect their economy.

The CBDR bears relevance to DACS as this policy attributes funding responsibility to different countries, for use to fund climate change measures, of which DACS is capable of doing. Despite this, it does not include a method to determine which technologies it should fund. CBDR can be thought of as back-end funding, while funding of technology can be thought of as front-end funding.

This results in a funding pot available for funding climate change technology. It also includes funding from private investment which is discussed in section 4.09 private investment into DAC. This is depicted in figure 18: Front-end/Back-end funding. The criteria developed in chapter 5, which is based on the objectives of the policies outlined in this chapter, can be used for DAC companies to attract investment and thought of as a front-end funding mechanism.

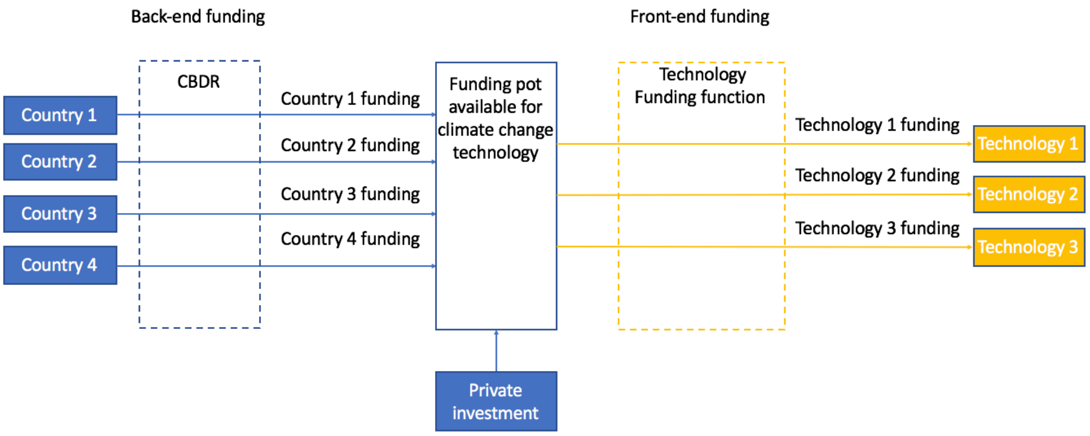


Figure 18: Front-end/back-end funding.

4.2 Clean development mechanism

The Clean Development Mechanism (CDM) is used to give flexibility to industrialised countries in meeting their Kyoto Protocol targets while giving developing countries sustainable development (UNFCC 2019). This is achieved by developing countries implementing projects which result in a reduction in emissions that would have occurred without the project. For every ton of CO_2 that is not emitted that would have, the developing country accrues credits which can be sold to industrialised and contribute to the industrialised countries Kyoto protocol targets (UNFCC 2019). By giving industrialised countries flexibility and developing one sustainable growth it can be seen as increasing equity and fairness between differing countries.

A range of projects exist under the CDM scheme. These include projects such as afforestation and reforestation (UNFCC 2019) which is used to remove CO_2 from the atmosphere and is therefore a CDR are process. DACS is also a CDR a process therefore such a system could be used to fund DACS.

4.3 European union policy objectives

The EU has developed the Energy Union which consists of five objectives (European Commission 2015). The purpose of this section is to understand what objectives drive policy within the EU. In this section the only EU policy that is discussed is the ETS. These objectives can be found with short description below (European Union 2019):

1. Energy security:
   Diversifying Europe's energy sources and more efficient use of energy produced within the EU.
2. The internal energy market:
   Energy to easily flow freely across the EU with no regulatory or technical barriers.
3. Energy efficiency:
   Consumption of less energy.
4. Decarbonisation of the economy:
   Create pressure for global deal for climate change and encourage private investment in new technology and infrastructure.

5. Research and competitiveness:

   Support development of low-carbon technologies through coordination and financing with partnership with private sector.

Objective 4 and 5 relate most closely with the development of DACS as DACS is an emerging technology which can remove emissions. It requires investment, which the EU would like to originate from the private sector.

4.4 European trading system (ETS)

J. Wurzbacher of Climeworks has identified that policies related to carbon pricing can encourage development of DAC (The National Academies of Science, Engineering and Medicine 2018).

The ETS is a carbon pricing mechanism used by the EU. It is a market-based system where there is a set number of credits which large emitters must buy to cover their own emissions. It covers 6% of total global emissions and 45% EU emissions from major emitters such as power stations (European commission 2019). This price range for ranges from almost 0[\$/t-CO_2] to 30[\$/t-CO2] (Market Insider 2019). They can both buy and sell these in a marketplace where the price is dependent on supply and demand. It is implemented across the EU where supply is slowly reduced over time (Haszeldine, et al. 2018). This system does not internalise a mechanism for carbon capture storage; rather it allows emissions with an increasing price of the carbon dioxide credits due a reduction in the supply of credits (Haszeldine, et al. 2018). This therefore has the objective to reduce carbon emissions and due to this contributes to objective 4 of the Energy Union (Decarbonisation of the economy). Furthermore, it does not allow for offsetting of emissions, which DACS can be used for.

The EU has stated that it has the objective to include all emissions sources in the ETS (Dimas 2005). The inclusion of road transport in the ETS has been analysed and it was found that the cost of emissions would need to be (217[\$/t-CO_2]) to achieve the 2030 target of 60[g/km] for cars in 2030 (Pollitt 2014). This was also shown to negatively affect the economy and therefore it is unlikely that in its current form, where emissions cannot be offset, that the ETS will cover all EU emissions.

If this would be adapted to include DACS it would need to be adapted so that it accounts for emissions that are offset. This is because DACS has been shown to be able to offset emissions in one location away from the point source it is accounting for (Keith, Heidel and Cherry 2010).

4.5 Contract for differences

A contract for difference (CfD), in terms of low carbon technology, involves paying the difference between the spot market price and a strike price. If the spot market price is below the strike price the difference is paid by the government. When the spot market price is higher than the strike price the producer pays the difference to the government. It is the UK main mechanism for supporting low-carbon electricity production (Department for Buisness, Energy & Industrial Strategy 2019). CfD has been implemented by the UK government by awarding contracts to low carbon technologies based on the difference between the market price for that product and the additional cost for the low carbon product. This has been deployed in the energy generation sector where energy generation from low carbon sources can ensure a minimum price (Low Carbon Contracts Company 2019). In the UK, funding has been passed onto the consumer by applying a levy on all electricity producers. CfD has been identified as a potential for use with CCUS where the strike price is based on the electricity production and cost of capture of the CO_2 (Department for Buisness, Energy & industrial Strategy 2019).

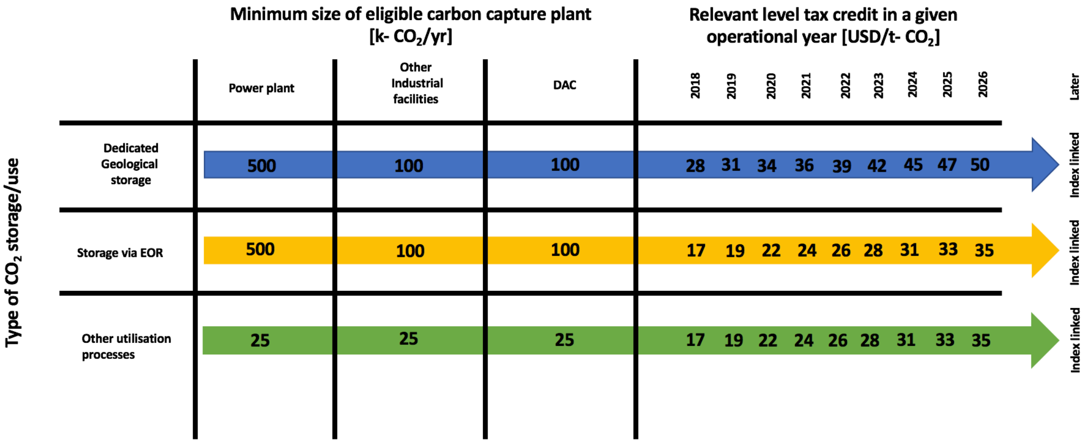
Incorporation of DACS into the electricity production could help this sector remove its emissions from the atmosphere. DACS is incorporated into a Natural Gas (NG) or coal power plant which uses an amine capture system which removes CO_2 from flue gas by using the waste heat from the re-boiler of the stripper of the amine system as the heat source for the DAC system. The DAC system captures CO_2 directly from the atmosphere and is used to top up the CO_2 captured from the flue gas by the amine system. This is described in more detail in sub-section 6.6 Using stripper re-boiler as heat source for de-humidification.

CfD could help the development of DACS in this sector as it is relatively easy to implement. This is because when using CfD the electricity is traded on the spot market and the value of the CfD is attributed at certain points each year. DACS could be included into these CfD where the price for the addition of the DACS system is incorporated into the strike price.

This could be limited as it puts the onus on power generators to reduce CO_2 emissions when other large emission sources originate from other sectors. The levels of emissions per sector is 49.04% for energy and heat production 20.45%, for transport, 19.96% for industry, 8.60% for building and other is 1.95% (WorldBank 2019). This could therefore be deemed unfair by the energy and heat production sector as they are operators removing CO_2 that is not only originating from their sector.

4.6 45Q legislation

The 45Q legislation directly addresses the use of DACS. This is where tax credits will be awarded to carbon capture systems. This includes capture from flue gas and air capture (Waltzer 2018). It would cost the US taxpayer a maximum of 800[$mill] (Bennet 2018). This is a development of previous policy which increased tax credits value and importantly removed the 75 million-ton CO_2 capture cap and allows tax credits to be claimed for 12 years after capture. This reduces uncertainty for developers as they can be more certain of been able to claim the tax credits. Figure 19: US tax breakdown of carbon capture (Bennet, 2018), outlines the 45Q legislation.



Type of CO_2 storage/use	Minimum size of eligible carbon capture plant [k- CO_2/yr]			Relevant level tax credit in a given operational year [USD/t- CO_2]									
	Power plant	Other Industrial facilities	DAC	2018	2019	2020	2021	2022	2023	2024	2025	2026	Later
Dedicated Geological storage	500	100	100	28	31	34	36	39	42	45	47	50	Index linked
Storage via EOR	500	100	100	17	19	22	24	26	28	31	33	35	Index linked
Other utilisation processes	25	25	25	17	19	22	24	26	28	31	33	35	Index linked

Figure 19: US tax breakdown of carbon capture (Bennet 2018).

This policy has yet to implemented as the IRS is developing guidance for attributing credits and have two key questions (Yang, Kennedy en Hausker 2019):

1. What kinds of carbon storage should qualify?
2. What happens if carbon escapes from previously secure storage?

The first question relates to the framework used to determine wether storage qualifies for the tax credits (Yang, Kennedy en Hausker 2019). The ISO has created a framework on geological storage (ISO CCS Storage working group 2017). This is designed to aid development on policy regarding CO_2 capture in differing regulation context (ISO CCS Storage working group 2017).

The second question pertains to how the stored CO_2 is monitored. This is because after the initial storage of CO_2 it should be monitored to see if it is still consistent with the 45Q legislation. The legislation states that if any CO_2 that was initially stored is no longer consistent with the 45Q legislation then additional CO_2 should be captured (Yang, Kennedy en Hausker 2019). The US Internal Revenue Service (IRS) has requested that the circumstances that would trigger this should be defined.

A potential solution which would not require re-capture of CO_2 is to have a buffer account which all receivers of the tax credit have to contribute to which they would not receive tax credits for. A similar mechanism is used in the Californian Carbon Capture and Sequestration Protocol where is a buffer account is used to account for CO_2 that is expected to be no longer compliant with the legislation (California Air Resource Board 2018).

The purpose of the 45Q legislation was to replace the use of natural CO_2 with anthropological CO_2 in the use of Enhanced Oil Recovery (EOR). This was then expanded to include CO_2 storage and CO_2 utilisation. Therefore, the objective of this policy is to increase CO_2 storage and utilisation of CO_2 (Latham and Watkin 2019).

The 45Q legislation is designed to directly affect the development of DACS. The only commercially available DAC system costs 600[\$/t-CO_2] (Climeworks 2020), this does not include the cost of storage. While the 45Q legislation currently applies a tax credit of 34[\$/t-CO_2] which will increase to 50[\$/t-CO_2] in 2026. This disparity means that until the cost of capture and storage falls below the value of the tax credit then it cannot be expected to encourage DACS and therefore is limited.

4.7 Private investment into DAC

J. Wurzbacher from Climeworks also identified that the economics regarding DAC has hindered its development (The National Academies of Science, Engineering and Medicine 2018). Therefore, for private investment a low cost for the system can be deemed an important objective.

DAC funding has primarily come from private investors which have funded the Companies Climeworks, Carbon Engineering and Global Thermostat to the tune of 30.8[\$Mill], 68[\$Mill] and 42[\$Mill] respectively (Hook 2019). This comparatively small when compared to other carbon capture investments which are outlined in Figure 20: Breakdown of carbon capture investment by sector (Bennet, 2018).

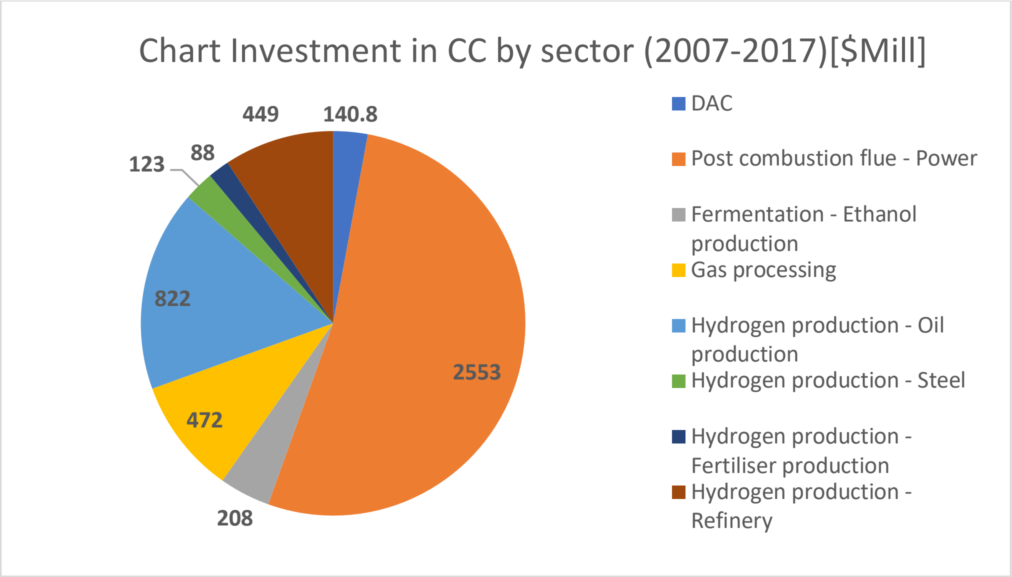


Figure 20: Breakdown of carbon capture investment by sector (Bennet 2018).

Other carbon capture processes are far more developed than DACS with a lower level of uncertainty and therefore a more reliable comparison can be made between different technologies. This makes them a more attractive investment than DACS for investors as there is lower risk involved.

To attract further investment from investors it should be possible for different DACS systems to be compared, and helping investors to select a DACS system that is most appropriate for them.

4.8 Policy recommendations

During this research there were no policy recommendations found regarding DACS specifically. Though policy recommendations regarding the CO_2 value chain are available. Therefore, the recommendations which relate to CO_2 storage value chain are discussed in this sub-section. These are categorised in table 6: Policy recommendations that relate to DAC and CO_2 storage (IOGP 2019), and described below.

Tax incentives, such as the 45Q legislation, can create economic value for DACS and therefore make it more attractive to investors, increasing market uptake of the process. For DACS there needs to be a storage option available and by funding of exploration and appraisal CO_2 storage locations this can support market development. The carbon capture value chain has been identified as a potential risk (IOGP 2019). The value chain consists of CO_2 capture, transport and storage. Particularly in the early stages of research and development failure in one part of the chain can result in a lack of development in the overall system. A DAC unit can be located directly at the point of storage. This therefore somewhat reduces the value chain risk as there is no need for transportation and therefore reduces risk for market uptake. This is not true for system proposed where the DAC system is incorporated into a carbon capture system at a NG or coal power plant, as this will still require transportation to the storage location. DAC and CO_2 storage can be decomposed into their distinct sub-systems; which if they fail will negatively affect the development of the other sub-system. It has been identified these systems should be disaggregated in terms of funding. This would de-risking the value chain (IOGP 2019) increasing market uptake for DACS.

DAC represents the starting point for the CO_2 supply chain and therefore downtime in downstream segments in the supply chain pose a risk to DAC companies. These downstream stakeholders include CO_2 storage. A contractual obligation should therefore be made by downstream stakeholders to be liable for any losses due to downtime on their part. The risks that DAC represent for downstream stakeholders can be minimised by having mandatory CO_2 stored in case of upstream downtime (IOGP 2019). This can therefore help support storage development. This can also be supported within the EU by member states sharing information about storage location.

Public support can be created by ensuring that projects are eligible across all stages of development. This is because if a project has to stop at a later stage of development then it could be deemed a waste of funds by the public and therefore not be supported. If a project is identified as not tackling climate change then it would not be meeting the objectives set out by the public and therefore would not be supported. Within the EU if member states create a climate change strategy and projects are designed to contribute to this then it is more likely to be supported by the public.

Value chain node	**Policy**
Market uptake	• Tax incentives • Funding of exploration and appraisal CO_2 storage locations • Absorbing risk due to physical CCS value chain
Storage	• Clarify liabilities for downstream components of value chain for upstream economic loss due to downstream down time. • Information sharing between member states on potential infrastructure.
Public support	• Ensure that projects are eligible across all stages of development. • Ensure projects are recognised as activities contributing to tackling climate change. • Ensure member states consider deployment strategies.

Table 6: Policy recommendations that relate to DAC and CO_2 storage (IOGP 2019).

4.9 Summary

The methods used to develop climate change technology are outlined in table 7: Description of methods analysed further in this research. The CBDR is not included in further research. This is because CBDR is designed to distribute funding responsibilities to different countries. In terms of policy, this research is focussed on measuring how methods will affect DACS development and therefore it does not concern how to distribute funding responsibilities. The research includes the objectives that relate to DACS. Objectives that are not related to DACS are not included.

Method to develop climate change technology	ETS	45Q	CDM	CfD	Private investment
Objective	Reduce CO_2 emissions	Support CO_2 storage sourced from atmosphere	flexibility to industrialised countries in meeting their Kyoto Protocol targets	supporting low-carbon electricity production	Low cost

Table 7: Description of methods analysed further in this research.

Chapter 5: Criteria definition

This chapter uses the policies outlined in chapter 4 to develop an objectives tree which is operationalised into measurable criteria. This can then be used to measure the technologies designed in chapter 6 and 7.

The operationalised objectives tree uses the methods outlined in (Enserink, et al. 2010). The method used here has similarities to multiple-criteria decision-making (MCDM), which is a method used to analyse options on a set of criteria, which can be conflicting, in a decision-making process. MCDM is widely used to make decisions regarding large complex infrastructure projects such as the sustainability and performance for nuclear power options where technical, social, environmental and economic concerns are taken into consideration (IAEA 2019).

5.1 Means-Ends diagram

This section considers the objectives of policies to determine the upper part of the means-end diagram where the lower part is made up of these policies which is the means to achieve these objectives. This also considers public acceptance of DACS. For this general concern for carbon capture storage is used and concerns that cross over to DAC is analysed.

5.1.1 Method to develop climate change technology

The upper part of the means-end diagram is determined by examining the goal of most significant climate change means. These are detailed in table 8: Determination of upper part of means-end diagram, with the associated goal of these policies. A detailed description of these can be found in chapter 4.

Method to develop climate change technology	ETS	45Q	CDM	CfD	Private investment
Objective	Reduce CO_2 emissions	Support CO_2 storage sourced from atmosphere	Flexibility for industrialised countries in meeting their Kyoto Protocol	Supporting low-carbon electricity production	Low cost

Table 8: Determination of upper part of means-end diagram.

Considering the policy goal, all policies relate to a **reduction in CO_2 emissions**. This can therefore be defined as the primary objective.

DAC companies would like to increase the use of their product therefore **increase of DAC** is a relevant means to achieve the primary objective.

The 45Q legislation promises a price for CO_2 captured via DAC used for storage or utilisation. Therefore, both creation of CO_2 market and **introduce long-term storage of CO_2** are relevant means to achieve the primary objective. By promising a price for CO_2 captured via DAC then it **reduces level of price uncertainty** and this an important means. Utilisation of CO_2 is deemed out of scope in this thesis, so is not included as an objective.

The CDM and CfD relate to funding of measure in which to tackle climate change. The CDM does this by having governments pay for climate change projects while CfD has consumers pay for this The CDM allows for emission reduction measures to be implemented where it is cheapest (Grubb World Economics). This reduces public finance requirements and it can be deemed fairer as funds are available for other projects. A CfD passes on the cost to energy consumer, with the energy sector being significantly larger than other sectors in terms of the emissions it creates (WorldBank 2019), it is therefore assumed that those who consume the most energy emit the most CO_2. This can therefore be deemed fair as those who emit the most pay the most towards green technologies through a CfD. From the CDM fairness in relation to DACS can be deemed to be defined as a system having low public finance requirement. From the CfD fairness for DACS be defined as a system which is funded by those emit the most. Furthermore, **increased fairness on funding emission reduction** can be deemed as a means to reduction in CO_2 emissions.

Private investors would like to see a return on their investment and governments would like to minimise public expenditure. Since only DACS is considered, and not utilisation where the CO_2 has no market economic value then a system which is cheap to build and operate is relevant to private investment and public finance. This means **low cost of system** is a relevant means to reduce CO_2 emissions.

These means are bullet pointed below. This is then followed by a description of how these means affect DAC development and how public acceptance can affect DAC development.

- Reduction in CO_2 emissions.
- Increase DAC use
- Introduce long-term storage of CO_2
- Increased fairness on funding emission reduction.
- low cost of system

5.1.1.1 ETS

The ETS carried out within the EU is the largest carbon pricing system, covering 6% total global emissions (European commission 2019). This analysis therefore considers how to the ETS and DACS can be incorporated.

The offsetting of emissions via DACS has been and was called “Physical Carbon Arbitrage” (Keith, Heidel and Cherry 2010). This exploits the ubiquitous nature of atmospheric carbon dioxide where the emission source and capture point do not need to be in the same location and therefore means it could be incorporated into the ETS if it is adapted where offset emissions are accounted for.

This analysis only considers the expansion of the ETS to more industrial processes. This is because the purpose of the inclusion of the ETS into the analysis is consider what affect the removal of CO_2 which does not include offsetting or abatement has on DACS.

5.1.1.2 CDM

The European Science Advisory Council states that there is no silver bullet technology available which can achieve effective reduction of CO_2 emissions (European Academies Science Advisory Council 2018). This is reflected in the variety of different projects using the CDM with 66 afforestation and reforestation projects out of a total 8169 projects (UNFCC 2019). Therefore, DACS could be incorporated as a CDM projects, though it would be competing with other projects which are cheaper.

The IPCC state that to achieve the 1.5-degree Celsius target then negative emissions will need to occur (IPCC 2018). This will need to be done via permanently removing CO_2 from the atmosphere and in accordance with the criteria set out by (Ramirez and Tanzer 2019) which is described in chapter 1. The CDM applies in relation to the Kyoto protocol targets. These targets still allow for emissions but at a reduced rate for industrialised countries (UNFCC 2019). Therefore, if the CDM was to be adapted to contribute to the 1.5-degree Celsius target set by the Paris agreement and an overshoot occurs then the total emissions allowed via the CDM would need to be negative. The CDM ran until 2020 and there is discussion as to whether it should be continued and if so how (UNFCC 2019).

This analysis assumes that the CDM is continued. Though it is adapted so that the total CO_2 emissions is further reduced and processes such as DACS which can be used to reduce emissions can still be used to achieve the new lower emission target though offsetting. Under the scenario where negative emissions are required due to overshoot then the CDM reflects this with CO_2 emissions targets that are below zero and that DACS is measured against the negative emission criteria (Ramirez and Tanzer 2019), can also contribute to these targets.

5.1.1.3 CfD

CfD has been identified as a potential for use with CCS where the strike price is based on the electricity production and cost of capture of the CO_2 (Department for Buisness, Energy & industrial Strategy 2019). An amine carbon capture system from flue gas has been shown to capture up to 90-95% of the CO_2 that would have been released (Knudsen, et al. 2009). This still leaves 5-10% emitted. Therefore, this system alone cannot achieve net-zero or negative emissions.

In section 6.6 Using stripper re-boiler as heat source for de-humidification, a system is described where an amine carbon capture system capture CO_2 from flue gas and the waste thermal energy is used by a humidity swing which captures CO_2 from the atmosphere. This is able to top up the CO_2 captured from flue gas by the amine system. The calculation shown in that section showed that 8% extra CO_2 was captured with the use of the humidity swing system than without. Though this analysis used an amine carbon capture system which removed 75% of the CO_2 from the flue gas. Therefore, further work should examine how much extra CO_2 can be captured from an amine system which captures 90-95% of the CO_2 from the flue gas.

Such a system could be funded by CfD where the strike price is based on the amine system, humidity swing system, transportation system and storage system. This would therefore fund this CO_2 supply chain and pass this onto the consumer of electricity. Example of CfD use is the conversion of coal power to BECCS plants within the UK (European Commision 2016).

Furthermore, current prediction put energy production from coal and lignite, petroleum products, natural gas, derived gasses and biomass at 7%, 1%, 18%, 1% and 8% respectively in 2050 (European Commission 2013). These energy production processes will require to be accounted for in some manner. The system described in the previous paragraph can do this and can be founded by a CfD.

5.1.1.4 45Q legislation

The 45Q legislation, discussed in section 4.05 45Q legislation, is applies tax credits for CO_2 that is geologically stored (Yang, Kennedy en Hausker 2019). It is designed so that by 2026 a tax credit of value 50[\$/t-CO_2] for geological storage, 35[\$/t-CO_2] to EOR or other utilisation process (Bennet 2018). Though current price of capture is 600[\$/t-CO_2] (Climeworks 2020). In section 4.06 45Q legislation, it was described how this legislation cannot encourage DACS until it applies a tax credit which is of a higher value than the cost of DACS. Therefore, for further analysis it is assumed that the tax credit is equal to at least the price of DAC plus the storage of CO_2, while the utilisation option is not included.

5.1.1.5 Public acceptance

Here, an outline of the public acceptance of DACS is considered. For this, only general concerns regarding CO_2 storage are examined. This is because there has not been a thorough specific analysis of the public acceptance of DAC or DACS. Crossover public concerns from CO_2 storage and DAC are considered. There are more specific views expressed by those in differing economic and demographic cohorts, environmental viewpoints and, countries. The factors discussed here are those which are at a high level of aggregation and can be generalised to CO_2 storage and extended to DACS.

It has been identified that there is a low public awareness of CO_2 storage (UK Energy Research center 2012). With DACS being a much newer and novel technology it can be expected that this factor is more extreme for DACS than it is for CO_2 storage alone. It has been identified that public engagement can increase the awareness of carbon capture storage and address concerns (Devine-Wright 2010). There a relevant factor contributing to the acceptance of DAC can expected to be a high level of public engagement.

The risk that leaks will cause to the environment has been identified as a general concern held by the public (Whitmarsh, Xenias and Jones 2019). In terms of DAC additional risks to the environment therefore should be considered. These environmental concerns could stem from potential pollution from the process, and concerns regarding the plant itself such as footprint area or the fact that it would be processing large volumes of air.

Other general concerns relate to safety of the process itself (UK Energy Research center 2012). These concerns are naturally addressed through the design of the system and are focused on minimising or removing the use dangerous processes or chemicals. This is often combined with having multiple layers protecting people and the environment.

The following bullet points summarise the generalised factors that contribute to a high level of public acceptance.

- **Increased public engagement in concerns/risk/benefit of technology.**
- **Identify and monitor consequences on the environment.**
- **Reduce use of and have multiple layers of protection from dangerous processes and chemicals.**
- **Reduce pollution levels for the process.**

5.1.2 Mean-end diagram

This analysis has resulted in the means-end diagram found in figure 21: Means end diagram for reducing CO_2 emissions. This shows how the means in which achieve a reduction in CO_2 emissions via DAC.

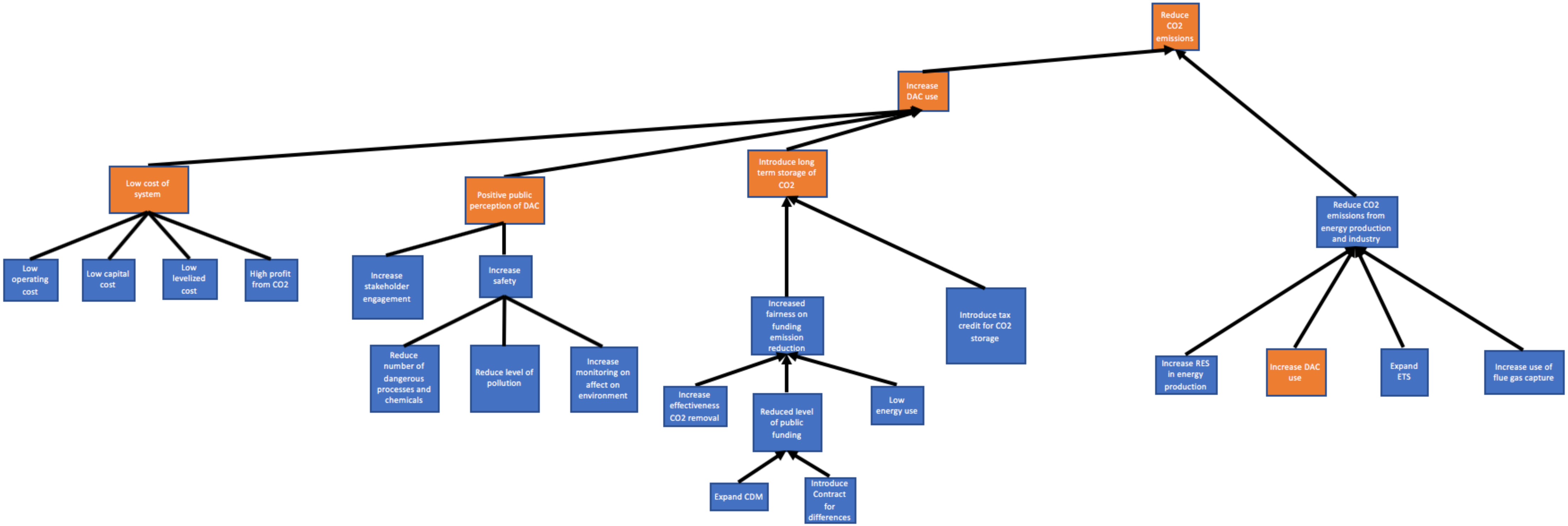


Figure 21: Means end diagram for reducing CO_2 emissions

5.2 Operationalised objectives tree

The concept of back-end and front-end funding was discussed in sub-section 4.01 Common but differentiated responsibilities. This described how back-end funding was determined through CBDR which, plus private investment, results in a funding pot for climate change technology. It also outlined that there was no formal method to attribute funding to different technologies. The purpose of this sub-section is to develop an operationalised objectives tree. This is done by categorise the KPIs outlined in table 5: Technical KPIs, into different objectives. These objectives are given in table 8: Determination of upper part of means-end diagram.

The KPIs outlined in table 5: Technical KPIs, are defined as criteria. Criteria are noun phrases that refers to a specific system property (Enserink, et al. 2010). An objective is defined as a noun phrase that indicates a desired change (Enserink, et al. 2010).

5.2.1 Increased fairness of financial funding of climate change measures

A low level of energy use per ton of CO_2 can contribute to a fair level of funding. Fair level of funding was defined in 5.1.1 Method to develop climate change technology. This is because if it has low energy requirements then there is less financial burden to generate that energy. This will make the system more attractive to private investors as they can use these funds for other matters. Furthermore, if the energy source is waste heat by using less it frees this energy to be used by other processes. This relevant for the humidity swing system as it uses low heat which means it can use waste heat from other processes. The low level of public investment requirement also contributes to fairness of financial funding of climate change measures as this is collected across the population and regardless of how much they are responsible for climate change.

5.2.2 Low emissions associated with DAC

Throughout the supply chain DACS emissions are released and captured. Analysis of the total supply chain is called cradle to grave analysis (Ramirez en Tanzer 2019). For this it is important to analyse the different components of the system. The focus in this research is the DACS system. Therefore, an important criterion is the net atmospheric CO_2 removed via capture by this system. For this it is assumed that all the atmospheric CO_2 is stored. In the analysis carried out in this thesis this included the CO_2 from the energy production required for the process. Other CO_2 sources include pumping to storage location if this is required, pumping for geological storage and from building the plant. For CO_2 utilisation then the criterion would be adapted for net CO_2 for the whole process and would require a full cradle to grave analysis of the proposed system.

5.2.3 Low level of investment requirements

The purpose of this criteria is to analyse the options and the likelihood of attracting private investment. Examples of DAC processes that are currently implemented such as Climeworks and Carbon Engineering exist in the private sector. This is because this is where the expertise is. Therefore, it can be expected that this will continue to develop in the private sector. To attract private investment the funding requirements should be low. The criteria in which this is measured is OPEX, CAPEX and levelised cost. The profit of created through the capture of the CO_2 with associated downstream processes is deemed out of scope for this analysis.

5.2.4 Technical criteria

The technical KPIs collected in chapter 3 are categorised as criteria contributing to relevant objectives for development of DAC in table 9: Categorising KPIs as per objective.

Technical KPIs / Criteria	OPEX	CAPEX	levelised cost of product (In this case per ton of CO_2)	energy requirements	CO_2 emissions from the plant
Objective	Low level of investment requirements			Increased fairness of financial funding of climate change measures	Low emissions associated with DAC

Table 9: Categorising KPIs as per Categorising KPIs as per objective.

5.2.5 Scope of front-end funding

Boxes marked in red are deemed out of scope of this research. A High level of stakeholder engagement is out of scope as this research does not have the resource to address meaningful stakeholder engagement. Despite this, chapter 9: stakeholder analysis can be used as a starting point. The high level of safety criterion set is also deemed out of scope. In chapter 6 and 7 the most significant contributors to this criterion are identified with a brief description. No specific modelling or system design is proposed therefore this criterion is deemed out of scope. The profit from carbon dioxide product is also out of scope as it would require a full economic analysis and LCA of the complete supply chain of CO_2 utilisation/storage process, while this research focus is on the DAC system itself.

The technical criteria, which is low energy use [MW/t-CO_2], OPEX [$/year], CAPEX [$], levelised cost of CO_2 capture [$/t-$CO_2$] and low emissions associated with DAC [kg-CO_2/year]$_{plant}$, are measured against both DAC systems in chapter 6 and 7. In chapter 8 low public finance required [$] where the affect different polices will have on this analysed. In chapter 9 the stakeholder analysis is considered separately from the criteria developed in this chapter.

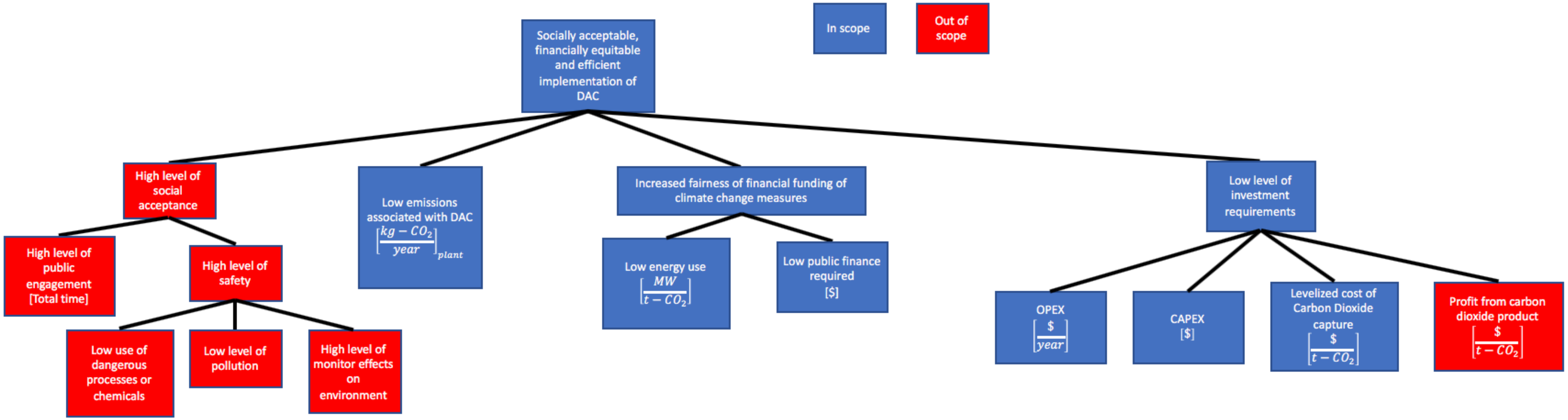


Figure 22: Definition of criteria using objectives tree.

5.3 Summary

This chapter used the methods in which to develop climate change technology outlined in chapter 4. This uses the objective of these methods which are explored in terms of how they can contribute to the primary objective which is a reduction in CO_2 emissions. In addition to the methods outlined in chapter 4 the contribution that public acceptance of a technology can have is explored. From this a means-end diagram is developed which can be used as a roadmap in which to achieve the primary objective.

An objectives tree is then used to define specifically what is needed to achieve the primary objective. For this the KPIs identified in chapter 3 are categorised as contributing to different objective which contribute to the primary objective. In addition to these, social acceptance, direct energy use and profit from CO_2 product were identified as contributing to the primary objective. Social acceptance and profit from CO_2 product were deemed beyond the scope of this thesis due to the increased complexity this would add. These were then operationalised by defining the units in which these are measured.

Chapter 6: Technical analysis of the humidity swing

The humidity swing was chosen for further analysis as it is the most poorly understood system. This chapter will analyse this system in terms of the criteria in figure 23: Chapter 6 criteria. The results of this are the output of this chapter.

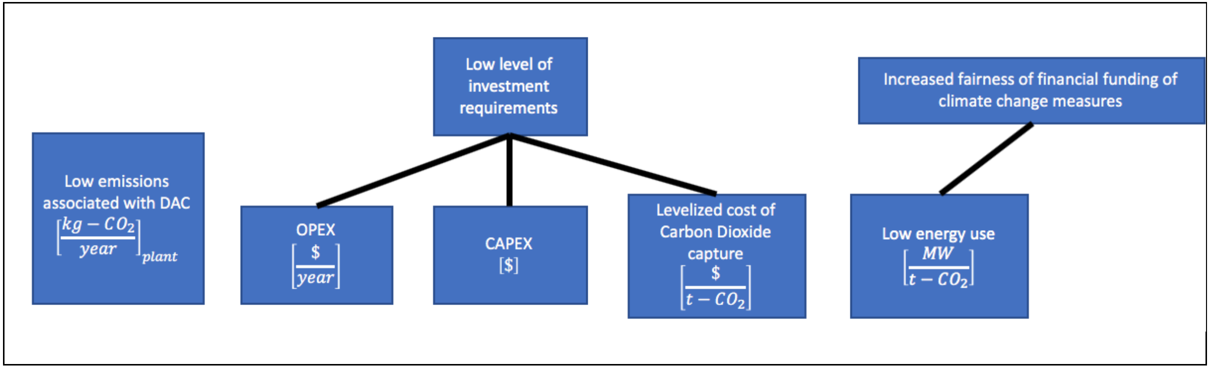


Figure 23: Chapter 6 criteria

6.1 System design

The system is split in three sub-systems: solar thermal, humidity swing and the amine sub-system. The air capture system has air which is pumped through a humidity swing process. The humidity swing is based on the results from the experimentation carried out and described by (Wang, Lackner & Wright 2011). The inlet concentration of CO_2 is 400 ppm, when the CO_2 is released from the sorbent it is at a partial pressure of 5%. This is then passed to a conventional amine carbon capture amine system which produces CO_2 at a mass concertation of 97%. The main energy requirements are from drying of the sorbent in the humidity swing and the stripper reboiler. The energy for these is supplied by a thermal solar energy system. Figure 24: High level description of humidity swing system, depicts a high-level description of this system. The system was designed to produce the same amount of CO_2 as (Keith, Holmes, et al. 2018) which is 166.08[t-CO_2/Hr] and mass fraction of 97%.

The system is based on a similar design proposed in (Zhai, et al. 2018). In this process a thermal solar system is used for the generation of energy used by the stripper re-boiler in an amine carbon capture system to capture flue gas from a coal powered power plant. The adaption in the proposed system is that there is no flue gas and that all the inlet flow rate for the amine system comes from the humidity swing system. Furthermore, there is no power plant and all the thermal energy for the humidity swing system and stripper re-boiler comes the solar thermal energy system. This is described in figure 24: High level description of humidity swing system. This includes the mass fraction of CO_2.

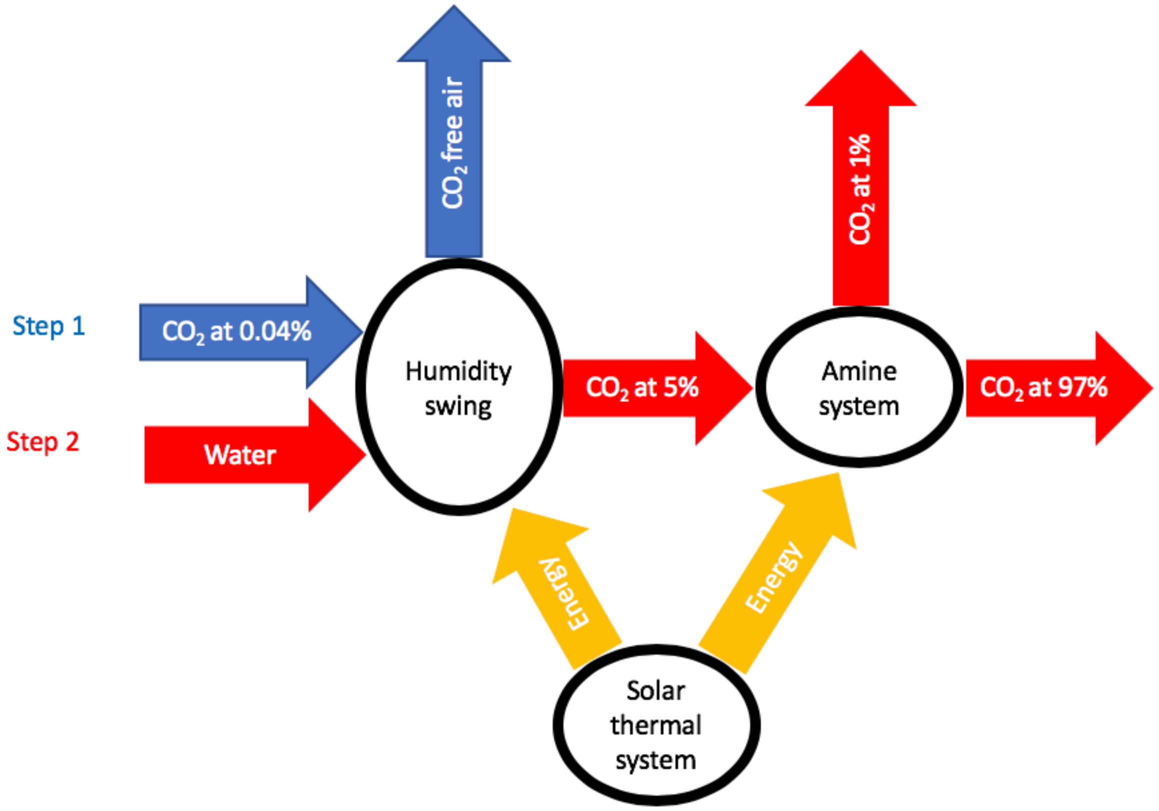


Figure 24: High level description of humidity swing system.

6.2 Humidity swing sub-system

The amine system was designed to have a capture rate of 74.63% of CO_2. This means that 222.54[t-CO_2/hr] have to be supplied to it from the humidity swing sub-system to match the outlet flow rate of the HT solvent system which is 166.08[t-CO_2/hr]. With a 5% mass fraction of air this is a total flow rate of 4450.8[ton/hr] of air. This is also the volume of water vapour that passes over the sorbent during step 2 of the process. A number of significant uncertainties exist for this system. These are outlined in table 10: Uncertainties for humidity swing sub-system, with a description given below:

Uncertainties
Cost of sorbent
Number of sorbent changes per year
Required time for adsorption/desorption
Ratio of mols of H_2O to CO_2 requirements

Table 10: Uncertainties for humidity swing sub-system

The sorbent can be bought commercially for 5.8[$/kg] (Ecosoft 2020) but costs included in this are packaging, delivery and sales which are not required for the type of system proposed. Therefore, was reduced to 3[$/kg].

Over time the sorbent will degenerate and require replacement. The actual degradation rate is unknown so a sensitivity analysis has been carried out and can be found in figure 25: Relationship between sorbent changes per year and cost per ton of CO_2.

The sorbent was exposed to CO_2 for 24 hours in (Wang, Lackner & Wright 2011). This was so that the sorbent comes to equilibrium with CO_2. It may be possible that the sorbent comes to equilibrium in less time, but experimentation would need to be carried out to determine this. The desorption time in (Wang, Lackner & Wright 2011) was 2 mins though the experiment did not optimise on this parameter. Due to these uncertainties a sensitivity analysis was carried out on these parameters

The ratio of mols of CO_2 to H_2O was assumed to be 1 mol to 1 mol. This is a rather large assumption and is based on the fact that there is 1 mol of CO_2 for every 1 mol of H_2O in the desorption reaction. To determine the actual mol ratio requirements experimentation will need to be carried out. It is hypothesised that this is an over estimation of water requirements and the reasoning for this is given in sub-section 10.3 Future research direction. Therefore, the sensitivity analysis carried out considers a mol ratio for H_2O to CO_2 to from 0.5-1.0

The sensitivity analysis on the affect these uncertainties will have on the cost of CO_2 are shown in figures 25-30. This also has a sensitivity analysis on the capacity of the sorbent as this is expected to change over time as the sorbent degrades. The cost of the sorbent range was selected to include the cost of the sorbent as it can be commercially bought at 5.8[$/kg] (Ecosoft 2020) and the estimated cost if purchased wholesale 3[$/kg]. For the number of sorbent changes per year there are is no information regarding the degradation rate and therefore the range was made very large. For the desorption time and adsorption time there is no information regarding when the sorbent comes into equilibrium to its surroundings and therefore the range for these parameters were also made very large. Despite, this it can be seen that adsorption contributes more to the cost per ton of CO_2. This is because the adsorption time is much larger than the desorption time. The ratio of mols of H_2O to CO_2 range was 0.5-1.0. The lower limit was selected as when water is added water is produced from the desorption reaction. For this it was assumed that if 1 mol of water is added then 1 mol of water is produced from this reaction. The upper limit was selected as it cannot be expected that if this reaction produces water in the presence of water that the requirement would exceed a 1 to 1 mol ratio. The capacity of the sorbent range was selected to see the effect of the sorbent degradation rate and the shape of the curve.

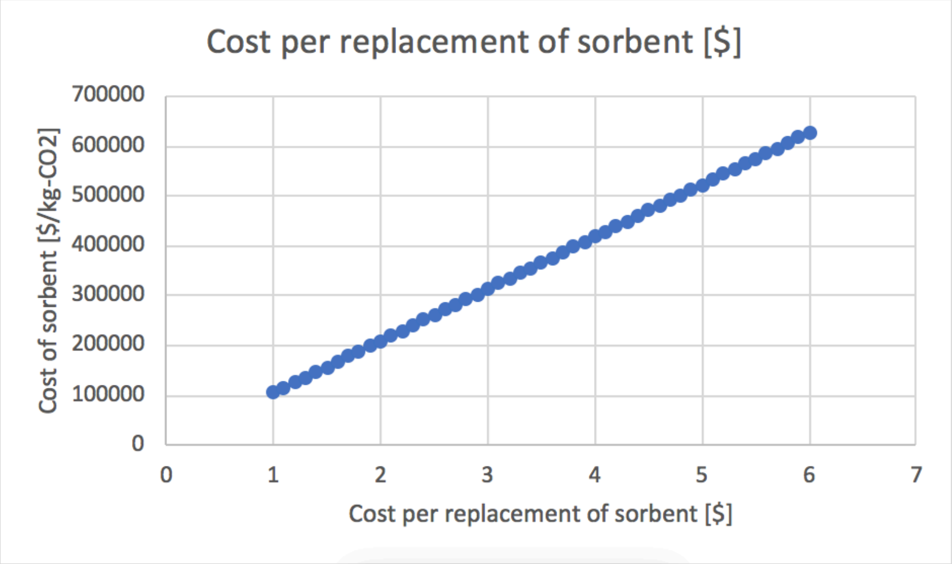


Figure 25: Relationship between cost of sorbent and cost per ton CO_2

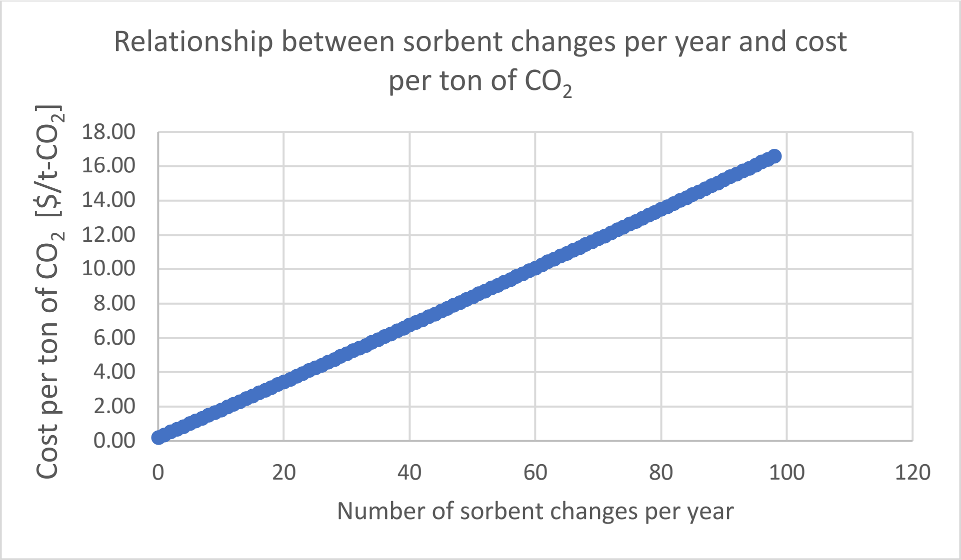


Figure 26: Relationship between sorbent changes per year and cost per ton of CO_2

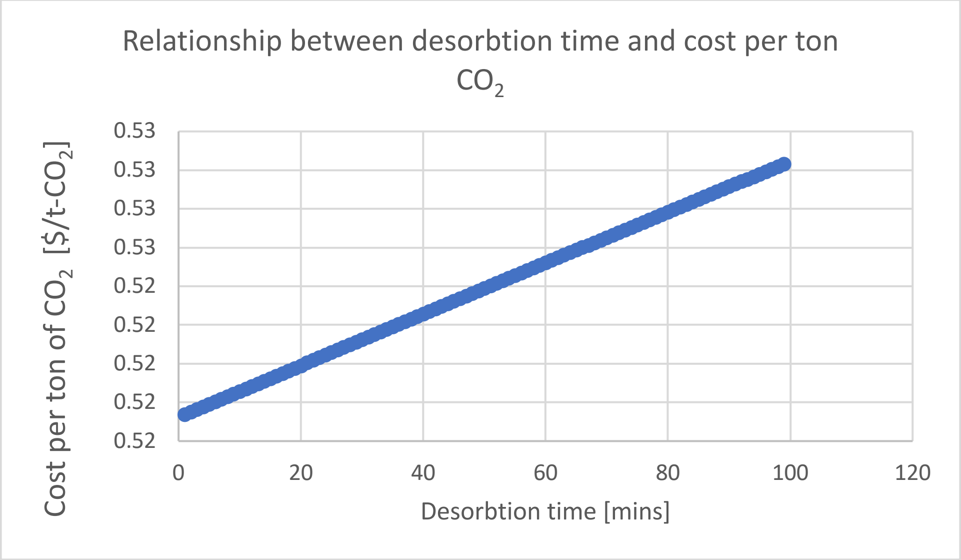


Figure 27: Relationship between desorption time and cost per ton of CO_2

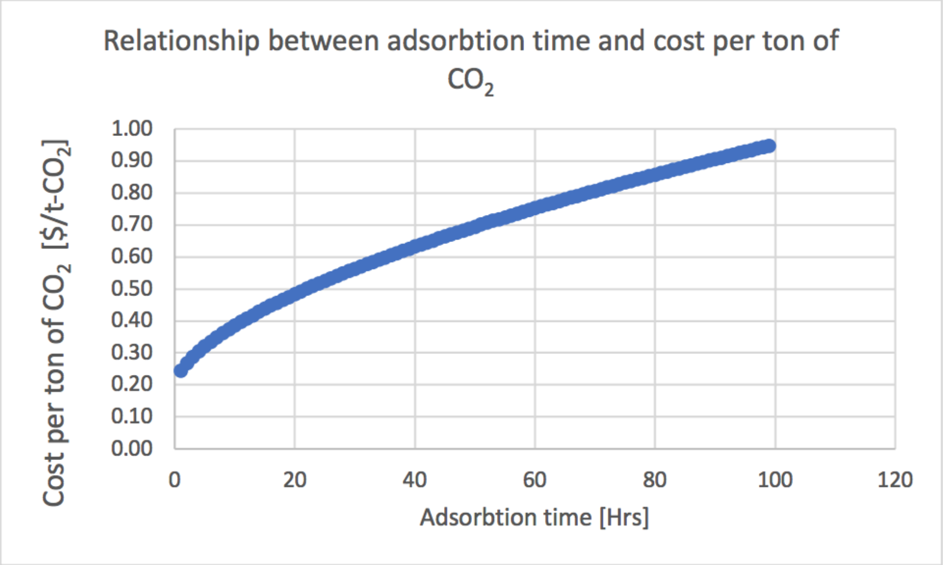


Figure 28: Relationship between adsorption time and cost per ton of CO_2

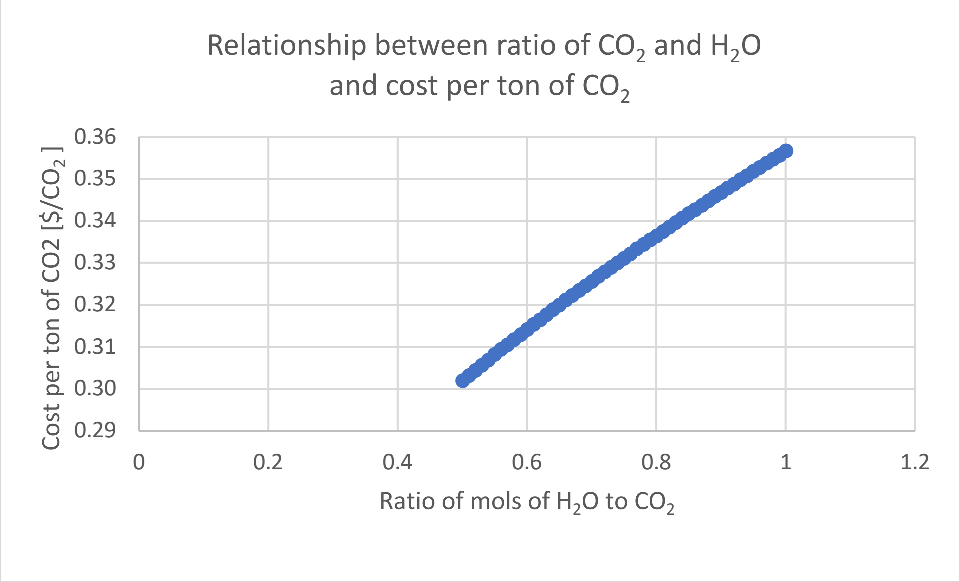


Figure 29: Relationship between adsorption time and cost per ton of CO_2.

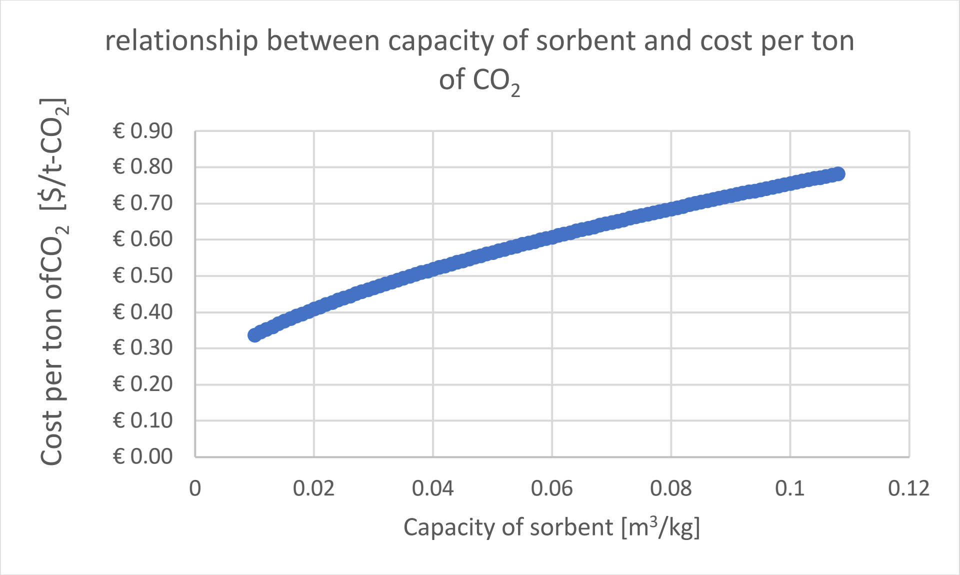


Figure 30: Relationship between capacity of sorbent and cost per ton of CO_2.

The main contributor to the OPEX is the fan energy requirements and the cost of changing the sorbent. Table 11: Levelized and OPEX for humidity swing, outlines the cost and parameters used to calculate this. A more detailed description of this calculation is given in appendix-A and appendix-B.

Table 11: Levelized and OPEX cost for humidity swing sub-system, outlines the values used for the uncertainties that were described in table 10: Uncertainties for humidity swing sub-system. Further to this the cost of electricity is given. This location was selected for the USA as the system is required to be located which has high solar thermal energy. Furthermore, the USA has the 45Q legislation which directly addresses DACS and therefore it can be expected that DACS is considered and option in the USA.

The fan energy requirements is also given in this table 11: Levelized and OPEX for humidity swing. The energy required for this is the same as (Keith, Holmes, et al. 2018) which is 61[kWHr/ton-CO_2]. This value comes from the deltaP across the fan and a 70% efficiency. The fan comes from SPX cooling technologies (SPX 2020).

In Table 11: Levelised and OPEX cost for humidity swing sub-system, the calculated value for levelized cost and OPEX are in bold. The OPEX is based on the production rate of CO_2 of 166.08[$/t-$CO_2$]

Parameter	**Value**
Cost of electricity (U.S. Energy Information Administration 2019)	0.03752 [$/kWHr]
Fan energy requirements (Keith, Holmes, et al. 2018)	61 [kWHr/t-CO_2]
Fan levelised cost	**0.26 [$/t-$CO_2$]**
Adsorption time	24 [hr]
De-adsorption time	0.033 [hr]
CO_2 at STP capacity of ion $CO_3{}^{2-}$	0.0398 [m^3/kg-CO_2]
Cost of sorbent	3 [$/kg-sorbent]
Sorbent changes per year	1
Ratio of mols H_2O to CO_2	1:1
Levelized sorbent cost	**0.36 [$/t-$CO_2$]**
Levelized cost of humidity swing	**0.62 [$/t-$CO_2$]**
OPEX of humidity swing sub-system	**900,000 [$/year]**

Table 11: levelised and OPEX cost for humidity swing sub-swing.

The CAPEX for the humidity swing consists of the sorbent and cost of the pumps used to pump the air. Details of the how the sorbent were costed are outlined in appendix-B. The CAPEX for the sorbent is **263,000[$]**. This is only based on the cost of the sorbent. Other costs are labour and land costs which are not included. The assumptions used to calculate this is the CO_2 at STP capacity of ion $CO_3{}^{2-}$ and Cost of sorbent [$/kg-sorbent] which are shown in table 11: levelised cost for humidity swing sub-swing.

The CAPEX required for the fans is determined from the flow rate. Figure 31: Relationship between pump capacity and purchase cost (Lyons & White, 2002). depicts the relationship between capacity of the fan and purchase cost. The flow rate of air is 4450.8[ton/hr] which at atmospheric conditions is 57375 [gallons/min]. Using the pumps at 70% capacity, 2500 [gallons/min], with one spare this requires 21 pumps. Each pump cost approximately 29,000 [$/pump] with the total cost is 609,000[$] in 2002. Accounting for inflation this would have a value of **719,000[$]** today.

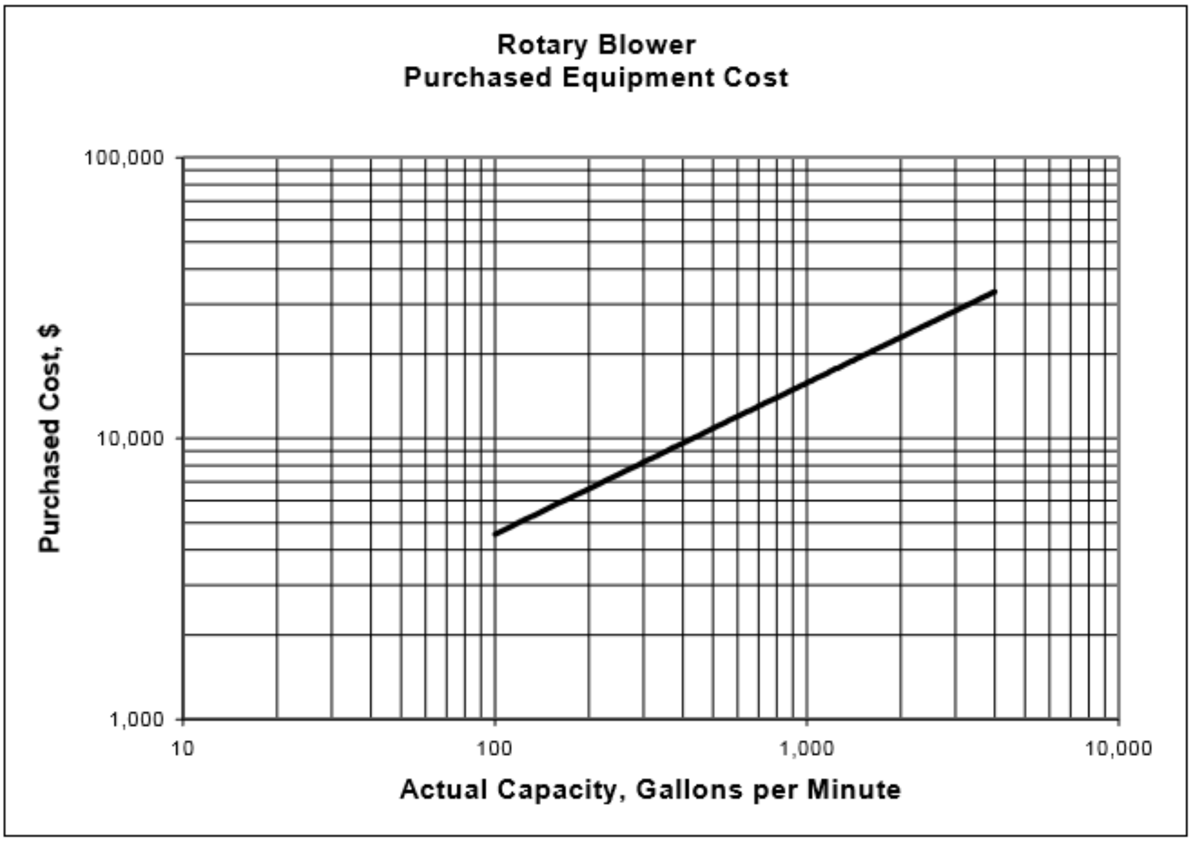


Figure 31: Relationship between pump capacity and purchase cost (Lyons en White 2002).

The breakdown of the CAPEX of the humidity swing sub-system are shown in table 12: Solar thermal sub-system CAPEX breakdown.

	Cost
Sorbent CAPEX	263,000 [$]
Pumping CAPEX	719,000 [$]
Total CAPEX	**982,000 [$]**

table 12: Solar thermal sub-system CAPEX breakdown.

In reality a number of other variables contribute to these values which have not been included in these calculations due to the high level of uncertainty in them. For OPEX the number of sorbent changes per year will is a contributing factor but not included as it is not known what the degradation rate of the sorbent is. For the CAPEX piping, small units and instrumentation have not been included. This is because a full P&ID has not been developed and therefore these are unknowns.

Table 13: Description of humidity swing, describes the humidity swing system. Details of the calculation can be found in appendices-A, B and C.

Inlet flow rate air [t/hr]	Outlet flow rate of CO_2 [t/hr]	Energy requirements for humidity swing [MW]	Volume of sorbent [m^3]	Mass of sorbent [kg]	Cost of Sorbent [$/change]
Input	Input	Output	Output	Output	Output
4451	222.54	6852	158860	173,157,342	263,000

Table 13: Description of humidity swing sub-system

6.3 Amine and solar thermal sub-system

The amine system that is used as a base case processes gas at a rate of 4450.8[t/hr] at a mass fraction of 5% CO_2. The relationship between the energy required by the stripper re-boiler and the inlet flow rate of gas is depicted in Figure 32: Relationship between inlet flow rate and duty of the stripper reboiler, it can be seen that this is non-linear. There is a linear relationship between the inlet flow rate and energy required by the humidity swing process, depicted in figure 33: Relationship between inlet flow rate and energy requirements of the humidity swing. The sum of the re-boiler duty and the duty required for the humidity swing results in the total thermal energy requirements. Figure 34: Relationship between inlet flow rate and total thermal energy requirements, shows that there is linear relationship between these components. This is because the re-boiler duty only makes up a small fraction of the total thermal energy requirements. This can visually be seen and also through the error terms for the fitted line to the data equalling 1. This linear relationship means that when the amine system is scaled it should be linearly scaled.

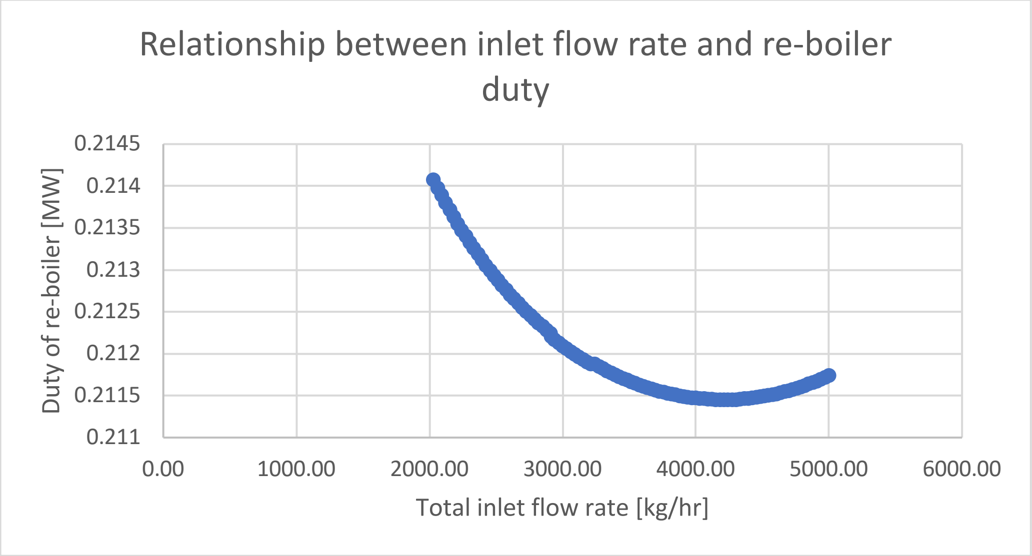


Figure 32: Relationship between inlet flow rate and duty of the stripper reboiler.

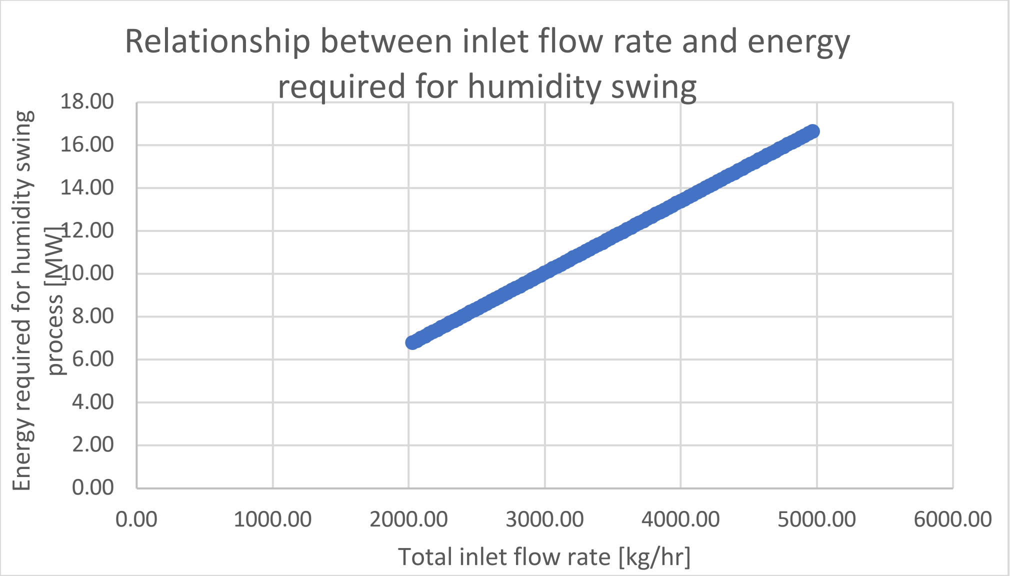


Figure 33: Relationship between inlet flow rate and energy requirements of the humidity swing.

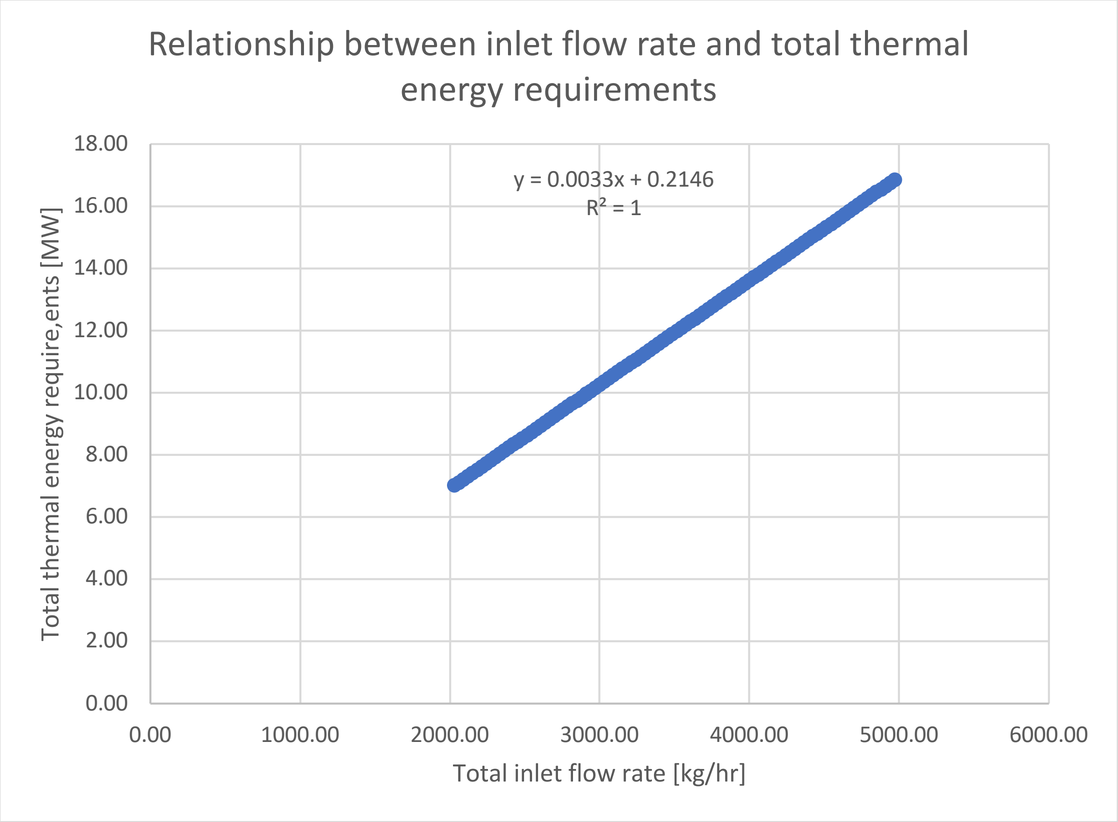


Figure 34: Relationship between inlet flow rate and total thermal energy requirements.

The area required for the solar thermal system is found via the following equation (Zhai, et al. 2018):

$$A_{net} = \frac{E_{reboiler} + E_{humidity\ swing}}{DNI * F_{opt} * k * F_{shading} * F_{end} * F_{wind} * F_{clean}}$$

Table 14: Solar thermal sub-system description, describes these parameters. This uses the same values for DNI, F_{opt}, k, $F_{shading}$, F_{end}, F_{wind}, F_{clean} as (Zhai, et al. 2018). It also defines the parameter. This also calculates the CAPEX of the solar thermal sub-systems. The CAPEX value comes from (Zhai, et al. 2018) and has a value of 308 [\$/m^2].

Parameter	Definition	Value
A_{net}	Total area required by thermal solar system	12,500,000 [m^2]
$E_{reboiler}$	Energy required by reboiler	151[MW]
$E_{humidity\ swing}$	Energy required by humidity swing	6852[MW]
DNI	Direct nominal irradiation	805 [W/m^2]
F_{opt}	Peak optical efficiency	0.73
k	Incident angle correction	1.00
$F_{shading}$	Shading losses	1.00
F_{end}	End loss effect	0.97
F_{wind}	Wind losses	0.99
F_{clean}	Cleanliness losses	1.00

CAPEX	Total capital costs of solar thermal sub-system	3.8[$Bill]

Table 14: Solar thermal sub-system description.

The area required is equivalent to the size of a relatively large city. For example, it the area represents approximately 80% of the metropolitan Glasgow area or around twice the size of the Rotterdam-Hague metropolitan area. Due to the low DNI, which is the solar energy supplied to a unit area (Blanc, et al. 2014), found in Europe and large land area required the process would need to be located outside Europe. Furthermore, such a large system may not be practical in terms of financing and operation. Therefore, smaller systems should be considered if this is the case.

For the calculation shown in Table 14: Solar thermal sub-system description a 1:1 mol ratio of H_2O to CO_2 was used. For the humidity swing sub-system this factor was analysed with a 0.5: 1.0 ratio of H_2O to CO_2. This has then been repeated and the effect on the solar thermal sub-system energy requirements, area and cost of solar thermal sub-system.

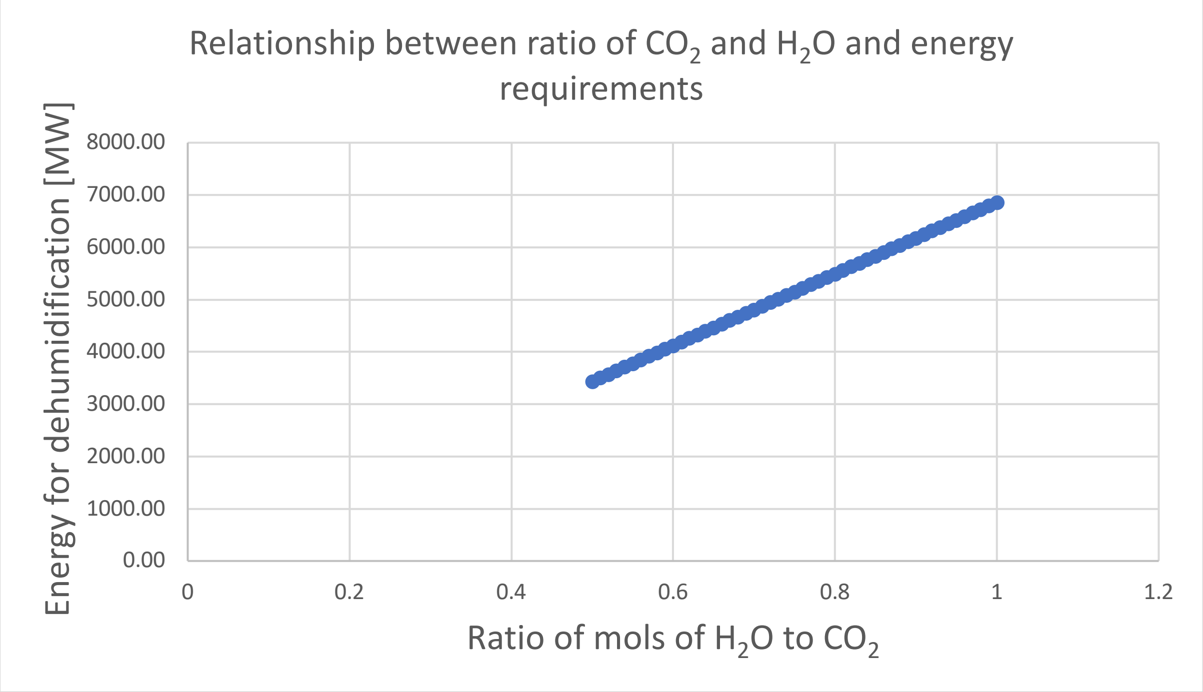


Figure 35: Relationship between ratio of CO_2 and H_2O and energy requirements

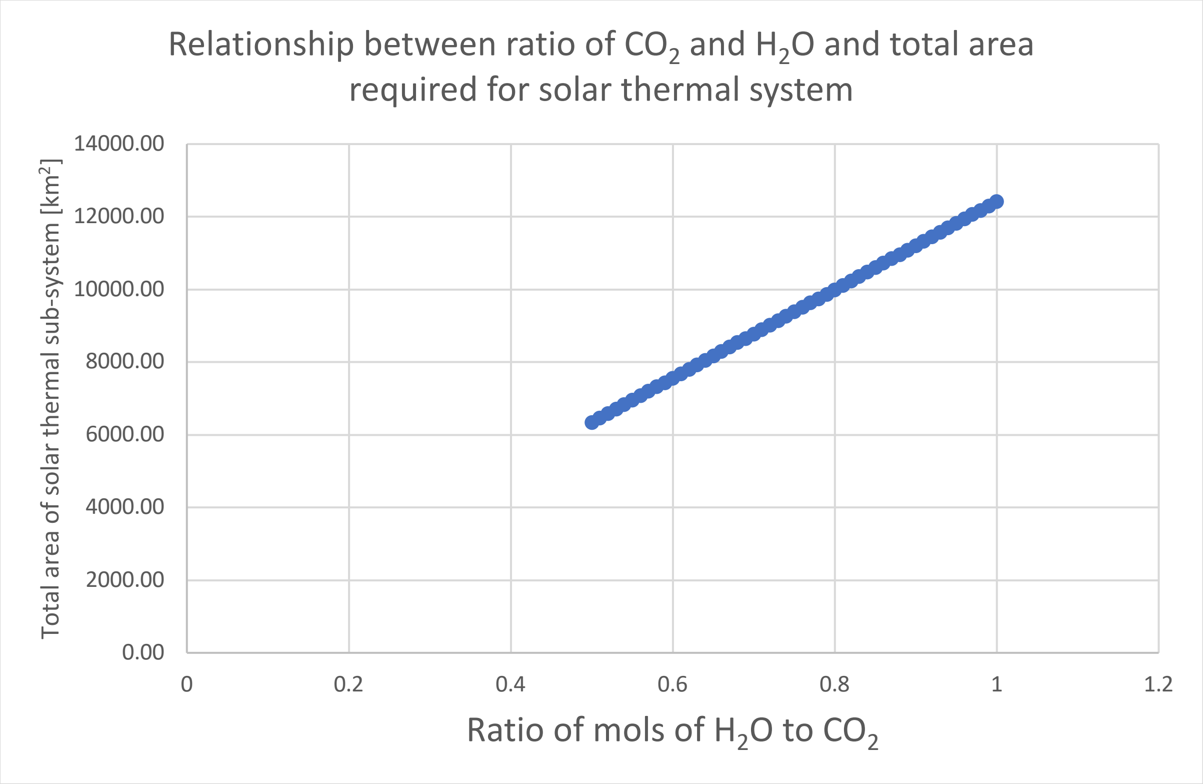


Figure 36: Relationship between ratio of CO2 and H2O and total area required for solar thermal system

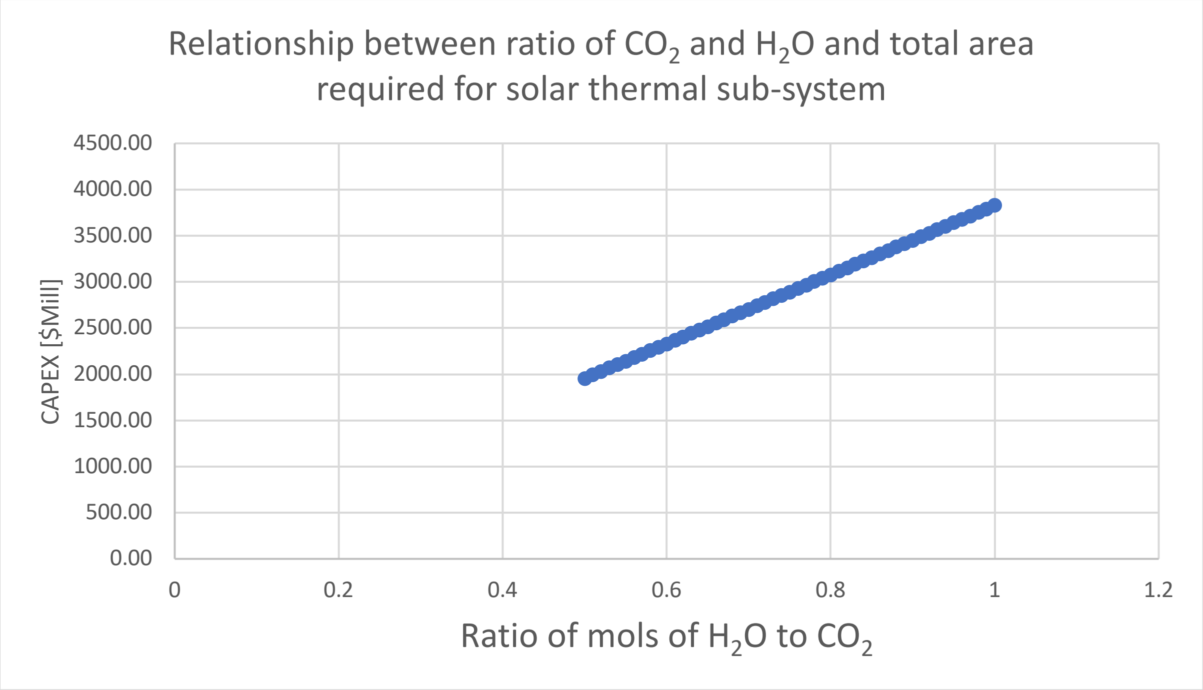


Figure 37: Relationship between ratio of CO_2 and H_2O and total area required for solar thermal system

The solar thermal energy requirements can be seen to be affected by the ratio of H_2O to CO_2 requirements. This is because the H_2O is required to be evaporated and this uses the energy from the solar thermal system. This in turn affects the requirements for the total area of the solar thermal sub-system which affects the CAPEX of this sub-system.

The cost of the solar thermal system was taken from (Zhai, et al. 2018) though uncertainty exists around this value as it has been possible to verify it. Therefore, a sensitivity analysis is carried out in figure 38: Relationship between cost of solar thermal per unit area and CAPEX of solar thermal sub-system.

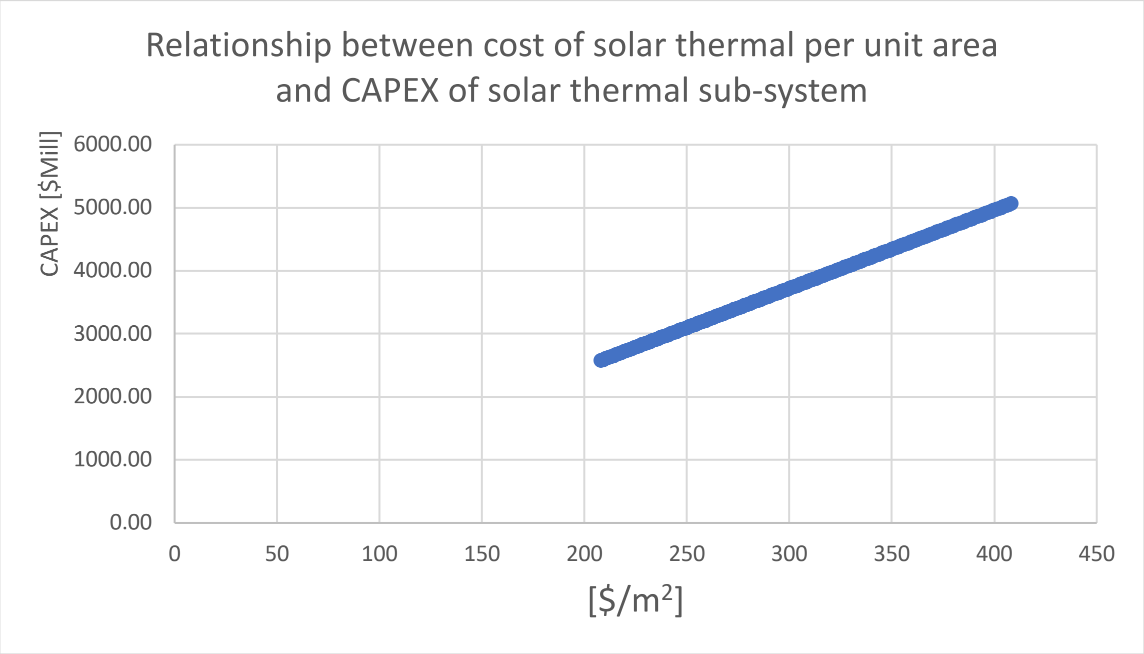


Figure 38: Relationship between cost of solar thermal per unit area and CAPEX of solar thermal sub-system.

Table 15: Levelised cost of electricity production from solar thermal system, describes the variables used in to determine the LCOE. These are also described below.

The levelized cost of electricity (LCOE) from the solar thermal energy sub-system is calculated as shown below (Zhai, et al. 2018). It is the sum of all the costs over the lifetime of the plant divided by the energy produced by the plant.

The total capital cost is based on the CAPEX of the humidity swing sub-system (982,000[$]) plus the humidity swing sub-system (3.8[$Bill]) plus amine sub-system (147[$Mill]).

The operating and maintenance cost come from the yearly operating and maintenance cost of the amine sub-system (135[$Mill]) plus the yearly operating cost of the humidity swing sub-system (0.9[$Mill]) plus the operating and maintenance cost of the solar thermal sub-system which is approximated as zero. For the operating and maintenance cost of the amine sub-system this was determined using Aspen Plus economic analyser. The operating cost of the humidity swing sub-system was taken from the calculations in sub section 6.2 Humidity swing sub-system. The solar thermal sub-system has no fuel requirements and therefore minimal operating costs. The maintenance cost was not included of the humidity swing and solar thermal sub-system sub-system, and this research could benefit from further analysis of this.

The fuel cost is zero as solar thermal energy is free. The interest rate comes from (Zhai, et al. 2018). The USA, where the system has been selected for, has an interest rate of 1.5% (Country Economy 2020) which is very low. As the interest increases the LCOE increases therefore the interest rate has been kept high to protect from estimating that the cost of electricity been low. This is because estimating future interest rates is extremely challenging.

The total energy requirements originate from the total solar thermal energy used which is the energy of the stripper re-boiler (151[MW]) plus the humidity swing energy requirements (6852[MW]). The lifetime of the plant has been taken as a standard lifetime a process plant.

The low LCOE means that for the levelised cost CO_2 and OPEX for this system is approximately zero.

$$LCOE = \frac{TCR * FCF + O\&M + FC}{E_{total}}; FCF = \frac{r(1+r)^t}{(1+r)^t - 1}$$

Variable	Description	Value
$LCOE$	Present value of price of electricity over lifetime of plant	0.07[$/J]
TCR	Total capital cost	4 [$Billion]
FCF	Fixed charge factor	0.09
$O\&M$	Operating and maintenance	136[$Mill]
FC	Fuel cost	$ 0
E_{total}	Energy of stripper reboiler and humidity swing	7003[MW]
r	Interest rate	0.08
t	Lifetime of plant	30[years]

Table 15: Levelized cost of electricity production from solar thermal system.

The amine sub-system uses standard units and therefore the total equipment cost is based on the standard built in units within Aspen Plus. Using Aspen Plus Economic Analysis software the cost of the amine system can be determined. This uses model-based estimations on capital and operating costs of a system (Aspen Plus 2020). The amine carbon capture sub-system uses standard equipment. Therefore using Aspen Plus Economic Analysis costing method was used.

This is then scaled using the 6^{th} 10^{th} rule. Table 16: Cost of amine system, describes the cost for the amine system for the required inlet flow rate of 4450.8[ton/hr]. A PFD taken from Aspen plus can be found in appendix-D.

The main raw material used in the amine carbon capture sub-system is amine which is used to capture the CO_2. Overtime this degrades and periodically has to be changed. This therefore contributes to the total raw material cost. This contribution of this is beyond the scope of this thesis and therefore has been set to zero. Product sales is also deemed out of scope as CO_2 utilisation which could be used for this is not considered in this thesis.

The total utility cost is made up of two utilities. All heating requirements for the humidity swing sub-system comes from the humidity swing which has a levelized cost of 0.07[$/J] and non-thermal energy requirements has a cost of 0.03752 [$/kWHr] (U.S. Energy Information Administration 2019).

Aspen results	Cost
Total Capital Cost	483 [$Mill]
Total Operating Cost	135 [$Mill/year]
Total Raw Materials Cost	0,00 [$/year]
Total Product Sales	0,00 [$/year]
Total Utilities Cost	2 [$Mill/year]
Equipment Cost	56 [$Mill]
Total Installed Cost	147 [$Mill]

Table 16: Cost of amine system.

With operating cost for the amine system being 135[$Mill/year] and a CO_2 production rate of 166.08 [t-CO_2/hr] then this a levelised cost in terms of CO_2 is 92.5 [$/t-$CO_2$]. Comparing this to other system such as carbon captured from NG and coal combustion costing 43-83[$/t-$CO_2$] with associated cost of transport and storage at 7-12[$/t-$CO_2$] (Irlam 2017), it can be seen that the amine system designed in this thesis cost more. The amine sub-system has air passed to it from humidity swing sub-system with a CO_2 concertation of 5%. When fuel for energy production is burned it has a higher concertation of CO_2 in the flue gas (US Energy information administration 2020). This goes some way towards explaining why these systems are cheaper than amine system designed in this thesis. The other systems have also been optimised to be incorporated into the energy production systems over a long period and have therefore been optimised to higher level.

6.4 Low level of investment requirements criteria

Table 17: Summary of OPEX and CAPEX for complete humidity swing system, levelised cost in terms of CO_2 and OPEX for the complete humidity swing system, outlines the CAPEX and OPEX for each of the subsystems. The OPEX is calculated as the plant working at a 100% capacity; though in reality there will be down. The fact that the solar thermal system has almost zero operating means that there is significant savings made. This saving in the operating costs is reflected in the high value CAPEX of the solar thermal sub-system. The cost of storage has not been included as this can be assumed the same for both the humidity swing system and the HT solvent system therefore would not help with the comparison between the systems.

CAPEX	
Humidity swing system	1 [$Million]
Amine system	483 [$Million]
Solar thermal system	3826 [$Million]
Total	**4310 [$Million]**
Levelized cost in terms of CO_2	
Humidity swing system	0.6 [$/t-$CO_2$]
Amine system	92.5 [$/t-$CO_2$]
Solar thermal system	0 [$/t-$CO_2$]
Total	**93.1 [$/t-$CO_2$]**
OPEX	
Humidity swing system	1 [$Mill/year]
Amine	135 [$Mill/year]
Solar thermal system	0 [$Mill/year]
Total	**136 [$Mill/year]**

Table 17: Summary of OPEX and CAPEX for complete humidity swing system.

6.5 Low emissions associated with DAC

The solar thermal system has no direct emissions associated to it. Electricity energy is used for the non-thermal energy requirements of the amine system. This includes aspects such as pumping of material round the system. Electricity energy is used in the non-thermal energy requirements of the amine sub-system. In the humidity swing sub-system the fan also uses electricity energy sourced from the grid. The associated emissions from the electricity production comes calculation using Aspen Plus. This uses the rules and regulations regarding emission reporting in the USA (Enviromental protection agency 2009). This was based on the base case emissions which has an outlet flow rate of CO_2 of 235.74[kg-CO_2/hr] and emissions of 1.73[kg-CO_2/hr]. Scaling this to an outlet flow rate of CO_2 of 166.08[t-CO_2/hr] results in emissions of 1.2[t-CO_2/hr]. This assumes that all CO_2 is captured from the air that is contacted with it. Other emission sources include the emissions associated with energy requirements for storage. This has not been included as this can be assumed to be the same for the humidity swing and HT solvent system as they produce the same amount of CO_2 product therefore would not help with the comparisons between the systems. Other sources include the building. This has also not been included as it has not been designed. It has also been assumed that the DAC plant is at the point of storage and therefore there are no emissions associated with transportation.

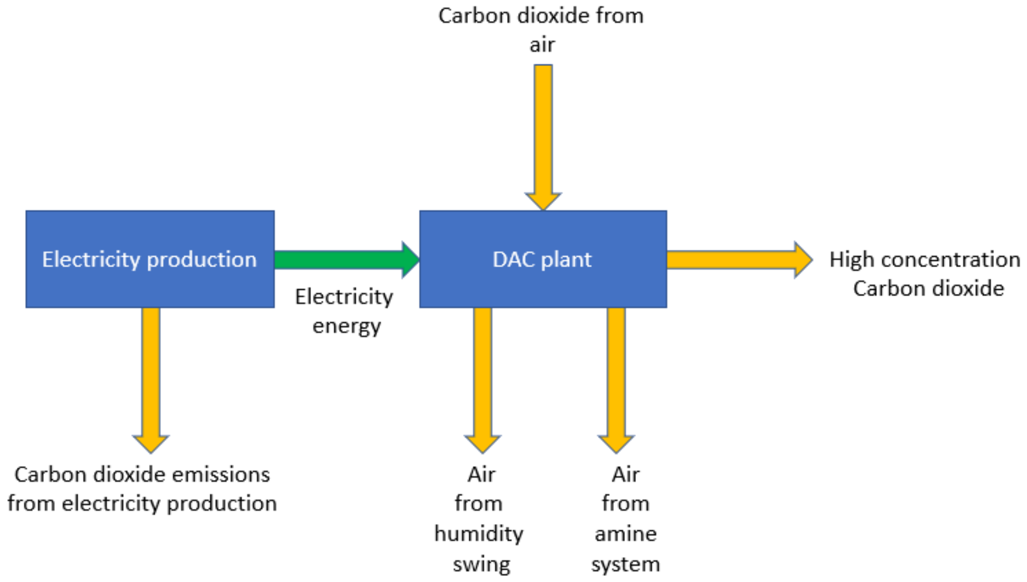


Figure 39: Flow rates of CO_2

Flow	**Flow rate [t-CO_2/hr]**
Carbon dioxide from electricity production	-1.17
Carbon dioxide from air	+222.54
Air from humidity swing	-0.00
Air from amine system	-54.02
High concentration carbon dioxide	-168.84
Net removal of CO_2 from atmosphere	**167.67**

Table 18: Inlet / outlet mass flow rates of CO_2.

6.6 High level of safety criteria

The DNI value used was the same as in (Zhai, et al. 2018) with a value 805 [W/m^2]. DNI in this range can be found in hot dry climates around the world. This resulted in a required area for the solar thermal system to be 12,421,372 [m^2]. Selection of the plant location should be optimised to minimise the area required and the affect the plant floor area will have on the local wildlife. Further to this the system requires the processing of 4450.8[ton/hr] of air which will affect local wildlife such as insects and birds. The effect this has on the local wildlife should be monitored and also any affect that this would have on the system.

The sorbent, Dowex Marathon A (Dow 2002), degrades over time and needs periodically replaced. It does not require any special disposal risks. Potential recycling opportunities of the sorbent has not been found. Table 19: Criteria risk / mitigation measures for humidity swing, outlines these risk and mitigation measures and connects these with the criteria for measurement.

Criteria	Low levels of pollution	Low use of dangerous processes or chemicals	Low levels of pollution
Risk	Total floor area of plant	Processing large volume of air and affect this will have on local wildlife	No recycling opportunities for derogated sorbent
Mitigation	Locate where DNI highest	Locate system where this risk is minimised and monitor effect on local wildlife and effect wildlife has on system	Search for recycling opportunities for derogated sorbent

Table 19: Criteria / risk / mitigation measures for humidity swing.

6.7 Energy use per ton of CO_2

Table 20: Energy use per ton of CO_2, shows the energy use per ton of CO_2. This shows the split thermal and non-thermal energy which uses grid electrical energy. The energy use per ton of CO_2 is based on the 166.08[t-CO_2/hr]. It can be seen that the thermal energy requirements are the most significant with the highest for the thermal energy of the humidity swing sub-system. A significant uncertainty for this was the ratio of mols of H_2O to CO_2. Therefore, further analysis should consider what the required ratio is as this heavily affects the energy requirements of the system.

Humidity sub-system	Non-thermal energy (Fan)	14[MW]
	Thermal energy (dehumidification)	6852[MW]
Amine sub-system	Non-thermal (Pumping)	4[MW]
	Thermal (heating requirements such as re-boiler)	151[MW]
Total		7021[MW]
Energy use per ton of CO_2 [MWHr/t-CO_2]		85

Table 20: Energy use per ton of CO_2

6.8 Using stripper re-boiler as heat source for de-humidification

Since the humidity swing system has low heat requirements, it is suitable to use to waste heat as a heat source (Fasihi, Efimova en Breyer 2019). In an amine carbon capture system, there is a large waste heat from the stripper re-boiler. The CO_2 captured from an amine system which captures CO_2 from flue gas could be toped up the CO_2 by a humidity swing system which captures CO_2 from the atmosphere and using the waste heat from amine system stripper re-boiler. This top-up would not require any extra thermal energy requirements. This would require the incorporation of a transport system to the storage location from the power station. This system setup is shown in figure 40: Humidity swing combined with amine carbon capture from flue gas.

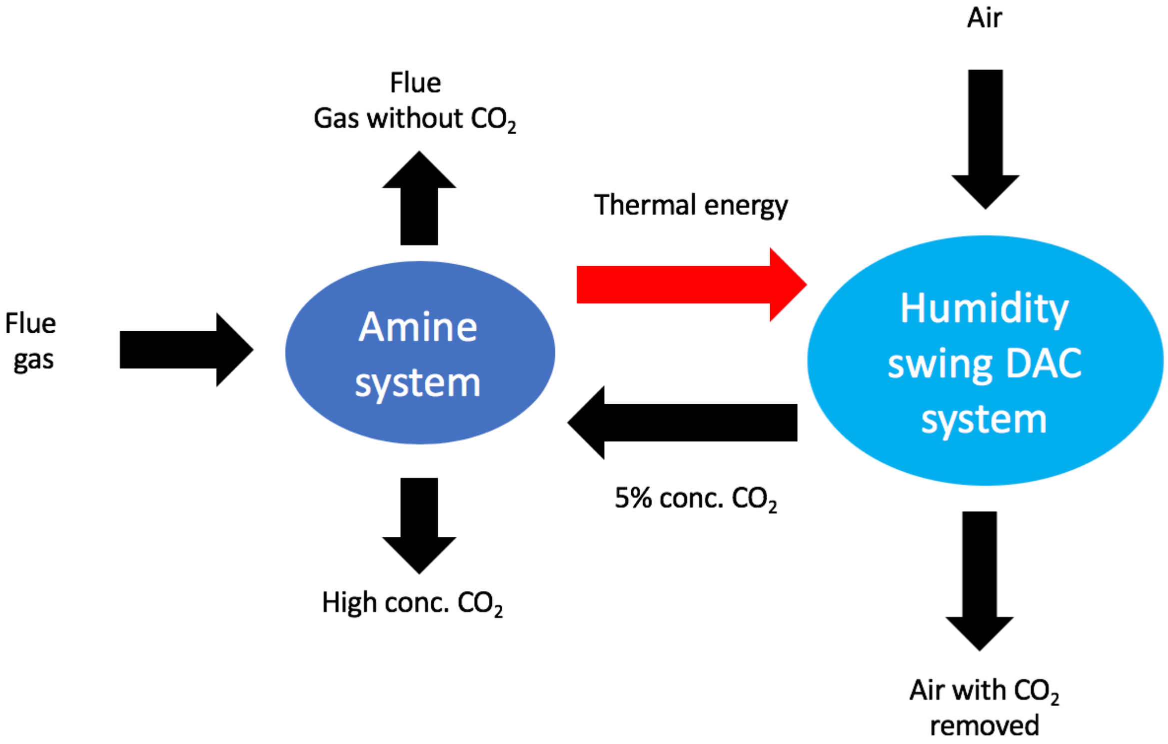


Figure 40: Humidity swing combined with amine carbon capture from flue gas.

In the amine base case there is a total cooling requirement of 770[kJ]. Using this to heat the air used for the dehumidification in the humidity swing system results in an increased capture rate of 8% with no extra-thermal energy requirements.

For the amine system base case without the cost for the humidity swing, the initial cost is 1,800,700[$]; while the cost for the addition of the humidity swing is 71,854,980[$]. The increased OPEX is dependent on how often the sorbent is changed. Since the degradation rate is not known this has not been calculated.

The base case is relatively small and captures 75% of all the CO_2 feed. Therefore, by using the system shown in figure 40: Humidity swing combined with amine carbon capture from flue gas, this could be increased to 83%. Most amine capture systems capture 90-95% of CO_2 fed to it. Therefore, further analysis should be carried to consider larger systems with a higher capture rate to see if such a system can capture the equivalent of the CO_2 released without the system shown figure 40: Humidity swing combined with amine carbon capture from flue gas.

Under a scenario where there is no other option available for the waste heat of the stripper re-boiler such a system would contribute to the Increased fairness of financial funding criteria as there would be no extra thermal energy requirement for the system and therefore a minimal increase in [MW/t-CO_2].

The calculation outlined here can be found in appendix-E.

6.9 Summary

Figure 41: Summary of findings of technical criteria for Humidity swing system, summarises the findings from this chapter which are analysed further in chapter 10. The emissions from

the plant result from the use of electrical energy for the non-thermal energy of the amine carbon capture sub-system. The OPEX almost solely comes from the operating of amine carbon capture sub-system which contributes 135[$Mill/year] and the humidity swing sub-system contributes 1[$Mill/year]. The solar thermal system contributes almost zero as there is no fuel requirement for this system. The CAPEX contribution is 1[$Mill], 483[$Mill] and 4310[$Mill] for the humidity swing, amine and solar thermal sub-system respectively. The solar-thermal sub-system CAPEX requirements is so high due to the high thermal energy requirements of the humidity swing sub-system. This calculation was based on the requirement for a 1:1 mol ratio of H_2O to CO_2 though experimentation of the sorbent will be needed the interrogate this assumption. For the levelised cost in terms of CO_2 the breakdown is as follows 0.6[$/t-$CO_2$], 92.5[$/t-CO_2] and 0[$/t-$CO_2$] for the humidity swing, amine and solar thermal sub-system respectively. The humidity swing sub-system levelised cost comes from the pumping of the air over the sorbent. The relationship between the exposed time to the sorbent and the removal rate of CO_2 from air is unknown. For this it was assumed that all the CO_2 is removed from the air. Further work should analyse what this relationship is. For the amine system when this price is compared to other systems this is relatively high. Therefore, work should consider optimising this sub-system further. Table 20: Energy use per ton of CO_2, breakdown the energy contribution per unit and if it is thermal or non-thermal energy used. Thermal energy use is the most heavily used with a total use of 7003[MW] from a total of 7021[MW]. This therefore increases the size and cost of the solar thermal sub-system. Since the non-thermal energy requirements comes from grid energy then minimising this reduces the emissions associated with grid electricity.

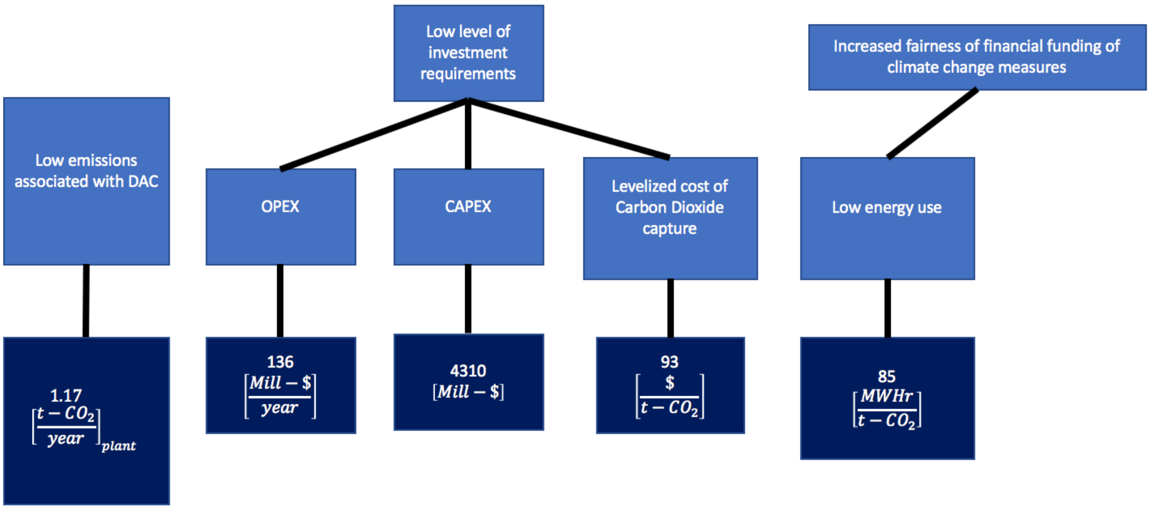


Figure 41: Summary of finding of technical criteria for humidity swing system

Chapter 7: Technical analysis of the high temperature solvent process

The objective of this chapter is to analyse the HT solvent process in terms of the criteria outlined in figure 42: Chapter 7 criteria. The results of this are the output of this chapter.

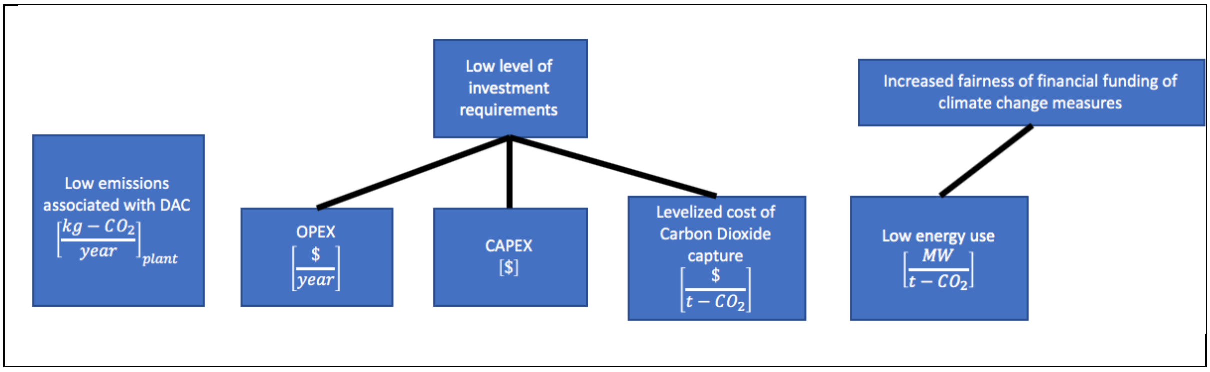


Figure 42: Chapter 7 criteria

As outlined in chapter 3: Review of literature on technical aspects of DAC, there are a number of units which are analysed further. These are outlined in table 21: HT solvent process units analysed, shown below. These will also be discussed in this chapter.

Unit	Air contactor	Pellet reactor	Calciner	Steam slacker

table 21: HT solvent process units analysed

7.1 Chemistry and mass balance

7.1.1 Chemistry

The HT solvent process reaction consists of an air contactor which contacts atmospheric air with the solvent Potassium Hydroxide. The air then leaves the air contactor CO_2 from the exhaust and the solvent which has reacted with the CO_2 to form Potassium Carbonate which is passed to the pellet reactor. The pellet reactor consists of pellets of Calcium Hydroxide which pass down the reactor while the Potassium Carbonate which a liquid flows up the pellet reactor. The calcium hydroxide pellets degrade over time and appropriate amount of make pellets are added as required. The solvent, potassium hydroxide, flows back to the air contactor to react with atmospheric CO_2. On the surface of the pellet reactor Calcium Carbonate is formed. This is washed off and travels to the calciner where it is heated at 900 degrees Celsius. Here, CO_2 is produced with a mass fraction of 0.97. In the slacker Calcium Oxide is also produced which is reacted with water in the slacker to produce Calcium Hydroxide. This is then used in the pellet reactor. This is depicted in figure 43: Reactions taking place in HT solvent DAC process (Keith, Holmes, et al. 2018), describes the reactions taking place in the process.

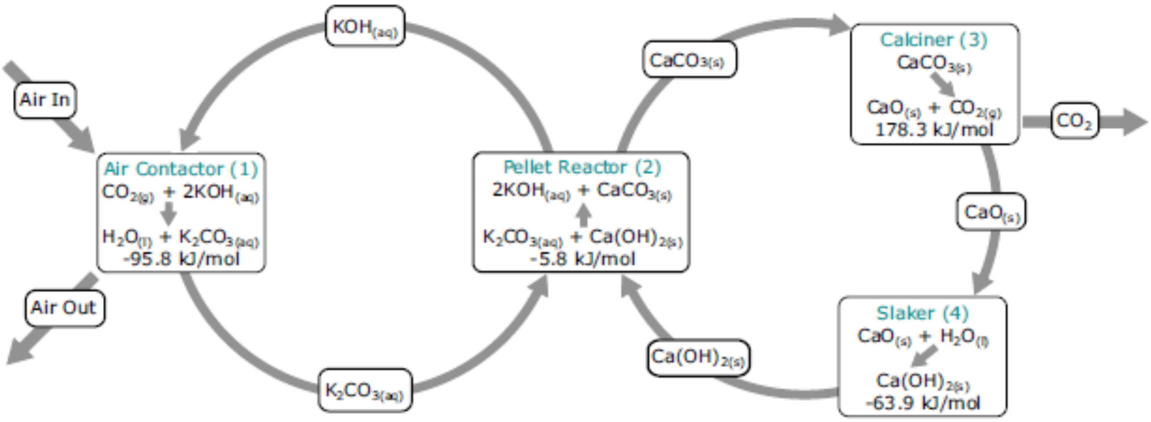


Figure 43: Reactions taking place in HT solvent DAC process (Keith, Holmes, et al. 2018).

7.1.2 Mass balance

Figure 44: PFD for HT solvent process, has been adapted by using colour circles to connect to the analysis in section 7.2 Unit analysis. In appendix-F a table of the full mass balance of the process can also be found.

The percentage error for the total mass balance of the compounds within the system can be found in in table 22: Mass balance error per component. It can be seen that the biggest error comes from compounds where atoms change to ionic state and back to the compound state. This results in extra calculations which make them more prone to error.

Carbon dioxide % error	Oxygen % error	Nitrogen % error	Water % error
9.4	0.2	0.1	14.0

Table 22: Mass balance error per component.

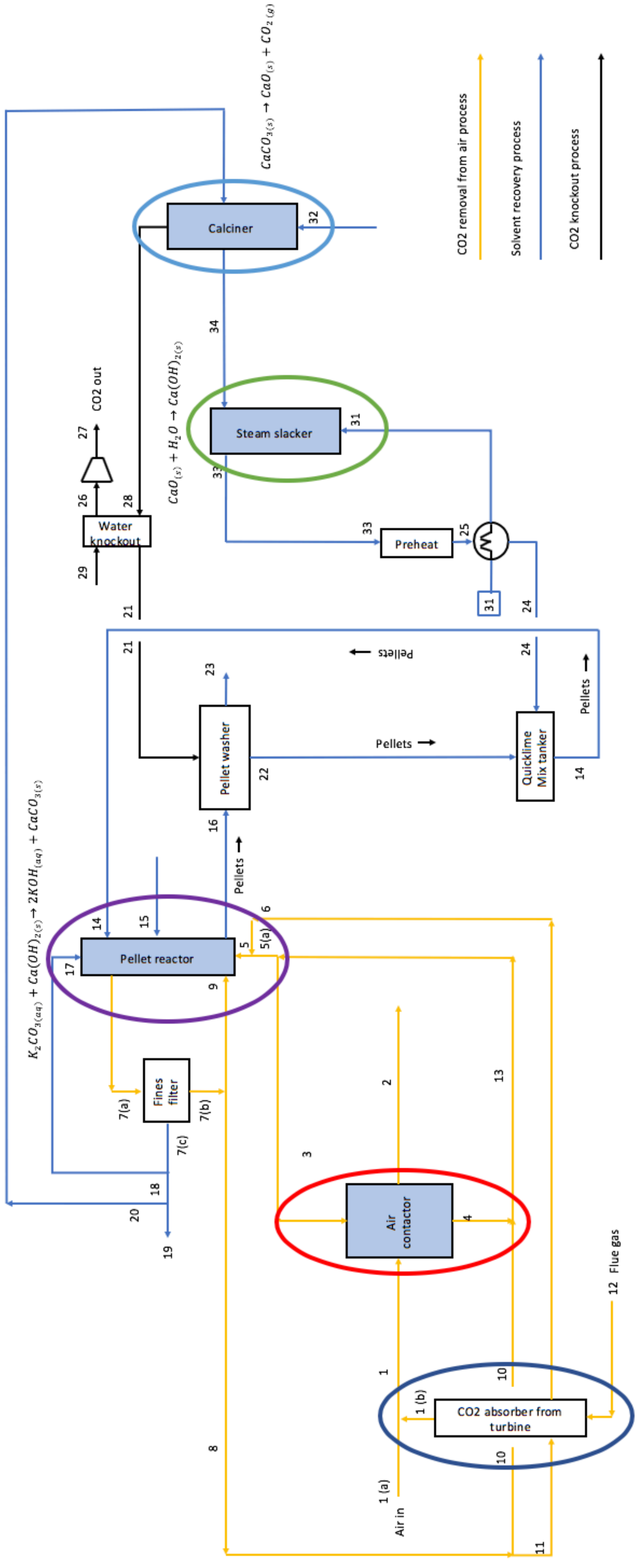


Figure 44: PFD for HT solvent process.

7.2 Unit analysis

CO_2 absorber

Note that stream 10 passes behind the absorber and does not interact with it.

The purpose of the CO_2 absorb from the flue gas is to absorb CO_2 from the burnt fuel used in the process. The turbine is gas turbine and if it is assumed that the gas used is methane at a rate of 6.3[t-methane/hr], when this is burnt in excess oxygen results in the same composition found in stream 12. This is the same as the value quoted in the paper. The calculation can be found in appendix-G.

The flow rate change across the CO_2 absorber was found not to be in line with what ion concertation change is reported as. The calculation for this can be found be found in appendix-H. This has a **gas/solvent ratio of 0.079**. This very low gas/solvent ratio can be explained by the high selectivity of potassium hydroxide has for CO_2. The OPEX costs of this piece of equipment relate to maintenance and pumping of the fluids. These are minimal when compared to the other OPEX costs and have not been included.

Air contactor

For the air contactor from the supplied data within (Keith, Holmes, et al. 2018) the solvent to gas ratio is not known. This is a key component for any solvent separation system as this determines the removal rate of CO_2 from the air. The air contactor, which is described in (Holmes & Keith 2012) and used in (Keith, Holmes, et al. 2018), is significantly larger than conventional absorbers. It is also of a different shape. It is not possible to input an absorber of the size and shape into conventionally used process modelling software.

To determine the solvent to gas ratio the software Aspen Plus was employed. The contactor described in (Holmes & Keith 2012) and used in (Keith, Holmes, et al. 2018) has a total cross sectional area 1700[m^2]. This was approximated by setting the dimensions of the absorber in Aspen Plus to the max possible. From this it was possible to determine the solvent to gas ratio required to capture the reported CO_2. This then made it possible to determine all the flow rates for the air contactor. Details of the process modelling can be found in appendix-I.

For the capture rate of CO_2 passive flow of air not sufficient. Therefore, a pump has to be used to pump the air through the contactor. The required energy was taken from (Keith, Holmes, et al. 2018) and the cost of electricity was taken as the wholesale electricity price for the USA (U.S. Energy Information Administration 2019).

Gas/ Solvent	**Pumping energy requirements**	**Air inlet flow rate**	**Energy requirements pumping**	**Electricity energy cost**	**Cost of pumping**	**Cost of pumping**	**Pressure**	**Temperature**
63.15	21 [kWHr/t-CO_2]	25110,5 [t-CO_2/hr]	3308.21 [kW]	0.03572 [\$/kWHr]	0.75 [\$/t-CO_2]	18833 [\$/Hr]	Atmospheric	20[^{O}C]

Table 23: Description of air contactor.

Pellet reactor

Within the pellet reactor the reaction taking place is potassium carbonate and calcium hydroxide to produce potassium hydroxide which returns to the air contactor. Calcium hydroxide is also produced, and this goes to the calciner to remove the CO_2 from the system. In Figure 14: PFD of the HT solvent process (Keith, Holmes, et al. 2018) the pellets are removed from the top when in reality the pellets fall down the reactor. This has been adjusted in figure 44: PFD for HT solvent process. For the pellet reactor there is no information regarding the pressure and temperature. A duty is supplied in Figure 14: PFD of the HT solvent process (Keith, Holmes, et al. 2018). A detailed description of modelling for modelling this reactor can be found in appendix-J. Table 24: Description of pellet reactor, outlines the characteristics for the pellet reactor

Pressure	Temperature	Volume	Duty
1[Bar]	20[^{O}C]	5[m^3]	26.1[MW]

Table 24: Description of pellet reactor.

Steam slacker

In Figure 43: Reactions taking place in HT solvent DAC process (Keith, Holmes, et al. 2018) the reaction in the steam slacker is indicated as a reaction between liquid water and Calcium hydroxide, though later it is described as gas due to giving better reaction kinetics. This is corroborated with other literature (Criado, Alonso en Abanades 2014). It is stated that the pressure of the steam slacker is 300 degrees Celsius and at 100[kPa]. From appendix-K, it is found that this results in a volume of 60.5[m^3]. The duty is calculated as the duty to heat the water up to 300 degrees Celsius as the Calcium Oxide is hot from the calciner. It is an exothermic reaction therefore there would likely be opportunity to recover some of this to heat up the water for this reaction.

Pressure	Temperature	Volume	Duty
1[Bar]	300[^{O}C]	60.5[m^3]	0.2[MW]

Table 25: Description of steam slacker.

Calciner

For the calcinatory from the supplied PFD it is not known what the recycle rate is, so this has been removed from the PFD created for analysis. Furthermore, the pressure and volume are not known though the temperature is quoted as 900 degrees Celsius. This is corroborated in literature which put this value at 900 degrees Celsius (Socolow, et al. 2011). In appendix-L the optimum pressure, temperature volume calculation is carried. Using the literature temperature of 900 degrees Celsius and at atmospheric pressure it is found that the volume of the calcinator is approximately 132[m^3]. The duty for the reactor was found to be 538.3[MW]. When this is compared to the energy available by the natural gas, which is 91[MW], it can be seen that there is an energy deficit. It should be noted the energy available from the natural gas was assumed to have no efficiency losses.

Pressure	Temperature	Volume	Duty
1[Bar]	900 [^{O}C]	132[m^3]	738.3[MW]

Table 26: Description of calciner.

7.3 Comparison of calculated values with reported values

For each unit the reported duty deviates from the calculated duty, these can be seen in table 27: Comparison of reported duty and calculated duty. For the reported energy requirements of the air contactor some of this can be attributed to pumping of the liquid which was not included in the calculation. The height of the air contactor is not known and therefore it is not possible to accurately calculate the energy requirements for this. For pellet reactor it was not possible to calculate the duty. The calculated energy requirements for the steam slacker were attributed to heating up water to 300 degrees Celsius. There may also be costs attributed to pumping of material which have not been included in the calculated number. For the Calciner the calculated value is significantly larger than the reported value. With such a significant deviation in these values there may likely be down to differing flow rates. The outlet flow rate calculated from the mass balance is 1096 [t/hr] while the reported value is 170 [t/hr]. The calculation was also carried out under isothermal conditions. In reality this is only true for the calciner. The other units require modelling under adiabatic conditions.

Unit	Reported Duty [MW]	Calculated duty [MW]
Air contactor	9.2	3.3
Pellet reactor	3.4	26.1
Steam slacker	3.6	0.2
Calciner	0.8	738.3

Table 27: Comparison of reported duty and calculated duty.

7.4 Low level of investment requirements criteria

The process used is complex and the air contactor is a novel unit. This makes it difficult to determine technical characteristics of the process and therefore cost them. Due to this, the values for these have been taken directly from the paper as this is the best estimate available. This is based on a gas fired energy source and a first-generation process. There is still a high level of uncertainty of the process as it was not possible to fully corroborate the findings outlined in their paper. Due to the large differences between the calculated values and reported values the uncertainty in the process description can be described as high. Considering the CAPEX for each unit it can be seen that the highest CAPEX comes from the air contactor. This is because it is an unusual and novel unit. The CAPEX only relates to the unit costing and not other CAPEX cost such as field costs and building costs. This has been done to remain consistent with the humidity swing system which did not consider these costs.

CAPEX	
Air contactor	212.2[$Mill]
Pellet reactor	130.7 [$Mill]
Calciner-slacker	77.7[$Mill]
Air separator	54.3[$Mill]
CO_2 compressor	19.9[$Mill]
Steam turbine	7.5[$Mill]
Power plant	35.0[$Mill]
Fines filter	30.9[$Mill]
Other units	102.9[$Mill]
CAPEX Total	**671.1[$Million]**
Levelized and OPEX cost	
Levelized cost in terms of CO_2	**232 [$/t-$CO_2$]**
OPEX	**34 [$Mill/yr]**

Table 28: Summary of HT solvent process.

7.5 Low emissions associated with DAC

A system boundary is set around the DAC plant and a CO_2 balance is done. The calciner supplies 13.4[t/hr] of CH_4 for combustion. This produces 36.86[t/hr] of CO_2. The balance of CO_2 around the system results in an error of -1.48[t/hr] of CO_2. The CO_2 produced via energy production by the combustion of CH_4 is captured and mixed with the CO_2 captured from air. This means that any downstream system that utilises the CO_2 can never achieve net zero emissions.

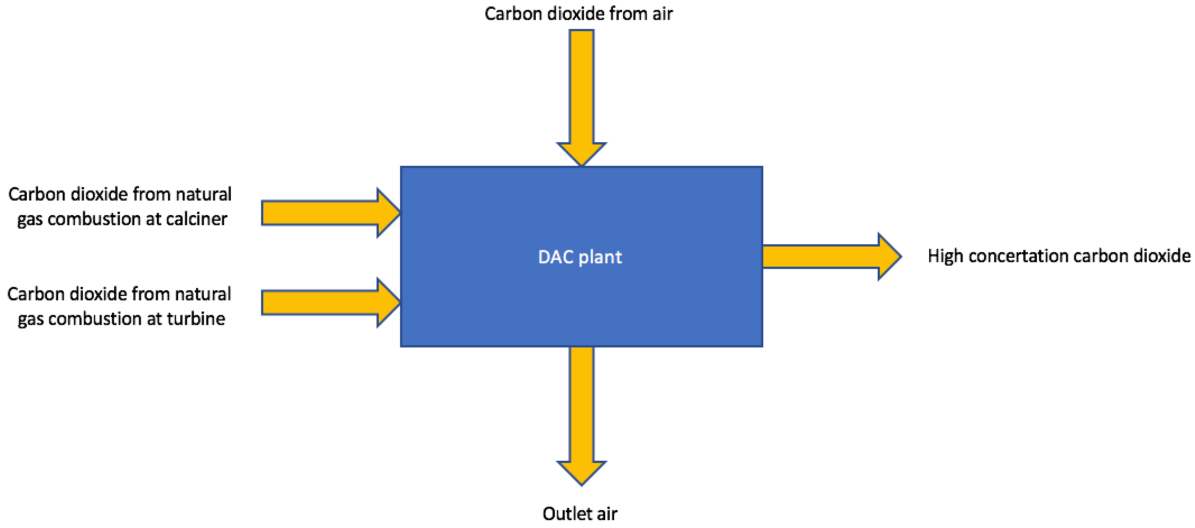


Figure 45: Boundary demarcation for CO_2 balance.

Flows	**Flow rates [t-CO_2/hr]**
Carbon dioxide from air	+150.6
Carbon dioxide from natural gas combustion at gas turbine	+17.46
Carbon dioxide from natural gas combustion at calciner	+36.86
High concertation carbon dioxide	-166.08
Outlet air	-40.32
Balance error	**-1.48**
Net removal of CO_2 from atmosphere	**166.08**

Table 29: Inlet / outlet mass flow rates of CO_2.

The upstream emissions are categorised as coming solely from the combustion of methane. The yearly emissions for this are shown in table 30: Upstream emissions from HT solvent process. These emissions are captured via CO_2 absorber and therefore are not emitted. The CO_2 captured from the air and the combustion of methane are mixed. Therefore, any utilisation method that has associated downstream emissions from the product that the CO_2 is converted to will result in a net increase in emissions and can never be assumed net-zero via this method. A full analysis of this is not carried out in this thesis.

Carbon dioxide from methane combustion [t-CO_2/hr]	475843
Upstream emissions [t-CO_2/yr]	0

Table 30: Upstream emissions from HT solvent process.

7.6 High level of safety criteria

The system uses the solvent potassium hydroxide. This poses as hazard to humans as it is acutely toxic if consumed orally, can cause sever irritation and corrosion to the skin and can cause serious eye damage (Caymen Chemicals 2018). To account for this in the APS system implemented a demister to remove any potassium hydroxide entrained in the exhaust of the air contactor. In the system designed in (Keith, Holmes, et al. 2018) paper they found that the exhaust met concertation limits for potassium hydroxide in air and therefore did not include any mitigation measure for this risk. Despite this due to these conflicting reports this risk should be monitored and the process should passively stop exhausting to the atmosphere if limits of potassium hydroxide are found to exceed air quality limits.

Potassium hydroxide also poses a risk to equipment as it can be corrosive to metals (Caymen Chemicals 2018). Therefore, material selection should account for this.

The system uses high temperatures which can pose a risk to humans if exposed to. Therefore, when implementing the inherently safe design philosophy during the design of the system this should be accounted for.

This system also poses a risk to local wildlife as it has to process a large volume of air. This can affect local bird and insect population. Therefore, the location of the system should be where this risk is minimised. Furthermore, if wildlife is drawn into the intake this may negatively affect the system itself and this risk should be monitored.

Table 31: Risk and mitigation measures for HT solvent process, outlines these risks a mitigation measures that should be taken.

Criteria	Low levels of pollution	Low use of dangerous processes or chemicals	Low use of dangerous processes or chemicals	Low use of dangerous processes or chemicals
Risk	Solvent entrained in exhaust	Solvent risk to equipment	High temperatures used within system	Processing large volume of air and affect this will have on local wildlife
Mitigation	Monitor solvent in exhaust and do not exhaust to atmosphere if concentrations exceed air quality limits	Select materials which will not be degraded by solvent	Account for this during implementation of inherently safe design philosophy during design of system	Locate system where this risk is minimised and monitor effect on local wildlife and effect wildlife has on system

Table 31: Risk and mitigation measures for HT solvent process.

7.7 Low energy use criteria

Table 32: Energy use per ton of CO_2, shows the energy use per ton of CO_2. This system used natural gas as the sole energy source with an outlet pressure for CO_2 at 15[MPa].

Process	HT solvent process
Duty [MW]	406
CO_2 production rate [t-CO_2/Hr]	166.08
Energy use [MWHr/t-CO_2]	2.4

Table 32: Energy use per ton of CO_2.

7.8 Summary

Figure 46: Summary of findings of technical criteria for HT solvent system, summarises the findings from this chapter which are analysed further in chapter 10. Since the HT solvent system generates all its own energy requirements and captures the associated CO_2 from this at has no emissions. This differs from the humidity swing sub-system which uses grid energy for non-thermal energy and therefore has an associated emission if 1.17[kg-CO_2/year]$_{plant}$. The HT solvent system also has a lower OPEX with the humidly swing system having an OPEX of 136[Mill&/year]. This is also true for the CAPEX with the CAPEX for the humidity swing system been 4310[$Mill]. The largest contributor to this is the air contactor at as this is a unique unit. For the levelised cost the humidly swing is cheaper at 93[$/t-$CO_2$]. This is because the thermal energy is the biggest contributor to the humidity swing energy requirements has almost no operating cost. While the HT solvent system uses NG, which has a cost of 3.5 [$/GJ]. The energy use is also significantly lower than the humidity swing system and is based on the use NG to power the system which supplies 8.81[GJ/t-CO_2].

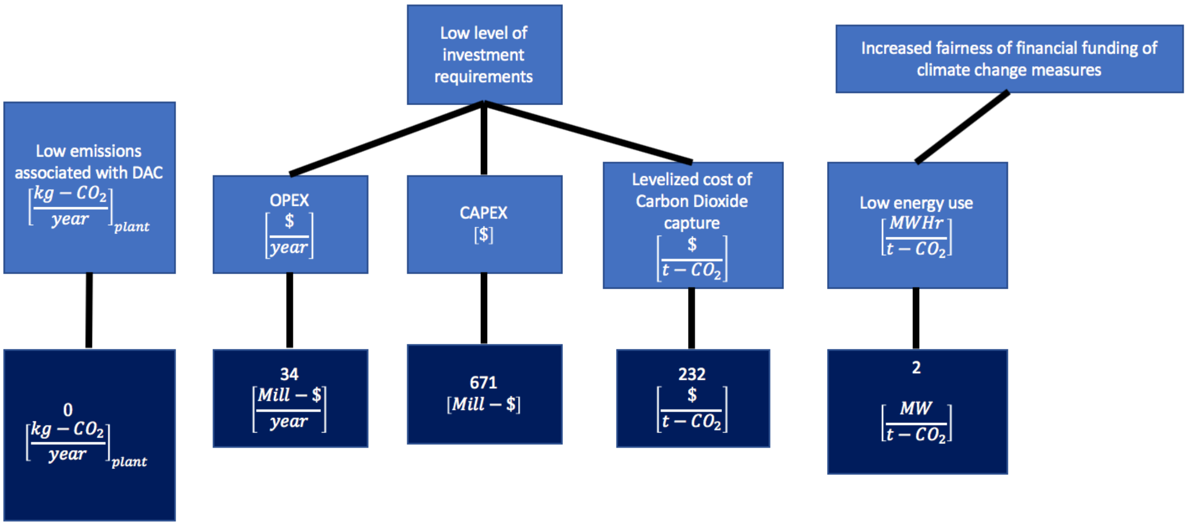


Figure 46: Summary of findings of technical criteria for HT solvent system

Chapter 8: Analysis of low public finance criteria

This chapter analyses the effect that policies regarding climate change measures will have on public finance requirements, and their contribution to the fairness of financial funding criterion. This is outlined in Figure 47: Fairness criteria. The output of this chapter is an analysis of the effect the policies outlined in chapter 4 will have on this criterion and the affect they can be expected to have on use of DACS.

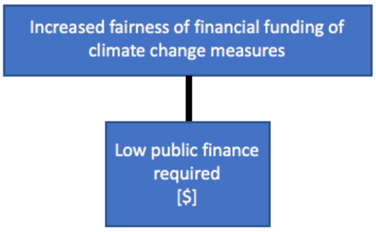


Figure 47: Fairness criteria

8.1 Low public finance required

The low public finance criteria are important to DAC companies as they need to demonstrate to the public authority that their system will not burden public finances. This, will therefore, mean that the public authority will see DACS as an attractive option to achieve their climate change targets while freeing up finances for other projects.

In the previous section, a means-end diagram was used. The purpose of this was to identify what to focus on and an objective tree was used to define the criteria which are used to evaluate solutions (Enserink, et al. 2010). The system diagram used in this section is used to identifies which policies result in a low public finance requirement and the casual relationship between the factors. There are a range of policies available which may affect DACS development. This section will consider the effect these policies will have on the level of public finance required for the technology, namely the ETS, CfD ,45Q legislation, CDM. This is done using a system diagram seen in Figure 48: System diagram, to evaluate the criterion low public finance. Further to this the relevant co-effects CO_2 emissions and use of DAC are analysed.

Box 2: How policies change

When selecting policies funding and research is required to determine the effects these policies will have. When the option taken is to maintain the status quo, uncertainty is reduced as predicting future outcomes is be based on past experience. This means that implementing any new policy increases uncertainty about the future. In minimising uncertainty by maintaining the status quo, a path dependency is created on policy decisions. At some point, the uncertainty about the effect of implementing new policy outweighs the predicted future with the current policies and the new policy is implemented.

Internal system description

- Increased use in DACS technologies can reduce atmospheric CO_2 and therefore reduce global emissions.
- Energy efficiencies can reduce energy demand for the transport sector and energy use in energy production.
- An increase in energy use in energy production and energy use in transport will increase the requirement for CO_2 offsetting.
- Efficiencies in the energy production sector can reduce CO_2 emissions for this sector.
- Develop storage location can encourage DAC.

Means description

- A CfD is the opposite of a penalty as it promises a minimum price and can be used for CO_2 offsetting.
- The ETS is a market-based penalty which is applied to large emissions sources such as power stations for the emissions they cause. It is designed to reduce emissions from large point sources.
- The CDM is a mechanism that allows for emissions to be offset in one location which is not the same the as the place where the emission took place. It, therefore, increases the offsetting of emissions.
- The CDM will require funding of emission offsetting projects. In this example, it is financed through tax rebates for such schemes.
- A tax rebate can be used to encourage CO_2 utilisation and storage. As this is a tax rebate there will be a cost to the taxpayer. This is how the 45Q legislation is designed to encourage DACS use.

External factor description

- Economic downturn will reduce energy use across the economy.
- A negative public perception of carbon dioxide emissions can force an increase in energy efficiency, resulting in consumers informing themselves of product and service emissions.
- As the population size increases, so will increase energy use across all markets.
- An unexpected increase in climate change could result in faster uptake in more efficient practices and CO_2 offsetting.

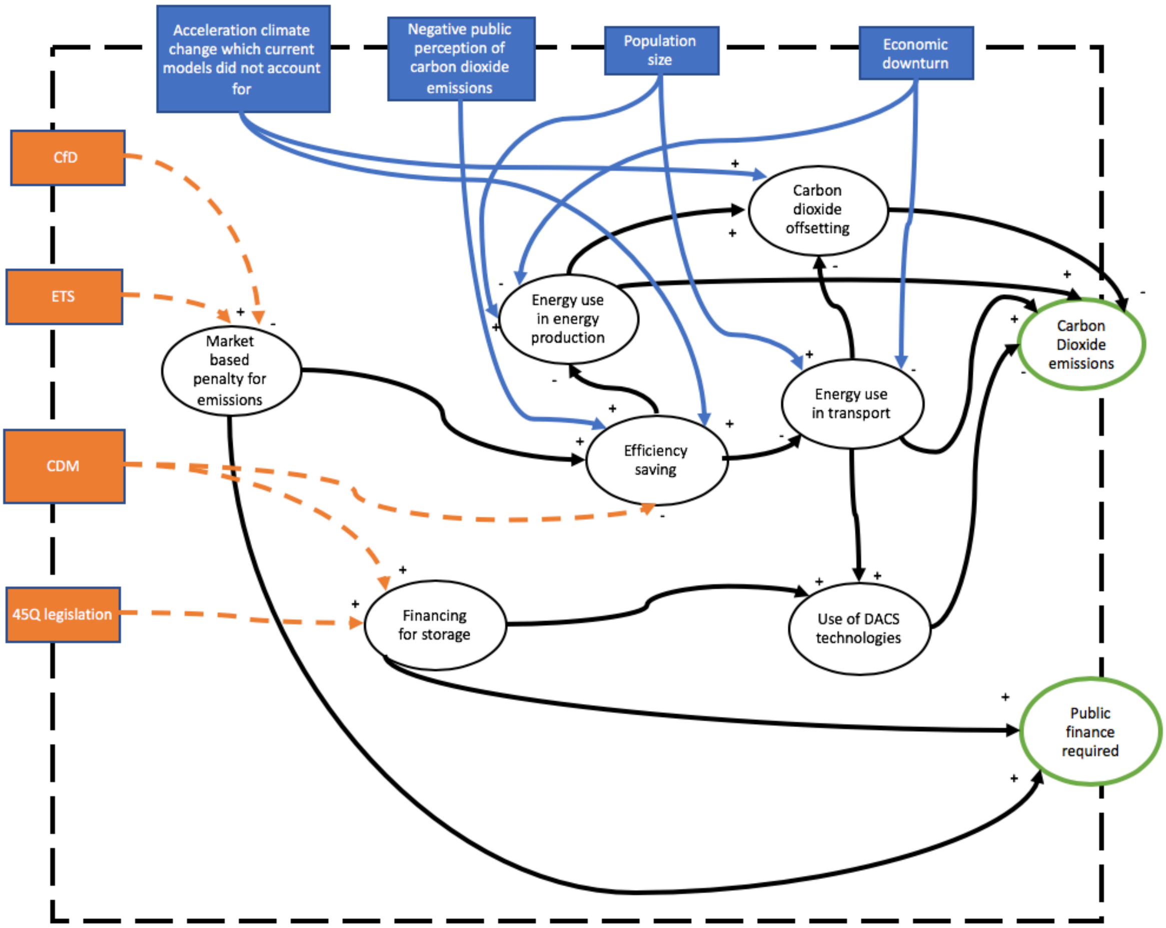


Figure 48: System diagram.

8.2 Consequence of system diagram

Table 33: System diagram consequence table, outlines the expected consequences of different policies.

Criteria → ↓Means	**Carbon dioxide emissions**	**Public finance required**	**Use of DACS technology**
ETS	-/-/-/+/+	+	-
CfD	-/+/+/-	-	+
CDM	-/+/+/-	+	+
45Q legislation	-	+	+

Table 33: System diagram consequence table.

The system diagram shows that the:

- ETS policy results in a lower likelihood of DAC technology being implemented, this is because this policy does not allow for emissions to be offset.
- CfD policy results in a higher likelihood of DACS implementation. This is because it can be used to directly fund green technologies such as DACS.
- CDM allows for the offsetting of emissions through the funding of projects in one location and accounting for the removal of emissions in another location. DACS has

been identified as being able to offset emissions (Fasihi, Efimova and Breyer 2019). Therefore, CDM policy can be used to fund DACS projects.
- The 45Q legislation encourages DACS through tax incentive and therefore attributing financial value to CO_2 captured using DACS.

DACS has been identified as a CDR technology (IPCC 2018). Furthermore, the IPCC has stated that CDR technologies will be required to meet climate change targets (IPCC 2018). Therefore, policies that include CDR technologies should be included in future policies. In the example given above the CDM allows offsetting through CDR.

The IPCC have also stated that CDR technologies are required to produce negative emissions (IPCC 2018). Negative emissions are those that remove greenhouse gases from the atmosphere, which are stored outside the atmosphere, that the upstream and downstream emissions are comprehensively estimated and included in the emission balance, and that the total quantity of atmospheric greenhouse gases removed and permanently stored is greater than the total quantity of greenhouse gases emitted to the atmosphere (Ramirez and Tanzer 2019). Therefore, the targets need to be adjusted to meet these requirements

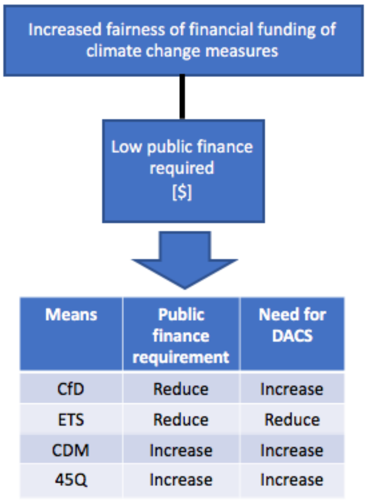
As the ETS does not allow for emissions to be offset, which DACS has been shown to be capable of (Fasihi, Efimova and Breyer 2019), then the ETS and DACS are not compatible in the ETS current form. DACS could be incorporated into the ETS, if it is adapted to allow for emissions to be offset by removal of CO_2 from the atmosphere. Despite this, it would not be able to account for negative emissions as the ETS applies a penalty for emissions. However, since the systems relies on a penalty system it does not require public funding.

The CfD policy can be used to fund green technology in the energy production sector. The technical analysis showed that the humidity swing system can be combined with a flue gas amine capture system for NG or coal energy production systems. Therefore, a CfD could be used to fund such a system. The cost is passed to the consumer and therefore does not require public investment. This can be deemed to be fair as the energy and heat sector is the largest emitter per sector at 49.04% (WorldBank 2019). It is assumed that those that use this sector the most emit the most and therefore pay the most. The other leading sectors in terms of emissions are transport (20.45%), industry (19.96%), buildings (8.60%) and others (1.95%) (WorldBank 2019).

The 45Q legislation can directly encourage DACS through the tax rebate it applies. Despite this there is a mismatch between the cost of capture and value the of the tax rebate. For the two systems analysed here the cost of capture was 94[\$/t-CO_2] for the humidity swing and 232 [\$/t-CO_2] while the tax rebate value is 34[\$/t-CO_2] (Bennet 2018). For this mechanism to be successful then the tax rebate should be higher than the cost of capture and storage. Furthermore, the fact that this cost is met solely by public finances it does not make this an attractive option for governments.

8.3 Summary

Figure 49: Summary of system diagram, which summarises the findings from this chapter. The Need for DACS column indicates if a policy would be supportive of DACS development. Those that increase the need for DACS can be deemed to be supportive of it while those reduce the Need for DACS are not supportive. The public finance requirement column indicates whether a policy is deemed to be fair. If the policy reduces the need for public finance, then it can be deemed fair and vice versa. From this it can be seen that the CfD increases the need for DACS and reduces the need for public finance. For the CfD the system would need to be the humidity swing incorporated into an amine system capturing CO_2 from the flue gas for energy production from NG or coal.



Means	Public finance requirement	Need for DACS
CfD	Reduce	Increase
ETS	Reduce	Reduce
CDM	Increase	Increase
45Q	Increase	Increase

Figure 49: Summary of system diagram.

The use of CDM to achieve negative emissions bodes relevance to this research as for the humidity swing system it is not appropriate to locate it in locations that are exposed to low solar thermal energy. The design for this system given in chapter 6 assumed a location exposed to high solar thermal energy. With the CDM it allows for targets to be met in one location while CO_2 is removed in another. This means that the humidity swing system could be located somewhere exposed to high solar thermal energy but account for targets of a country with low solar thermal energy.

As mentioned in chapter 1, J. Wurzbacher from the company Climeworks identified that carbon pricing can be used to encourage DACS development (The National Academies of Science, Engineering and Medicine 2018). However as outlined chapter 5, the ETS in its current form is not capable of encouraging DACS as the ETS does account for emission offsetting which DACS is capable of doing (Fasihi, Efimova and Breyer 2019). Therefore, if the ETS is to be adapted to encourage DACS it should account for emission offsetting.

The CfD can be used to fund a humidity swing system combined with an amine capture system capturing CO_2 from coal or NG combustion for energy production. Further by passing this cost on to the consumer of electricity it is deemed fair as the electricity and heating sector is the largest emission sector. This assumes that those who consume the most electricity emit the most and therefore pay the most for green technology. Furthermore, for the 45Q legislation, the tax rebate should be higher than the cost of capture and storage to make it an attractive option which is currently not the case.

Chapter 9: Stakeholder analysis

The purpose of this chapter is to analyse where coalition between DAC companies and other actors is possible and those actors that would be opposed to DAC companies been successful in developing DAC technology. The output of this chapter is a list of stakeholders categorised as either potential threats or allies.

The analysis carried out relates solely with regards to the EU. For other jurisdictions such as the USA, the relationship and resources of actors may be different. Therefore, for other jurisdictions, another stakeholder analysis should be completed. The actor analysis is carried out under the scenario where the EU is implementing the energy transition towards net-zero emissions and ultimately negative emissions.

9.1 Mapping relationships between stakeholders

The current formal relationships between actors within Europe related to large emission sources is shown in figure 50: Relationship between different actors related to DAC. This should not be viewed in a hierarchal manner, but rather in terms of formal relationships between actors. This is followed by a set of text boxes which describe this figure.

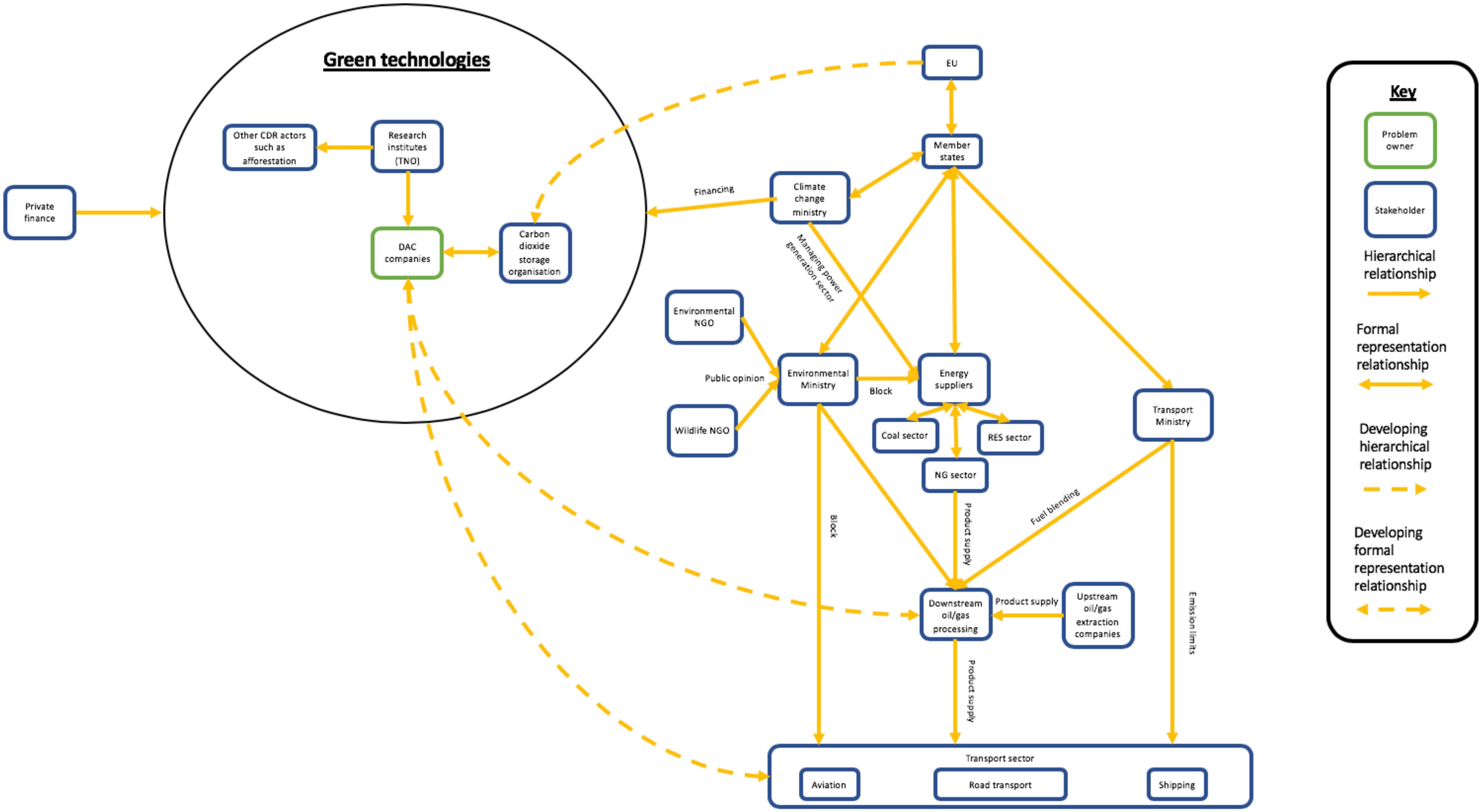


Figure 50: Relationship between different actors related to DAC.

Box 3: EU Actor

The EU is the umbrella organisation for a range of actors such as the European Commission and European Parliament. The main actor related to climate change is the Director General (DG) for Climate Action. This DG sets targets, examples of this include the Renewable Energy directive which sets targets for renewable energy use within the EU (European Commission 2019). They are also responsible for managing programmes such as the ETS. These rules and regulations are passed by the European Parliament where member states can vote on them. Once they are passed these states have to adhere to these regulations. Another relevant DG is for the Environment. This DG is relevant as they are responsible for risk assessing legislation related to environmental matters.

Box 4: National government actor

At a government level, there are also a range of relevant actors. These include the ministry for the environment which is responsible for environmental protection and standards. Most countries also have a separate ministry for climate change. These actors are used to develop and apply regulations. The transport ministry is also relevant as they set regulations regarding fuel blending. An example of this in the UK is that fuel will require a minimum 12.4% biofuel (Department for Transport 2018). This can be used to reduce emissions and will affect downstream oil/gas processors.

In the UK a relevant actor is the Department for Business, Energy & Industrial Strategy (UK Government 2020). In relation to DAC and climate change, this department is responsible for financing new green technology, managing emission targets and energy suppliers.

The environment ministry can block infrastructure development when it is deemed not to be in line with legislation or if it excessive causes environmental damage. In the UK this ministry is the Department for Environment, Food and Rural Affairs (DEFRA) (UK government 2019). DEFRA is subject to pressure from organisations such as environmental and wildlife NGOs. One example of this is WWF working via the Scottish environment LINK, which brings together volunteer organisations related to environmental protection to represent the environment and influence governmental organisations (Scottish Enviroment Link 2020).

Box 5: Energy market

The downstream oil/gas sector has to abide by legislation imposed by the Environment Ministry. This could, for instance, concern the expansion of plants to include new processes which DEFRA could block on environmental grounds.

Downstream oil/gas suppliers require product from upstream oil/gas companies. This oil/gas is then sold the transport sector or used for electricity production.

Box 6: Green technologies

DAC companies sit within the green technology sector. Due to the high level of uncertainty regarding this technology, no significant formal relationship or dependency has developed with the actors mentioned above. Due to the potential of DAC actors have started to research how these relationships can be developed.

Currently there is a relationship developing between DAC companies and downstream oil/gas sector. An example of this is Exxon Mobil, which has started joint venture with the DAC company Global Thermostat (Exxon Mobil 2019).

Other developing relationships exist between the aviation sector and DAC companies to develop non-biological synthetic fuel. Example of this is the company Climework collaborating with Transavia Airlines at Rotterdam the Hague airport (Virardi 2019) to develop this process.

Development of CO_2 storage has also been supported by climate change ministries. Examples of this include in Iceland where the EU has supported underground storage of CO_2 through projects captured from DAC through the Horizon 2020 program (Climeworks 2017).

Development in this sector can be assisted through research and collaboration with research institutes, such as TNO.

There are other CDR options available in the green technology sector such as afforestation, which DAC is in competition with to attract funding from. Such options include afforestation, land management, BECCS, enhanced weathering and ocean fertilisation. The formal relationship of these have not been included in figure 50: Relationship between different actors related to DAC.

The relationship between DAC companies and energy suppliers shows technical potential but has not been formally developed. The technical relationship comes from the demonstration in section 6.5: Using stripper re-boiler as heat source for de-humidification, that an amine carbon capture system used to capture CO_2 from flue gas with associated DAC system can help energy suppliers achieve net-zero or negative emissions with no extra heating requirements. This relationship could never have been developed till technical analysis of such a process has been carried out. After reviewing literature this is the first time such a system has been analysed.

9.2 Actors problem formulation

Table 35: Stakeholder resource and objectives table, outlines the objective and resource to achieve this for each actor. This can then be used to categorise actors into allies or threats as this is based on if their objective is conflicting or similar that of DAC companies. Furthermore, their ability to block or not DAC technology determines if they are critical or not to DAC companies. Potential allies are marked in green while threats are marked in red.

Looking at table 35: Stakeholder resource and objectives table, it can be seen that all governmental organisations are potential allies. Examples of interaction between Government and DAC companies can be seen from the company Climeworks been a member of the group “Alliance for Efficient Solutions to Move the Carbon Removal Agenda” which took place at during the 2018 United Nations Climate Change Conference (Climeworks 2017). Governments are also extremely relevant to DAC as they can propose/implement legislation such as fuel blending standards which could directly affect DAC companies.

Within the energy sector all those benefiting from burning of fossil fuels are potential allies. This is because the humidity swing DAC system can be combined with an amine carbon capture system. This would mean that there are no additional thermal energy requirements for the humidity swing. Furthermore, the system described here would legitimise the continued use of fossil fuels and could damage the public perception of DAC companies. The only actors which are potential threats for this sector is the renewable energy sector used for electricity generation as this removes emissions from the system and therefore reduces the need for DAC.

Other CDR options used by other actors pose a threat to DAC companies. This is because they compete with DAC companies to achieve the same objective, which is to remove CO_2 emissions from the atmosphere. Table 34: CDR processes comparison (European Academies Science Advisory Council 2018), outlines the capacity of these systems and cost of them. These other CDR options do not pose the same potential in terms of CO_2 utilisation as DAC and in this respect, DAC is a more attractive option.

Technology	Afforestation	Land management	BECCS	Enhanced weathering	DACS	Ocean fertilisation
$\Delta CO_{2,process}$ $[Gt - CO2/year]$	1.1-3.3	2-3	3.3	1	3.3+	<1
Cost ***[\$/t-CO₂]***	<100	<100	100-400	100-400	100-400+	0-400

Table 34: CDR processes comparison (European Academies Science Advisory Council 2018)

Within the oil/gas supply chain, downstream oil/gas processing companies represent a potential ally. This is because the CO_2 from DAC can be used as a sustainable feedstock for processes taking place in this sector. While up-stream oil/gas suppliers are a potential threat. This is because DAC companies can be part of the consortium which replaces upstream oil/gas supplier’s product. Example of collaboration between these actors is with ExxonMobil and the DAC company Global Thermostat (Exxon Mobil 2019). Despite showing potential to replace oil/gas products for sectors such as transport (Royal Society 2019), this could be seen by the public as an attempt by oil/gas companies to greenwash their product. Greenwashing is a process where companies mislead consumers about the environmental performance of their product (Delmas and Burbano 2011).

Within the transport sector there is the Aviation, Road transport and shipping sub-sectors. All these sub-sectors are potential allies this is because CO_2 from DAC can be used to in production of sustainable synthetic fuel production. This fuel can be used in all transport types without adaption to current motors (Royal Society 2019). This means that DAC companies can help the transport sector reduce its CO_2 emissions.

Wildlife and environmental NGOs represent a threat to DAC companies. This is because both systems require large processing large volumes of air which could adversely affect the natural environment and wildlife. Examples of this is the Royal Society for the Protection of Birds (RSPB) blocking windfarms in areas of natural importance (RSPB 2020) due to concerns regarding the adverse effect on bird populations. Despite this issue, which can be regarded as a local issue, on macro-scale DAC can be regarded as a benefit to the environment and wildlife due to the positive affect it can have on global warming. DAC companies can therefore nullify the threat posed by their system on local wildlife and environment by locating it where the negative affect is minimised.

CO_2 storage organisations are required for DACS. Examples of CO_2 organisations are companies such as Drax (European Commision 2016) or SaskPower (SaskPower 2020) who are electricity producers who capture the CO_2 from their electricity production and permanently store it underground. Other CO_2 storage organisations include those that use CO_2 for EOR, an example of this is Ocidential Petroleum gas processing facility (MIT 2016). The CO_2 storage is always done in collaboration with another system where there is a CO_2 source (MIT 2016). Therefore, unless DAC companies wish to diversify their specialisation to include CO_2 storage, they will need to collaborate with one of these CO_2 storage organisations. Example of this type of collaboration can be seen in Iceland where DAC is combined with a storage system at a power plant (Climeworks 2017).

Research institutes, such as TNO, are also a potential ally. This is because TNO purpose is to advance technology which enhances society. DAC is a developing technology which has potential to reduce CO_2 emissions and therefore can support society. This means that DAC is a potential ally for DAC companies.

Actor		**Important resource**	**Desired situation/Objective**
EU	**DG for climate action**	Development and implementation of ETS, set emission targets per member state and fund low carbon technologies (European Commision 2020)	Lower greenhouse gas emissions and be carbon neutral by 2050 (European Commision 2020)
	DG for environment	Can take member state to European court of justice if in breach of legislation and member state does not rectify this (European Union 2008)	Increase protection of biodiversity and wildlife (European Commission 2019)
Member states	**Climate change ministry**	Choose where to distribute funding for green technology/impose emission limits on energy generators (UK Government 2019)	Ensure a reliable, low cost and clean energy system (UK Government 2020)

	Transport ministry	Select emission limits & control fuel blending (Department for Transport 2018)	Increase development of a clean and sustainable transport sector (UK Government 2020)
	Environment ministry	Block development of projects based on affect it has on environment (UK government 2019)	Increase protection and harnessing value from the natural environment (UK government 2019)
Energy sector	**RES Sector**	Non-finite resource used and falling price of electricity production from RES. Also has a high level of social acceptance (Stigka, Paravantis and Mikalakakou 2014)	Reduction in CO_2 emission limits and reduced fossil fuel used as energy source
	NG Sector	Can respond to electricity demand quickly (Marini 2017)	Increase market share in energy production while supplying clean energy
	Coal Sector	Block new players in energy production due to large market share (Offgem 2019). Also, a known technology which is reliable	Increase market share in energy production while supplying clean energy
	Energy suppliers	Control where energy source comes from	Increase affordable energy from a clean source.
Transport sector	**Aviation**	Demand for service (World Bank 2018) and only sustainable non-biological synthetic fuel suitable alternative which reduces emissions while maintaining demand for service	A reduction in CO_2 emissions while not affecting growth in sector (IATA 2016)
	Road transport	Current infrastructure heavily based on road transport and suitable alternatives such as electric vehicles can reduce CO_2 emissions (Department for Transport 2010)	Maintain reliance on road transport while reducing CO_2 emissions
	Shipping	Demand for service (World Bank 2018) and suitable alternatives such as ammonia based shipping can reduce CO_2 emissions (Ash and Scarbrough 2019).	Introduction of zero-emission ships by 2030 (Saul 2019)
NGO	**Wildlife NGO**	Pressure decision makers by influencing public opinion.	Increase protection of wildlife (UNESCO 2019)
	Environmental NGO	Pressure decision makers by influencing public opinion	Increase protection for the environment (Enviromental protection society 2013)
Oil companies	**Downstream**	World has high dependency on processed oil/gas products	Venturing into multiple low carbon energy sources (BP 2020)
	Upstream	No suitable alternative product for downstream companies	Protect market share for crude oil/gas (BP 2020)
Other CDR actors		Cheap alternative for CO_2 removal (Gale, et al. 2013)	In the case of company Drax it is to reduce emissions via a BECCS power plant which can achieve negative emissions (Drax 2020)

DAC companies	Sustainable feedstock for processes requiring CO_2 and can remove CO_2 directly from atmosphere and stored therefore contributing to negative emissions	Increase implementation of technology and reduction in CO_2 emissions (Climeworks 2019)
CO_2 Storage organisations	When combined with DAC it can be used for offsetting of emissions (Climeworks 2020)	Reduction of atmospheric CO_2 by storing CO_2 underground
Research institutes (TNO)	Expertise on range of different processes	“To create innovations that boost the competitive strength of industry and the well-being of society in a sustainable way” (TNO-Mission statement 2020)

Table 35: Stakeholder resources and objectives table.

Table 35: Stakeholder resource and objectives table, does not analyse if actors dedicated to the problem. A dedicated actor is one who is affected by the solution to the problem that DAC companies would like to solve (Enserink, et al. 2010). Here, DAC companies would like to solve the problem of CO_2 emissions by widespread implementation of their product. This can be either to remove CO_2 from the atmosphere for long term storage or as a feedstock for other processes such as the sustainable production of non-biological synthetic fuel. Therefore, those who directly affected by climate change are dedicated and those not non-dedicated. This is shown in table 36: Actor dedication to problem.

Actor	Dedicated/non-dedicated	Reason
EU	Dedicated	Citizens are directly affected by climate change (IPCC 2018).
Member states		
Climate change ministry	Dedicated	As climate change is core part of the objective of this ministry (UK Government 2019) then they will be directly affected by actions taken to tackle climate change.
Transport ministry	Dedicated	As climate change is core part of the objective of this ministry (UK Government 2020) then they will be directly affected by actions taken to tackle climate change.
Environment ministry	Dedicated	As responsible for the local environment (UK government 2019), they are affected by climate change or the affect that a DAC plant would have on the local environment.
RES Sector	Non-dedicated	RES will likely still be required to meet CO_2 emission targets even with DAC with CO_2 utilisation or DACS.
NG Sector	Non-dedicated	Almost all CO_2 can be captured with conventional amine carbon capture system and when this is combined with a DAC system the DAC system tops up the CO_2 captured.
Coal Sector		
Energy suppliers		
Other CDR actors	Dedicated	They are in competition with DAC using alternative method to achieve the same goal as DAC companies
Downstream oil processing	Dedicated	DAC use for production of non-biological synthetic fuel can be in fuel blending for fuel products (Royal Society 2019). This will require process changes for downstream oil/gas companies.
Upstream oil companies	Dedicated	Development of DAC for sustainable non-biological fuel can affect their market share if demand for this increases.
Aviation	Dedicated	Non-biological synthetic fuel can be used in all transport modes used today and is one of the most promising utilisations options available for DAC (Royal Society 2019).
Road transport		
Shipping		
DAC companies	Dedicated	They will be affected by their own actions.
Wildlife NGO	Non-dedicated	Wildlife and environmental themselves will not be affected by climate change or DAC plants
Environmental NGO		
CO_2 Storage organisations	Non-dedicated	CO_2 Storage organisations do not require DAC as most CO_2 will be sourced from industrial sources or electricity production.
Research institutes (TNO)	Non-dedicated	Research institutes are not affected and cannot block DAC development.

Table 36: Actor dedication to problem.

9.3 Summary

Table 37: Interdependency table for actors related to DAC, summarises which actors are critical/non-critical, dedicated/non-dedicated and have similar/conflicting interests. This is viewed from the point of view of the DAC company.

Within the transport industry no suitable alternative is available which can reduce emissions and maintain growth in industry apart from non-biological synthetic fuel, though this is at a low TRL (Royal Society 2019). This is also one of the most promising options available for DAC as a utilisation option. To explore this option the airline Transavia in the consortium exploring using DAC in production of sustainable non-biological fuel as jet fuel at the Rotterdam-Hague airport (Virardi 2019). Due to this, the aviation sector has been categorised as a critical actor. For the road transport sector other options such as electric and hydrogen vehicles are options (Royal Society 2019). For shipping options such as ammonia fuel options are available (Royal Society 2019). Due to this these have actors have been categorised as non-critical actors. For TNO support will need to be actively perused due to no inherent benefit of DAC to TNO. Those with conflicting objectives will need to be appeased. This can already be seen to be happening from upstream oil/gas companies who are collaborating with the DAC company Global Thermostat with Exxonmobil (Exxon Mobil 2019). This can be seen as a sharing of potential profits for the upstream oil/gas section of Exxonmobil with DAC companies in production of sustainable non-biological fuel. Those who are strong allies are more likely to naturally align themselves with DAC companies.

	Dedicated actors		**Non-dedicated actors**	
	Critical actors	**Non-critical actors**	**Critical actors**	**Non-critical actors**
Similar / Supportive interests and objectives	**(Strong allies)** • EU (DG for climate action, DG for environment) • Member states (Transport ministry, Climate change ministry, Environment ministry) • Downstream oil / gas • Aviation transport sector	**(weak allies)** • Road transport sector • Shipping transport sector	**(allies, hard to activate)** • Coal sector • NG sector • Energy suppliers • CO_2 storage organisations	**(Not required involvement)** • Research institutes (TNO)
Conflicting interests and objectives	**(Blockers of change)** • Upstream oil/gas • Other CDR actors	**(Potential critics)** • Environmental NGO • Wildlife NGO	**(Potential blockers)** • RES	**(Need little attention)**

Table 37: Interdependency table for actors related to DAC.

Chapter 10: Conclusion and recommendations

In this final chapter the research question and sub-questions are evaluated against the findings from this research. This also outlines the main methods in which these questions were answered. It is finalised by a set of recommendations for Direct Air Capture (DAC) companies.

10.1 Research question development

Technological implementation into wider society can be described through Multi-Level Perspective (MLP) theory (Loorbach, Frantzeskaki en Avelino 2017). This says that such systems sit in a regime with components that relate to technology and society, making such systems stable. These components are: market user preference; industry; policy; culture; and technology. For new technology to be implemented into society there needs to be change in these components which allows for a new social-technical regime.

It has been identified that Direct Air Capture and Storage (DACS) is technically possible (House, et al. 2011), though limitations exist regarding a lack of validation of DAC technologies. (The National Academies of Science, Engineering and Medicine 2018). This can be thought of as the technological component of the socio-technical regime. This limitation is not just limited to validation but there is also missing research which allows for comparison of DAC technologies.

It has also been identified that there are limitations regarding the policy component of the regime to support DAC development by industry experts (The National Academies of Science, Engineering and Medicine 2018). These policies relate to a funding mechanism for DAC and carbon pricing.

Further limitations for DAC include, the fact that there is no available analysis of the stakeholder environment in which DACS sits (The National Academies of Science, Engineering and Medicine 2018). This relates to the cultural component of the socio-technical regime.

From these three significant issues, which relate to technical, policy and stakeholder domains, the following research question is composed.

Research question:

"How do the most promising available DAC systems described in literature compare, where does DAC fit in current policy and how can DAC companies best position themselves to maximise the potential of their technology?"

Sub questions:

1. *Of the representative DAC technologies currently outlined in literature, how do these technologies compare with each other?*
2. *Of the policies that already exist regarding the tackling of climate change, what potential affect will they have on development of DACS?*

3. *What are the resources and interdependencies between the main actors which are related to DAC and can DAC companies leverage their position as stakeholders reduce their CO_2 emissions?*

10.2 Methods and results

To answer the main research, question a number of common criteria for DAC systems were taken from literature. These were then categorised as contributing to particular objectives which can contribute to an increase likelihood of DAC having widespread implantation. These objectives are High level of social acceptance, Low emissions associated with DAC, Increased fairness of financial funding of climate change measures and Low level of investment requirements. This therefore allows for the analysis of each DAC system and different policies against each other by measuring them against these criteria. A detailed analysis of the social acceptance criteria is not included as this requires a detailed description of each system using the design philosophy of inherently safe design and layers of protection which is beyond the scope of an Engineering and Policy Analysis Master thesis. In relation to how DAC companies can position themselves a stakeholder analysis is carried out.

10.2.1 Sub-question 1

To answer the first research sub-question two DAC systems are selected for further analysis: the humidity swing and the HT solvent system. These were selected because there is relativity little information regarding the humidity swing, though it is a promising alternative (Fasihi, Efimova and Breyer 2019). An HT solvent system such a system has been described that shows promise in terms of CO_2 production rate and cost, though there are a number of technical unknowns in the description given.

For the humidity swing a system was developed that can produce CO_2 at the same rate and concentration as described for the HT solvent system in (Keith, Holmes, et al. 2018). The humidity swing system used solar thermal energy as the energy source and an amine system to concentrate the CO_2 from the humidity swing sub-system.

For the HT solvent system, a Process Flow Diagram (PFD) was developed from the description given in (Keith, Holmes, et al. 2018). This then modelled the CO_2 absorber, air contactor, pellet reactor, calciner and steam slacker. The flow rates calculated and used in the PFD were higher than reported values in (Keith, Holmes, et al. 2018). This is because the extent of the reactions are unknown and it was assumed that the reaction went to completion which means that there was no recycle requirements. The higher flow rates were hypothesised as resulting in the larger energy requirements for the units calculated and used in the developed PFD than in reported in (Keith, Holmes, et al. 2018).

The results from the humidity swing process modelling were measured against the developed criteria. For the HT solvent system, the reported values were measured against these criteria. This was done for the HT solvent system because many of the units used in this system are unusual and require non-standard modelling processes therefore making the reported values in (Keith, Holmes, et al. 2018) having a lower uncertainty than those calculated through the modelling of the system. The results of this can be found Figure 51: Results from technical analysis of both systems, where dark blue is the results from the humidity swing system ad orange the HT solvent system.

For the humidity swing system significant uncertainty lies in OPEX, levelised cost, low energy use and CAPEX value. This is because the degradation rate of the sorbent been unknown. Another significant uncertainty was the ratio of mols of H_2O to CO_2 required to regenerate the sorbent. To determine these values, experimentation on the sorbent used in humidity swing will need to be carried out. This contributes to the OPEX and levelised cost uncertainty as it is not known how much water is required to liberate the CO_2 from the sorbent which determines the thermal energy requirements and, in turn, the size of the solar thermal system and therefore the CAPEX value.

These uncertainties do not exist for the low emissions associated with DAC. For this it can be seen there are associated emissions for the humidity swing, resulting from the use of grid electricity. The HT solvent system has no emissions associated with DAC as it captures the CO_2 released from the NG which is used as the energy source.

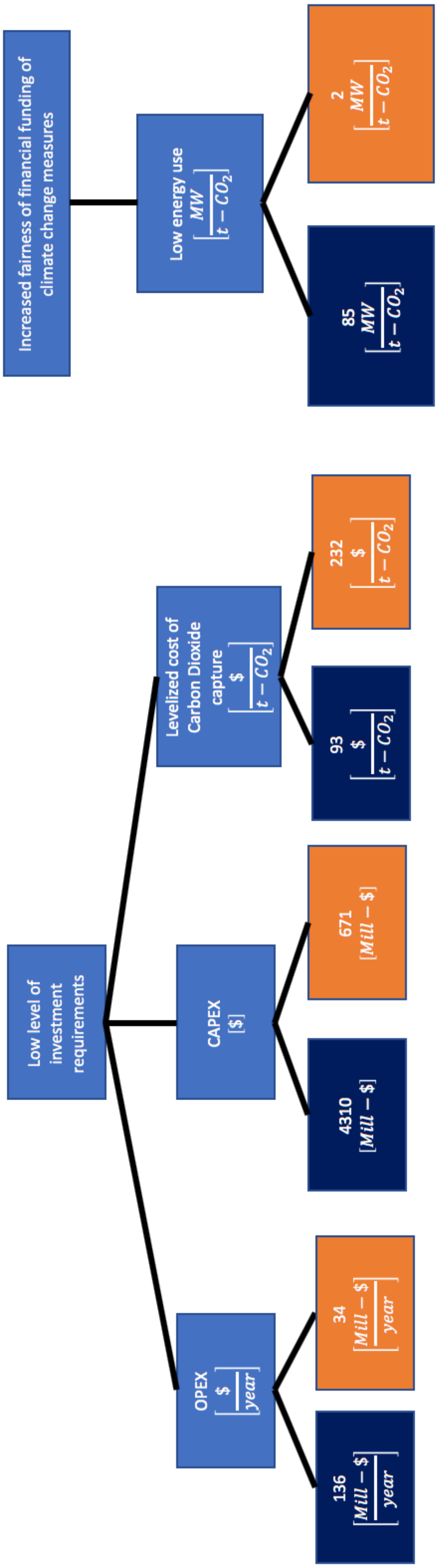


Figure 51: Results from technical analysis of both systems.

During the technical analysis, the research explored combining an amine carbon capture system which captures CO_2 from flue gas with a humidity swing system which captures CO_2 from the atmosphere and uses the waste heat from the stripper re-boiler as a heat source for the humidity swing system was explored. This type of system is depicted in figure 52: Humidity swing combined with amine carbon capture from flue gas. From this it was found that such a system could capture an extra 8% more CO_2 than the amine system alone. Under a scenario where there is no other waste heat option available this would contribute to fairness of financial funding as it would require almost no extra energy requirements [MW/t-CO_2]. The only increase in this value would come from the pumping of air, which only contributes to 1% of the operating cost. Such a system was also considered in sub-question 3 of the research question in relation to the potential between DAC companies and energy producers from NG or coal.

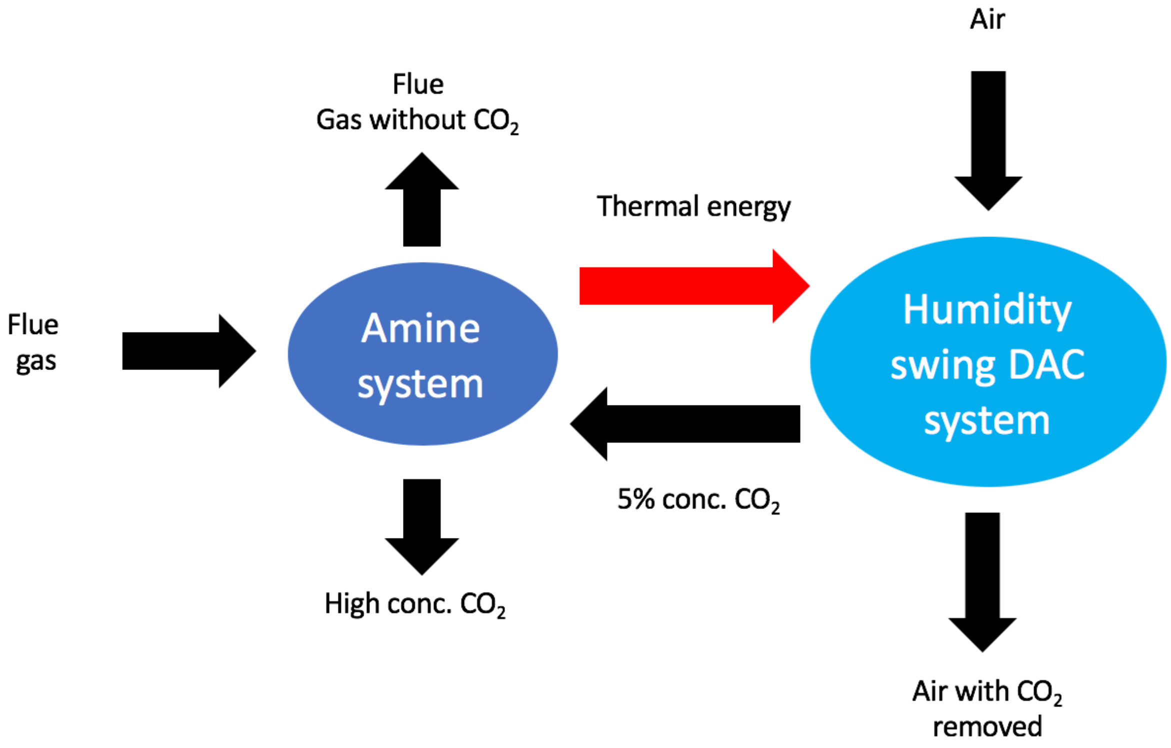


Figure 52: Humidity swing combined with amine carbon capture from flue gas.

10.2.2 Sub-question 2

For sub-question 2 a number of polices were selected which are designed to help tackle climate change and analysed in what affect they will have on DACS development. This analysed the following contract for differences (CfD), European Trading System (ETS), Clean Development Mechanism (CDM) and the 45Q legislation. These were selected as they are all significantly different. The CfD is a funding mechanism where consumers of electricity pay for green technology, the ETS is a system where large emitters pay a penalty for emissions, the CDM allows emissions to be offset to account for climate change targets and the 45Q legislation applies a tax credit for DACS***.***

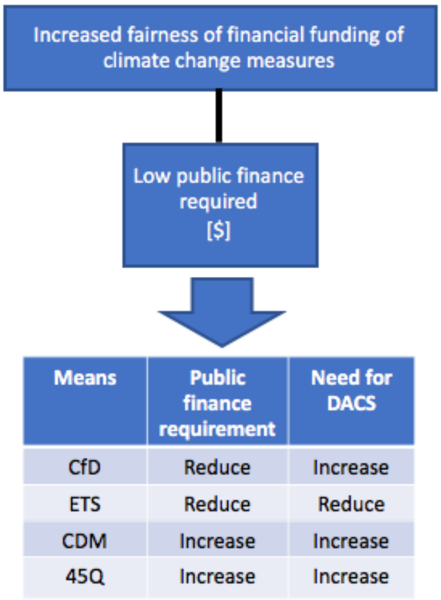
The purpose of the analysis was to determine whether these policies will be supportive of DACS development. Furthermore, the question of whether the policies are deemed fair was analysed. This was analysed because policies that are deemed fair are more likely to be implemented. The criterion of fairness for policy was identified as policy which does not require a significant public funding. This connection between low public finance requirements and fairness was selected as by reducing public finance requirement it allows funding for other projects.

To answer this a system diagram is developed using the method outlined in (Enserink, et al. 2010). The results of this can be found in figure 53: Summary of system diagram. The results showed that all policies except the ETS can be used to support DACS. The reason the ETS cannot be used to support DACS is that the ETS does not account for emissions which are offset which DACS is capable of doing (Fasihi, Efimova and Breyer 2019). The ETS would need to be adapted to allow for offsetting of emissions if it is to support DACS development.

The CDM can be used to support DACS development as it allow for offsetting of emissions. The CfD can also be used to support DACS development as the humidity swing DACS system can be combined with amine capture system for NG or coal combustion used in electricity production. Such a system can be funded through a CfD which passes the cost of such a system to the energy consumer. The 45Q legislation can be used to support DACS development though the tax credit would needs to be of greater value than the cost of capture for it to be effective.

The CDM requires public funding as it is a system which funds emission mitigation projects via public funding. The 45Q legislation also requires funding as it applies a tax rebate for DACS. The ETS does not require public funds as it applies a penalty for emissions. A CfD also does not require public funds as it passes on the cost to the energy consumer.

Looking at both the public finance requirements and need for DACS it can be seen that the CfD policy is the most attractive. This policy requires the humidity swing system to be implemented into an amine capture system which captures CO_2 from flue gas from the combustion of NG or coal for electricity production. This type of system can be used to top up the CO_2 captured from the flue gas by the humidity swing system capturing CO_2 from air with no extra thermal energy requirements and the cost is passed onto the consumer. This reduces the burden on public finances. Also, if it is assumed that those consumers who use the most electricity emit the most CO_2 those who emit the most should pay the most.



Means	Public finance requirement	Need for DACS
CfD	Reduce	Increase
ETS	Reduce	Reduce
CDM	Increase	Increase
45Q	Increase	Increase

Figure 53: Summary of system diagram.

10.2.3 Sub-question 3

For this question an actor analysis was carried out. This identified examples of actual and potential collaboration between DAC companies and other stakeholders. This also mapped the relationship between these actors and analysed their objectives and resources. This was used to identify wither an actor was an ally or a threat and how hard they will be to activate in relation to implementing DAC.

This analysis found that, apart from those directly affected by climate change, that stakeholders within the energy sector are most likely to be interested by DAC. This is because DACS can be used to offset their emissions. From the technical analysis on using waste heat from the amine system in the humidity swing system to capture CO_2 from the atmosphere bears relevance to energy producers from natural gas and coal. This is because they can use such a system to capture CO_2 from flue gas with the amine system and with no extra thermal energy requirements to capture CO_2 directly from the atmosphere with the humidity swing system. Therefore, the main findings from this is that DAC companies should look to collaborate with this energy producers from NG and coal. Though there is threat that this will damage the public image of DAC companies as it continues societies dependency on fossil fuels.

10.3 Future research direction

As stated in sub-section 10.2.1 Sub-research question 1, a significant uncertainty for the humidity swing was the ratio of mols of H_2O to CO_2 required to regenerate the sorbent. In this sub-section the assumption that there is a 1:1 mol ratio requirement will be re-evaluated. The reaction taking place in the humidity swing is shown through eqn. (1) to eqn. (3) and described as before in box 7: Humidity swing reaction mechanism.

Box 7: Humidity swing reaction mechanism

$$OH^- + CO_2 \leftrightarrow HCO_3^- \; eqn.(1) \; carbon \; dioxide \; adsorption \; (dry)$$
$$CO_3^{2-} + CO_2 + H_2O \;\leftrightarrow HCO_3^- + HCO_3^- \; eqn.(2) \; water \; adsorption \; (dry)$$
$$CO_3^{2-} + CO_2 + H_2O \;\leftrightarrow HCO_3^- + HCO_3^- \; eqn.(3) \; carbon \; dioxide \; de-adsorption \; (wet)$$

When water is added to the system it would be expected that CO_2 is adsorbed and released when removed water is removed in accordance with mass action law. Though in reality the opposite is true. This peculiarity is explained through Gibbs free energy of CO_3^{2-}. Gibbs free energy is the thermodynamic potential energy available. When water is exposed to CO_3^{2-} the available energy to react with CO_2 reduces. Which promotes the backwards reaction in *eqn. (3)*. This means that when CO_3^{2-} has this water removed it has an increased amount of energy to react with CO_2 which promotes the forward reaction in *eqn. (2)* (Wang, Lackner and Wright, 2011).

Looking at the reaction taking place in accordance with equation 3 when water is added CO_2 is released. Though also through this reaction water is also released. This means that in theory only one water molecule is required to release all CO_2 through a chain reaction mechanism as every time a CO_2 molecule is released then a water molecule is released. Despite this, in reality the water molecule would need access to each carbonate ion and therefore the reaction is not limited by the ratio of H_2O to CO_2 molecules.

Due to this it is hypothesised that the 1:1 mol ratio of CO_2 and water is an over-estimation and that less water is required.

If this is true, it will result in a smaller OPEX, CAPEX and levelised cost of CO_2 for the humidity swing. Interestingly this will mean that when using the system proposed where the humidity swing is combined with a flue gas amine CO_2 capture system to top up the CO_2 captured from the flue gas it will be able to capture more CO_2 from the atmosphere than the 8% calculated in this thesis.

Therefore, an extremely interesting area of future research would be to determine the relationship between the number of mols of H_2O added to the system and CO_2 released during the de-adsorption step.

10.4 Recommendations

The most novel findings of this research is that using waste heat from an amine system that captures CO_2 from flue gas can be topped up in terms of the CO_2 captured if the waste heat from the stripper re-boiler is used as a thermal energy source for a humidity swing DAC system which captures CO_2 from the atmosphere. This can be funded through a CfD which passes the cost of this process onto the consumer. This would could therefore be deemed fair as it does not burden public finances.

The following list gives more specific recommendations in terms of cost, fairness and stakeholder analysis.

1. Recommendations cost:
    a. For the humidity swing future work should determine degradation rate of sorbent and therefore the optimum number of sorbent changes per year.
    b. For the humidity swing the number mols of H_2O required per mols of CO_2 captured should be determined.
    c. The thermal solar system should be placed outside Europe to maximise the DNI.
    d. Focus should be on use of renewable energy as energy source for the systems. This is because under a scenario where the CO_2 is utilised this makes the process more sustainable. For the humidity swing, solar thermal is the most suitable renewable energy source. For the HT solvent process the system will need to be adapted so that the solvent is regenerated via electrolysis. This can then use a range of different renewable energy sources.
    e. Energy requirements for the humidity swing system are very high and therefore smaller plants should be considered.
    f. Regularly update associated CO_2 emissions for the DAC plant.
    g. Consider using the waste heat from stripper re-boiler as heat source for humidity swing system.
2. Recommendations fairness:
    a. Policy related to CO_2 offsetting align most closely with development of DACS. Policies which reduce CO_2 emissions such as ETS reduce the need for capture of CO_2. CO_2 offsetting policies will reduce the need for change to the current systems and are therefore more likely to be deemed fair though it may have negative affect on public image by continuing dependency on fossil fuels.
    b. Develop a framework to helpl which to determine CO_2 emissions from cradle to grave of long-term storage of CO_2.
    c. Monitor the effect processing a large volume of air has on the local environment and the process itself. This will make it less likely to be blocked by local environment agencies.
3. Recommendation stakeholders:
    a. Large emitters can become allies as DAC companies can offset their emissions through CO_2 arbitrage or by using CO_2 as a feedstock material for utilisation methods.
    b. Other actors which can mitigate emissions such as afforestation pose threat as they can mitigate at lower price, though they do not show same potential for CO_2 utilisation.
    c. DAC companies should look for investment where it is de-risked from other parts of the value chain of CO_2 utilisation and storage. This can be done by looking for investment solely on DAC and has no dependency on other parts of the value chain for CO_2 utilisation and storage.
    d. There should be a CO_2 storage buffer so that downstream stakeholders are not negatively affected by DAC downtime.
    e. A contractional obligation from downstream stakeholders that downtime on their part does not negatively affect DAC companies.

Appendix

Appendix-A: Fan energy requirements for humidity swing.

- For 222.54 [t-CO_2/hr], this would require a total flow rate of air of 4451 [t-CO_2/hr] for 5% capture rate.
- $61\left[\frac{kWhr}{ton}\right]*3321.6\left[\frac{ton}{hr}\right]=202618[kW]$
- $0.0357\left[\frac{\$}{kWhr}\right]*61\left[\frac{kWhr}{ton}\right]*3321.6\left[\frac{ton}{hr}\right]=7233\left[\frac{\$}{hr}\right]$
- $\frac{7233\left[\frac{\$}{hr}\right]}{166.08\left[\frac{t-CO_2}{hr}\right]}=43.55\left[\frac{\$}{hr}\right]\rightarrow\frac{43.55\left[\frac{\$}{hr}\right]}{166.08\left[\frac{t-CO_2}{hr}\right]}=\mathbf{0.26}\left[\frac{\$}{\boldsymbol{t-CO_2}}\right]$

Appendix-B: Sorbent sizing and costing.

- $24.033[Hrs]*0.0398\left[\frac{m^3-sorbent}{kg-CO_2}\right]*166.08\left[\frac{t-CO_2}{hr}\right]*10^3\left[\frac{kg-CO_2}{t-CO_2}\right]$ $=158860[m^3-sorbent]$
- $1090\left[\frac{kg-sorbent}{m^3-sorbent}\right]*158860[m^3-sorbent]=173157342[kg-sorbent]$
- 6th 10th rule: $3\left[\frac{\$}{kg-CO_2}\right]\left(\frac{173157342[kg-sorbent]}{1[kg-sorbent]}\right)^{0.6}=263{,}139[\$]$
- $Sorbent\ changes\ per\ year=1;2\left[\frac{min}{cycle}\right];262800\left[\frac{cycles}{year}\right]$
- $1\left[\frac{sorbent}{year}\right]*263139\left[\frac{\$}{sorbent}\right]=263{,}139\left[\frac{\$}{year}\right];This\ also\ the\ CAPEX$
- $\frac{526.279[\$]}{166.08\left[\frac{t-CO_2}{hr}\right]*365[days]*24\left[\frac{hours}{day}\right]}=0.36\left[\frac{\$}{t-CO_2}\right]$

Appendix-C: Solar thermal system.

- $inlet\ flow\ rate\ of\ CO_2 = 222\left[\frac{t-CO_2}{hr}\right]$
- $222.54\left[\frac{t-CO_2}{hr}\right] * \frac{2[mins]}{60\left[\frac{mins}{hr}\right]} = 7418[t-CO_2]$
- Water replaces CO_2 at ratio of 1 [mol]: 1 [mol] and 40650 Joules required to evaporate 1 mol of water.
- $\frac{7418[t-CO_2]}{0.04401\left[\frac{t-CO_2}{mol-CO_2}\right]} * 40650\left[\frac{J}{mol-CO_2}\right] = 6852[MJ]$
- $173157342[kg-sorbent] * 3\left[\frac{\$}{kg-sorbent}\right] = 519{,}472{,}026[\$]$
- $12{,}421{,}372[m^2-solar] * 308\left[\frac{\$}{m^2}\right] = 3826[\$mill]$

Appendix-D: PFD of Aspen plus carbon capture system.

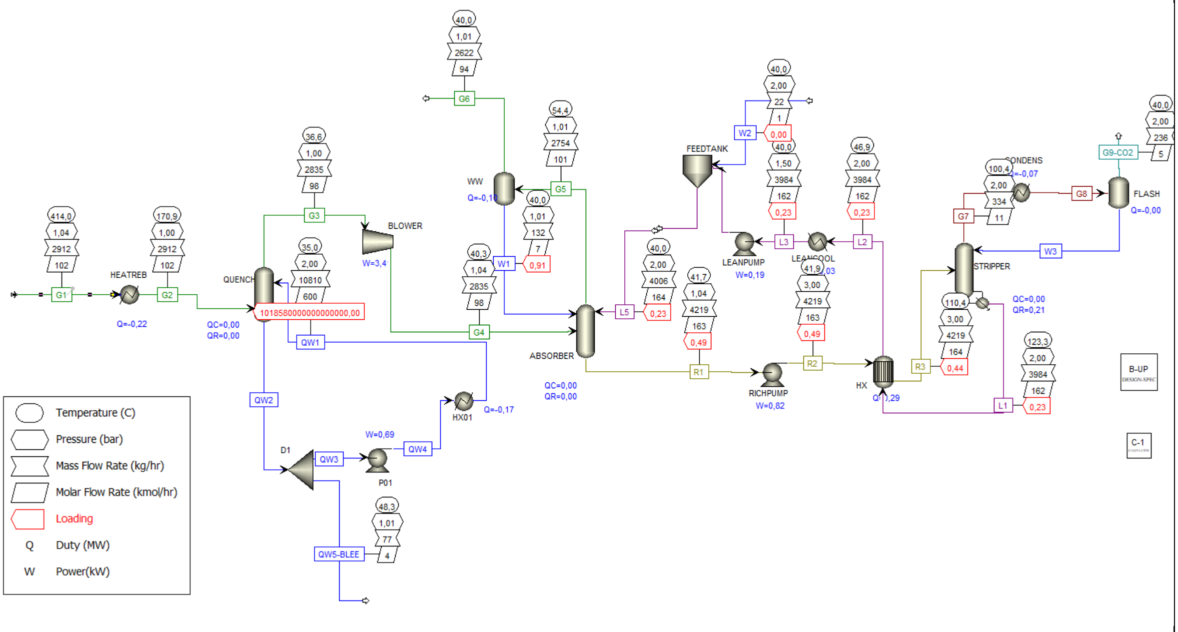


Figure A-1: Screenshot of Aspen Plus model for amine system

Appendix-E: Waste heat calculations.

The sorbent has a capacity:

$$1[m^3 - sorbent] \rightarrow 0.0398[kg - CO_2]$$

Heat of vaporisation of water:

$$1[kg - water] \rightarrow 2.3[MJ]$$

From Aspen model there is 770[kJ] cooling requirement for the whole system.
If it is assumed that the water replaces CO_2 on 1:1 mol ratio then it can be determined how much CO_2 can be captured by the system:

$$1[kg - H_2O] \rightarrow 55.5[mol - H_2O]$$
$$55.5[mol - H_2O] \rightarrow 2.3[MJ]$$

The total cooling requirements of the system is 770[kJ] and this can therefore evaporate 18.4 [mol-H_2O] which is 0.81[kg-CO_2] if it is assumed that the water replaces CO_2 on 1:1[mol] ratio.
It takes 2[mins] for the desorption and 24[hours] for the adsorption process to happen. Therefore, the adsorbent and desorption sorbent should switch every 24 hours. The following calculation determines the flow rate:

Sorbent volume:

2 [mins] happens 540 times over 24 hours. The following calculation calculates the volume requirements:

$$\frac{0.81[kg - CO_2]}{0.0398\left[\frac{kg - CO_2}{m^3 - sorbent}\right]} = 20.35[m^3 - sorbent]$$

$$20.35[m^3 - sorbent] * 540 = 10987[m^3 - sorbent]$$

Total volume is therefore 21974[m^3] as there should be the same of sorbent adsorbing and desorbing.

Flow rate:
Every 2 mins 0.81[kg-CO_2] is released which in an hourly basis is **24.30[kg-CO_2/hr]**.

With the production rate at 232 [kg-CO_2/hr] and inlet flow rate of 310.85[kg-CO_2/hr] the use of the condenser to heat the air to dehumidify the system results in an increase 7.82% capture rate. With a sorbent cost of 3[$/kg] and a density of 1090[kg/m^3] then the cost of the humidity swing system would be 71,854,980[$]. The cost of the amine system without the humidity swing is 1,800,700[$]. The increased operating cost would be dependent on how often the sorbent needs replaced, which is a significant unknown.

Appendix-F: Mass balance for HT

	Stream	1(a)	1(b)	1	2	3	4	5	5(a)	6	7(a)	7(b)	7(c)	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	Stream
	Flow rate	251000,0	110,5	251110,5	252000,0	3976,6	3087,1	5188,3	1988,3	3200,0	7000,0	6615,8	384,2	6056,4	559,4	2877,8	3178,6	252,0	5964,9	3365,7	6,0	3102,3	3,4	380,8	0,0	380,8	561,0	567,0	3096,3	2798,7	2798,7	171,0	171,0	201,0	531,0	0,0	0,0	2619,3	2798,7	2799,0	Flow rate
Mass flow [t/hr]	CO2	150,6	6,9	157,5	40,3	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	36,4	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	-0,1	0,0	-0,1	0,0	0,0	166,1	166,1	166,0	0,0	0,0	0,0	0,0	0,0	0,0	CO2
	O2	57730,0	1,2	57731,2	57859,2	0,0	3,7	3,7	3,7	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	2,3	3,7	0,0	0,0	3,7	0,0	0,0	0,0	0,0	0,0	0,0	3,7	0,0	0,0	2,3	2,3	2,3	0,0	0,0	0,0	2,3	0,0	0,0	O2
	N2	190659,6	87,1	190746,7	191091,6	0,0	4,5	4,5	4,5	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	181,4	4,5	0,0	0,0	4,5	0,0	0,0	0,0	0,0	0,0	0,0	4,4	0,0	0,0	2,6	2,6	2,6	0,0	0,0	0,0	2,6	0,0	0,0	N2
	H2O	2459,8	15,3	2475,1	3024,0	3528,1	2704,5	4482,4	1697,6	2784,8	5875,3	5875,3	0,0	5306,0	569,4	2521,2	2784,8	31,9	5225,7	566,2	382,0	124,7	0,0	0,0	0,0	0,0	561,1	566,2	119,5	0,0	0,0	0,0	0,0	30,1	531,0	0,0	0,0	710,8	0,0	680,7	H2O
	[K+]	0,0	0,0	0,0	0,0	283,0	223,9	417,4	165,9	251,5	547,4	547,4	0,0	473,6	73,8	225,0	248,6	0,0	448,9	0,6	0,0	246,2	0,0	0,0	0,0	0,0	0,0	0,6	245,6	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	[K+]
	[OH-]	0,0	0,0	0,0	0,0	67,7	27,9	51,0	14,0	37,0	138,8	138,8	0,0	113,3	25,5	53,8	59,5	0,0	81,7	0,1	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,1	-0,1	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	[OH-]
	CaCO3	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	384,2	0,0	384,2	0,0	0,0	0,0	0,0	0,0	0,0	0,0	6,0	50,3	3,4	380,8	0,0	380,8	0,0	0,0	50,3	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	CaCO3
	[CO3,2-]	0,0	0,0	0,0	0,0	97,7	122,6	229,3	102,6	126,7	175,4	175,4	0,0	163,5	11,8	77,7	85,8	0,0	200,3	0,0	0,0	189,0	0,0	0,0	0,0	0,0	0,0	0,0	189,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	[CO3,2-]
	[Ca(OH)2]	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	2798,7	0,0	2484,0	0,0	0,0	0,0	0,0	0,0	0,0	2484,0	2798,7	2798,7	0,0	0,0	0,0	0,0	0,0	0,0	0,0	2798,7	0,0	[Ca(OH)2]
	CaO																					0,0								0,0	0,0		0,0	0,0	0,0	0,0	0,0	0,0	0,0	2118,4	CaO
	K2CO3																					0,0								0,0	0,0		0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	K2CO3
	sum	251000,0	110,5	251110,5	252015,1	3976,6	3087,1	5188,3	1988,3	3200,0	7121,0	6736,8	384,2	6056,4	680,4	2877,8	3178,6	252,0	5964,9	3365,7	388,0	3102,3	3,4	380,8	0,0	380,8	561,0	567,0	3096,3	2798,7	2798,7	171,0	171,0	201,0	531,0	0,0	0,0	715,7	2798,7	2799,0	sum
	Charge	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	Charge
Composition (mass fraction)	CO2	0,1	6,3	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	14,4	0,0	0,0	0,0	0,0	0,0	0,0	0.0	0,0	0,0	0,0	0,0	0,0	0,0	97,1	97,1	82,6	0,0	0,0	0,0	4,4	0,0	0,0	CO2
	O2	23,0	1,1	0,2	23,0	0,0	0,0	0,1	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,9	0,1	0,0	0,0	0,1	0,0	0,0	0.0	0,0	0,0	0,0	0,1	0,0	0,0	1,4	1,4	1,2	0,0	0,0	0,0	95,6	0,0	0,0	O2
	N2	76,0	78,8	0,8	75,8	0,0	0,0	0,1	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	72,0	0,1	0,0	0,0	0,1	0,0	0,0	0.0	0,0	0,0	0,0	0,1	0,0	0,0	1,5	1,5	1,3	0,0	0,0	0,0	0,0	0,0	0,0	N2
	H2O	1,0	13,9	0,0	1,2	0,9	0,9	86,4	0,9	87,0	87,6	0,9	0,0	87,6	101,8	87,6	87,6	12,7	87,6	16,8	6366,7	4,0	0,0	0,0	0.0	0,0	100,0	99,9	3,9	0,0	0,0	0,0	0,0	15,0	100,0	0,0	0,0	0,0	0,0	24,3	H2O
	[K+]	0,0	0,0	0,0	0,0	0,1	0,1	8,0	0,1	7,9	7,8	0,1	0,0	7,8	13,2	7,8	7,8	0,0	7,5	0,0	0,0	7,9	0,0	0,0	0.0	0,0	0,0	0,1	7,9	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	[K+]
	[OH-]	0,0	0,0	0,0	0,0	0,0	0,0	1,0	0,0	1,2	2,9	0,0	0,0	1,9	4,6	1,9	1,9	0,0	1,4	0,0	0,0	0,0	0,0	0,0	0.0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	[OH-]
	CaCO3	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,1	0,0	1,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	100,0	1,6	100,0	100,0	0.0	100,0	0,0	0,0	1,6	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	CaCO3
	[CO3,2-]	0,0	0,0	0,0	0,0	0,0	0,0	4,4	0,1	4,0	2,7	0,0	0,0	2,7	2,1	2,7	2,7	0,0	3,4	0,0	0,0	6,1	0,0	0,0	0.0	0,0	0,0	0,0	6,1	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	[CO3,2-]
	[Ca(OH)2]	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	83,2	0,0	80,1	0,0	0,0	0.0	0,0	0,0	0,0	80,2	100,0	100,0	0,0	0,0	0,0	0,0	0,0	0,0	0,0	100,0	0,0	[Ca(OH)2]
	CaO																																			0,0	0,0	0,0	0,0	75,7	CaO
	K2CO3																																			0,0	0,0	0,0	0,0	0,0	K2CO3
	sum	100,0	100,0	100,0	100,0	100,0	100,0	100,0	100,0	100,0	101,1	101,8	100,0	100,0	121,6	100,0	100,0	100,0	100,0	100,0	100,0	100,0	100,0	100,0	100,0	100,0	100,0	100,0	100,0	100,0	100,0	100,0	100,0	100,0	100,0	0,0	0,0	100,0	100,0	100,0	sum

Table 1-A: HT Solvent full mass balance.

In		Out	
Stream	Flow	Stream	Flow
1(a)	150,60	2	40,32
12	36,36	19	0,00
31	0	23	0,00
32	0,00	27	166,08
15	0,00	-	
Total	186,96	**Total**	206,40

Table 2-A: CO_2 inlet/out balance.

In		Out	
Stream	Flow	Stream	Flow
1(a)	57730,00	2	57859,20
12	2,29	19	0,00
31	0,0	23	3,70
32	2,33	27	2,33
15	0,00	-	
Total	57734,62	**Total**	57865,22

Table 3-A: O_2 inlet/out balance.

In		Out	
Stream	Flow	Stream	Flow
1(a)	190659,60	2	191091,60
12	181,44	19	0,00
31	0,0	23	4,44
32	2,57	27	2,58
15	0,00	-	
Total	190843,61	**Total**	191098,62

Table 4-A: N_2 inlet/out balance.

In		Out	
Stream	Flow	Stream	Flow
1(a)	2459,80	2	3024,00
12	31,90	19	0,00
31	0,0	23	119,54
32	710,81	27	0,02
15	382,00	-	
Total	3584,52	**Total**	3143,56

Table 5-A: H_2O inlet/out balance.

Appendix-G: Methane composition.

$$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O$$

- $\dot{m}_{CH_4,in} = 6.3 \left[\frac{t-CH_4}{hr}\right]$; $\dot{n}_{CH_4,in} = 392768 \left[\frac{mol-CH_4}{hr}\right]$
- $\dot{n}_{CO_2,out} = 392768 \left[\frac{mol-CH_4}{hr}\right]$; $\dot{m}_{CO_2,out} = 17.29 \left[\frac{mol-CH_4}{hr}\right]$

Reported flow rate is $\dot{m}_{CO_2,out} = 17.46 \left[\frac{mol-CH_4}{hr}\right]$ which is approximately the same as the calculated value.

Appendix-H: Flow rates around CO_2 absorber.

Calculation absorbed CO_2		
Stream	**8**	**6**
OH^- conc. [mol/L]	1,1	0,68
mass (water)		2784775,36 [kg/hr]
Description	**Value**	**Unit**
mol difference OH^-	0,42	[mol/L]
CO_2 concentration (see reaction kinetics)	0,21	[mol/L]
CO_2	584802,8256	[mol/hr]
Atomic mass water	18	[g/mol]
Change in flow rate water	10,52	[t/hr]

` Table A-6: Ion change and flow rate across CO_2 absorber.

Concertation calculated by the following based on density of water:

$$\left[\frac{mol}{L}\right] = (mass\%) * \frac{1000 \left[\frac{g}{L}\right]}{\left[\frac{g}{mol}\right]}$$

Appendix-I: Air contactor modelling description.

The fluid package chosen was electrolyte NRTL using figure A-2:

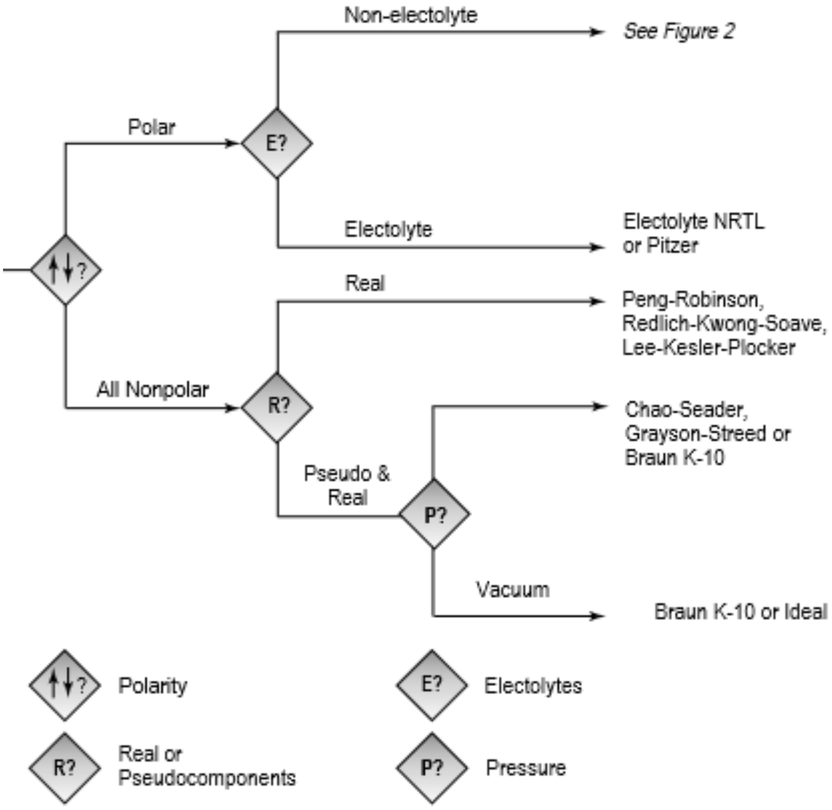


Figure A-2: Fluid package decision tree.

The CO_2 absorber was set up as follows:

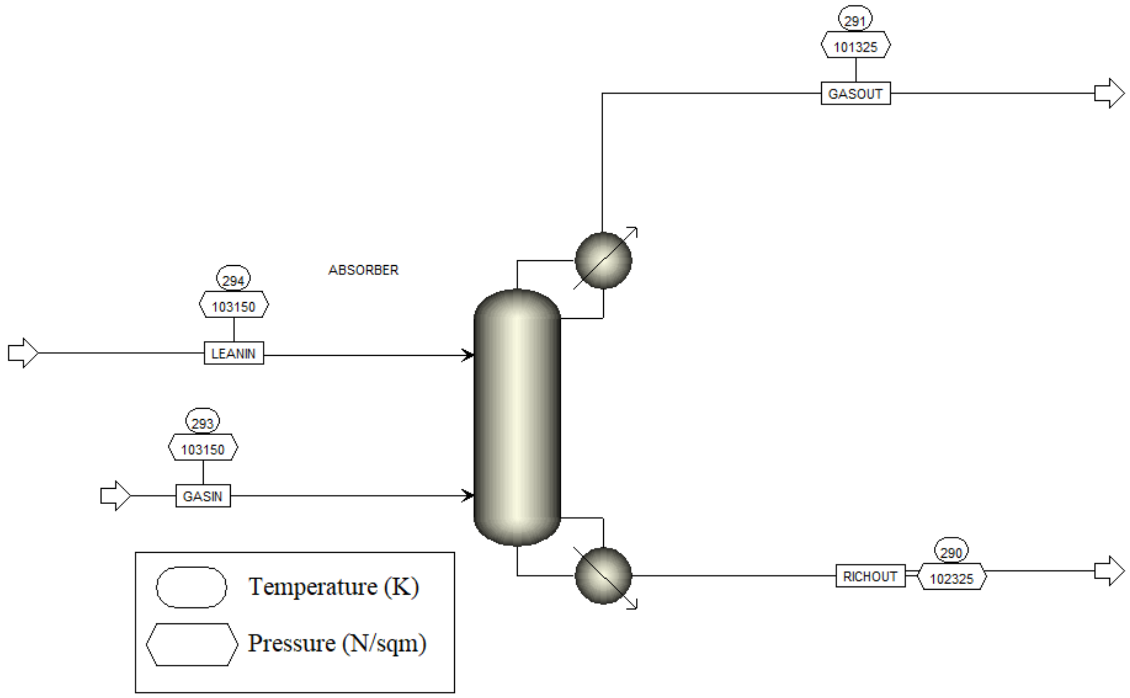


Figure A-3: Screenshot of Aspen plus model.

A calculator was set up to determine the flow rate of KOH required to capture the CO_2 quoted in (Keith, Holmes, et al. 2018). The contactor within (Keith, Holmes, et al. 2018) was approximated with the following characteristics:

Type	**Diameter**	**Number of theoretical trays**	**Tray spacing**	**Packing type**
Packed bed	30.48[m]	18	8[m]	Generic berl ceramic ½ inch

Table A-7: Absorber column description.

Appendix-J: Pellet reactor modelling.

$$\frac{dF_{K_2CO_3}}{dW} = -k * y_{K_2CO_3} * P;\ eqn\ (1)$$

$$Mole\ frac.: y_{K_2CO_3} = \frac{F_{K_2CO_3}}{F_T}; eqn\ (2)$$

$$eqn\ (2)\ \rightarrow eqn\ (1): \frac{dF_{K_2CO_3}}{dW} = -k * \frac{F_{K_2CO_3}}{F_T} * P; eqn\ (3)$$

$$F_T = F_{KCO_{3(aq)}} + F_{Ca(OH)_{2(S)}} + F_{KOH} + CaCO_{3(S)}; eqn\ (4)$$

$$eqn\ (4) \rightarrow eqn\ (3): \frac{dF_{K_2CO_3}}{dW} = -k * \frac{F_{K_2CO_3}}{F_{KCO_{3(aq)}} + F_{Ca(OH)_{2(S)}} + F_{KOH} + F_{CaCO_{3(S)}}} * P$$

$$collect\ terms: \left(1 + \frac{F_{Ca(OH)_{2(S)}}}{F_{K_2CO_3}} + \frac{F_{KOH}}{F_{K_2CO_3}} + \frac{F_{CaCO_{3(S)}}}{F_{K_2CO_3}}\right) dF_{K_2CO_3} = -k * P * dW; eqn\ (5)$$

$$Flow\ rates\ in\ terms\ F_{K_2CO_3}: F_{Ca(OH)_{2(S)}} = F_{K_2CO_3};\ F_{KOH} = 2 * F_{K_2CO_3};\ F_{CaCO_3} = F_{K_2CO_3}$$

$$subsitute\ these\ values\ into\ eqn\ (5)\ and\ integrate\ from\ inlet\ to\ outlet\ flow\ rate.$$

$$5\int_{dF_{K_2CO_3}(@inlet)}^{dF_{K_2CO_3}} dF_{K_2CO_3} = -kP\int_0^W dW$$

$$5 * (F_{K_2CO_3} - F_{K_2CO_3}(@inlet)) = -kPW$$

$$W = -5\frac{F_{K_2CO_3} - F_{K_2CO_3}(@inlet)}{k * P}$$

$$V_{K_2CO_3} = -5 * \frac{F_{K_2CO_3}}{k * P * \rho_{K_2CO_3}}$$

Repeat step in box for each component in reaction:

$$V_{Ca(OH)_2} = -5 * \frac{F_{Ca(OH)_2} - F_{Ca(OH)_2}(@inlet)}{k * P * \rho_{Ca(OH)_2}}$$

$$V_{CaCO_{3(S)}} = -5\frac{F_{CaCO_{3(S)}} - F_{CaCO_{3(S)}}(@inlet)}{k * P * \rho_{CaCO_{3(S)}}}$$

Repeating for KOH as mass balance differs for this component due to stoichiometry of reaction.

$$F_{K_2CO_3} = \frac{1}{2} F_{KOH};\ F_{Ca(OH)_{2(S)}} = \frac{1}{2} F_{KOH};\ F_{CaCO_{3(S)}} = \frac{1}{2} F_{KOH}$$

$$\frac{dF_{KOH}}{dW} = -k * \frac{F_{KOH}}{F_{KCO_{3(aq)}} + F_{Ca(OH)_{2(S)}} + F_{KOH} + F_{CaCO_{3(S)}}} * P$$

$$\frac{dF_{KOH}}{dW} = -k * \frac{F_{KOH}}{\frac{1}{2}\,F_{KOH} + \frac{1}{2}\,F_{KOH} + \frac{1}{2}\,F_{KOH} + F_{KOH}} * P$$

$$\frac{dF_{KOH}}{dW} = -k * \frac{3}{2} * P$$

$$7dF_{KOH} = -k * P * dW$$

$$2\int_{F_{KOH}(@inlet)}^{F_{KOH}} dF_{KOH} = -3 * k * P * \int_{0}^{W} dW$$

$$2 * F_{KOH} = -3 * k * P * W$$

$$V_{KOH} = -\frac{2 * [F_{KOH} - F_{KOH}(@inlet)]}{3 * k * P * \rho_{KOH}}$$

$$V_T = -\frac{5}{k * P}\left(\frac{F_{Ca(OH)_2} - F_{Ca(OH)_2}(@inlet)}{\rho_{Ca(OH)_2}} + \frac{F_{CaCO_{3(S)}} - F_{CaCO_{3(S)}}(@inlet)}{\rho_{CaCO_{3(S)}}} + \frac{F_{K_2CO_3} - F_{K_2CO_3}(@inlet)}{\rho_{K_2CO_3}}\right) - \frac{2[F_{KOH} - F_{KOH}(@inlet)]}{3k * P * \rho_{KOH}}$$

Total volume of reactor as a function of pressure temperature:

$$V_T = -\frac{1}{P * A * e^{\frac{-E_A}{R*T}}}\left(5 * \left[\frac{F_{Ca(OH)_2} - F_{Ca(OH)_2}(@inlet)}{\rho_{Ca(OH)_2}} + \frac{F_{CaCO_{3(S)}} - F_{CaCO_{3(S)}}(@inlet)}{\rho_{CaCO_{3(S)}}} + \frac{F_{K_2CO_3} - F_{K_2CO_3}(@inlet)}{\rho_{K_2CO_3}}\right] + \frac{2 * [F_{KOH} - F_{KOH}(@inlet)]}{3 * \rho_{KOH}}\right)$$

Note extent of reaction has not been included in mass balance.

The activation energy and Arrhenius constant are given by (Wiechers, Sturrock and Mairais 1975):

$$k = 6.57 * 10^3 e^{\left(\frac{43.10*10^3}{R*T}\right)}$$

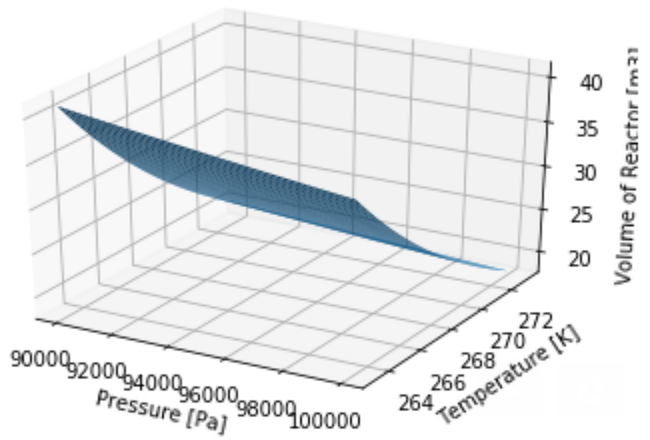


Figure A-4: Relationship between pressure, temperature and volume for pellet reactor.

Associated python code for sizing reactor:

```
import pandas as pd
import numpy as np
import math
from mpl_toolkits.mplot3d import Axes3D
import matplotlib.pyplot as plt

# Volume in terms of solid

def fun(P,T):
    global V

    V               =               (1/(P*(6.57*10**7)*math.exp((-
43.10*10**3/(8.314*T)))))*(5*(((335327084.62-
37773653.66)/2210)+((4340494.52-93915.48)/2710)+((0-
4246579.04)/2430))+(2/3)*((16824863.9-1257824.90)/2120))

    return V

fig = plt.figure()
ax = fig.add_subplot(111, projection='3d')
P = np.linspace(90000, 100000, num=50) #1[Bar] to 15[Bar]
T = np.linspace(263.15, 293.15, num=50) #0 Celsius to 220 celsius
X, Y = np.meshgrid(P, T)
zs = np.array([fun(x,y) for x,y in zip(np.ravel(X), np.ravel(Y))])
Z = zs.reshape(X.shape)

ax.plot_surface(X, Y, Z)

ax.set_xlabel('Pressure [Pa]')
ax.set_ylabel('Temperature [K]')
ax.set_zlabel('Volume of Reactor [m3]')

combined = np.ravel(X)*np.ravel(Y)*zs

d = {'Pressure [Pa]':np.ravel(X),
     'Temperature [K]':np.ravel(Y),
     'Volume [m3]':zs,
```

```
     'Combined[Pa*K*m3]':combined}

df    =    pd.DataFrame(d,columns=['Pressure    [Pa]','Temperature
[K]','Volume [m3]','Combined[Pa*K*m3]'])

row_number = df['Combined[Pa*K*m3]'].argmin()
min_vol = df['Volume [m3]'].argmin()

print(df.iloc[[row_number]])
print(df.iloc[[min_vol]])


plt.show()
```

Note, for the steam slacker and calciner many of the steps are the same as for the pellet bed and therefore have not been included in their derivations.

Appendix-K: Steam slacker modelling.

$CaO_{(s)}$ is solid therefore does not have partial pressure and therefore does not contribute to gas phase volume.

$$CaO_{(s)} + H_2O_{(g)} \rightarrow Ca(OH)_{2(g)}$$

$$\frac{dF_{H_2O_{(g)}}}{dW} = -k * y_{H_2O_{(g)}} * P$$

$$mole\ frac.: y_{CaO_{(s)}} = \frac{F_{CaO_{(s)}}}{F_T}$$

$$\frac{dF_{H_2O_{(g)}}}{dW} = -k * \frac{F_{H_2O_{(g)}}}{F_{H_2O_{(g)}} + F_{Ca(OH)_{2(g)}}} * P$$

$$\left(1 + \frac{F_{Ca(OH)_{2(g)}}}{F_{H_2O_{(g)}}}\right) dF_{H_2O_{(g)}} = -k * P * dW$$

Flow rates in terms of $H_2O_{(g)}$:

$$2 * dF_{H_2O_{(g)}} = -k * P * dW$$

Integrating from inlet to outlet:

$$2 * \int_{F_{H_2O_{(g)}}@inlet}^{F_{H_2O_{(g)}}} dF_{H_2O_{(g)}} = -k * P * dW$$

$$2 * \left(F_{H_2O_{(g)}} - F_{H_2O_{(g)}}@inlet\right) = -k * P * W$$

$$W = -2 * \frac{F_{H_2O_{(g)}} - F_{H_2O_{(g)}}@inlet}{k * P}$$

$$V_{H_2O} = -2 * \frac{F_{H_2O_{(g)}} - F_{H_2O_{(g)}}@inlet}{k * P * \rho_{H_2O}}$$

$$V_{H_2O} = -2 * \frac{F_{H_2O_{(g)}} - F_{H_2O_{(g)}}@inlet}{k * P * \left(\frac{P * molar\ mass}{8.314 * T}\right)}$$

For $Ca(OH)_{2(g)}$ the process is the same and yields:

$$V_{Ca(OH)_{2(g)}} = -2 * \frac{F_{Ca(OH)_{2(g)}} - F_{Ca(OH)_{2(g)}}@inlet}{k * P * \left(\frac{P * molar\ mass}{8.314 * T}\right)}$$

For the volume of the solid the reactor volume is calculated by the following:

$$V = \frac{F_{CaO_{(s)},inlet}}{k * \rho_{CaO_{(s)}}}$$

$CaO_{(s)}$ is solid therefore no dependency of volume on pressure or temperature.

$$V_T = -\frac{2}{A * e^{\frac{-E_A}{R*T}} * P}\left(\frac{F_{H_2O_{(g)}} - F_{H_2O_{(g)}}@inlet}{\left(\frac{P * molar\ mass}{8.314 * T}\right)} + \frac{F_{Ca(OH)_{2(g)}} - F_{Ca(OH)_{2(g)}}@inlet}{\left(\frac{P * molar\ mass}{8.314 * T}\right)} + \frac{F_{CaO_{(s)},inlet}}{\rho_{CaO_{(s)}}}\right)$$

From paper Pressure is equal 100[k-Pa] and temperature range is 300 Celsius to 520 Celsius. Activation energy and Arrhenius constant are (Criado, Alonso en Abanades 2014):

$$E_A = 59.4\left[\frac{kJ}{mol}\right]; A = 2.5 * 10^6\left[\frac{1}{s}\right]$$

Appendix-L: Calciner modelling.

$$CaCO_{3(s)} \rightarrow CaO_{(s)} + CO_{2\,(g)}$$

$$\frac{dF_{CaCO_{3(s)}}}{dW} = -k * y_{CaCO_{3(s)}} * P;\ eqn\ (1)$$

$$Mole\ frac.:\ y_{F_{CaCO_{3(s)}}} = \frac{F_{F_{CaCO_{3(s)}}}}{F_T}; eqn\ (2)$$

$$eqn\ (2) \rightarrow eqn\ (1): \frac{dF_{F_{CaCO_{3(s)}}}}{dW} = -k * \frac{F_{F_{CaCO_{3(s)}}}}{F_T} * P; eqn\ (3)$$

$$F_T = F_{CaCO_{3(s)}} + F_{CaO_{(s)}} + F_{CO_{2\,(s)}}; eqn\ (4)$$

$$\frac{dF_{F_{CaCO_{3(s)}}}}{dW} = -k * \frac{F_{F_{CaCO_{3(s)}}}}{F_{CaCO_{3(s)}} + F_{CaO_{(s)}} + F_{CO_{2\,(s)}}} * P$$

$$collect\ terms: \left(1 + \frac{F_{CaO_{(s)}}}{F_{CaCO_{3(s)}}} + \frac{F_{CO_2}}{F_{CaCO_{3(s)}}} + \right) dF_{CaCO_{3(s)}} = -k * P * dW$$

Flow rates in terms of CaO_s yields:

$$3dF_{CaCO_{3(s)}} = -k * P * dW$$

Integrating from inlet to outlet:

$$3\int_{F_{CaCO_{3(s)}}(@inlet)}^{F_{CaCO_{3(s)}}(@outlet)} dF_{CaCO_{3(s)}} = -k * P * \int_0^W dW$$

$$3\left[F_{CaCO_{3(s)}}(@outlet) - F_{CaCO_{3(s)}}(@inlet)\right] = -k * P * W$$

$$W = -\frac{3 * \left[F_{CaCO_{3(s)}}(@outlet) - F_{CaCO_{3(s)}}(@inlet)\right]}{k * P}$$

$$V_{CaCO_{3(s)}} = -\frac{3 * \left[F_{CaCO_{3(s)}}(@outlet) - F_{CaCO_{3(s)}}(@inlet)\right]}{k * P * \rho_{CaCO_{3(s)}}}$$

Repeat for each component:

$$V_{CaO_{(s)}} = -\frac{3 * \left[F_{CaO_{(s)}}(@outlet) - F_{CaO_{(s)}}(@inlet)\right]}{k * P * \rho_{CaO_{(s)}}}$$

$$V_{CO_{2\,(g)}} = -\frac{3 * \left[F_{CO_{2\,(g)}}(@outlet) - F_{CO_{2\,(g)}}(@inlet)\right]}{k * P * \rho_{CO_{2\,(g)}}}$$

Using ideal gas law to approximate the density of carbon dioxide yields:

$$V_{CO_{2\,(g)}} = -\frac{3 * \left[F_{CO_{2\,(g)}}(@outlet) - F_{CO_{2\,(g)}}(@inlet)\right]}{k * P * \left(\frac{P * molar\ mass}{8.314 * T}\right)}$$

A better approximation of carbon dioxide density is the (Basu, Achayra and Dutta 2011) EOS.

$E_A = 180.56 \left[\frac{kJ}{mol}\right]; A = 2.12 * 10^6 \left[\frac{1}{s}\right]$ (Basu, Achayra en Dutta 2011)

Total volume of reactor expressed as:

$$V_T = -\frac{3}{A * e^{\frac{-E_A}{R*T}} * P} * \left(\frac{F_{CaO_{(s)}}(@outlet) - F_{CaO_{(s)}}(@inlet)}{\rho_{CaO_{(s)}}} + \frac{F_{CaCO_{3(s)}}(@outlet) - F_{CaCO_{3(s)}}(@inlet)}{\rho_{CaCO_{3(s)}}} + \frac{F_{CO_{2\,(g)}}(@outlet) - F_{CO_{2\,(g)}}(@inlet)}{\left(\frac{P * molar\ mass}{8.314 * T}\right)} \right) * 3600$$

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