Energieforschungsprogramm

Publizierbarer Endbericht

Programmsteuerung:
Klima- und Energiefonds

Programmabwicklung:
Österreichische Forschungsförderungsgesellschaft mbH (FFG)

Endbericht
erstellt am
05/2/2020

Projekttitel:

Energy efficient CO_2 capture and carbon neutral CO_2 supply chain for greenhouse fertilization at Wien Simmering (ViennaGreenCO_2)

FFG Projektnummer: 845022

Ausschreibung	4. Ausschreibung e!Mission.at
Projektstart	01/01/2015
Projektende	31/12/2019
Gesamtprojektdauer (in Monaten)	42 Monate
ProjektnehmerIn (Institution)	Technische Universität Wien
AnsprechpartnerIn	Prof. Hermann Hofbauer
Postadresse	Getreidemarkt 9/166, 1060 Wien
Telefon	+43-1-58801-166300
Fax	+43-1-58801-16699
E-mail	hermann.hofbauer@tuwien.ac.at
Website	http://www.vt.tuwien.ac.at

ViennaGreenCO_2

Energy efficient CO_2 capture and carbon neutral CO_2 supply chain
for greenhouse fertilization at Wien Simmering

AutorInnen:
Johannes Fuchs (TU Wien)
Florian Zerobin (BOKU)
Melina Infantino (Shell Global Solutions International B.V.)
Otmar Bertsch (Bertsch)
Ernst Höckner (Wien Energie)
Markus Saxinger (M-TEC)
Egon Zehetner (TU Wien)
Florian Dietrich (TU Wien)
Julius Pirklbauer (TU Wien)
Stefan Penthor (TU Wien)
Andreas Klingler (BOKU)
Magdalena Wolf (BOKU)
Gerhard Hofer (BOKU)
Gerhard Schöny (TU Wien)
Tobias Pröll (BOKU)
Sander van Paasen (Shell Global Solutions International B.V.)
Hermann Hofbauer (TU Wien)

1 Inhaltsverzeichnis

1 Inhaltsverzeichnis 4
2 Introduction 5
2.1 Problem definition 5
2.1.1 Short description of the TSA process 5
2.1.2 Problem definition 6
2.2 Relevance of the project to the call 6
2.3 Project consortium 7
3 Project summary („Inhaltliche Darstellung“) 9
3.1 Key aspects 9
3.2 Methods 9
4 Results and conclusions 11
4.1 Preliminary studies 11
4.1.1 BSU Tests 11
4.1.2 Cold flow model (CFM) tests 14
4.1.3 Heat transfer tests 16
4.1.4 Heat pump tests 17
4.1.5 Process modelling 18
4.2 Pilot unit campaign 19
4.2.1 Description of the pilot plant 19
4.2.2 Design and manufacturing 23
4.2.3 Summary of the pilot unit experimental campaign 25
4.3 Greenhouse fertilization study 29
4.4 Technoeconomic assessment 29
5 Outlook and recommendations 31
6 Literature 31
7 Contact 32

2 Introduction

2.1 Problem definition

2.1.1 Short description of the TSA process

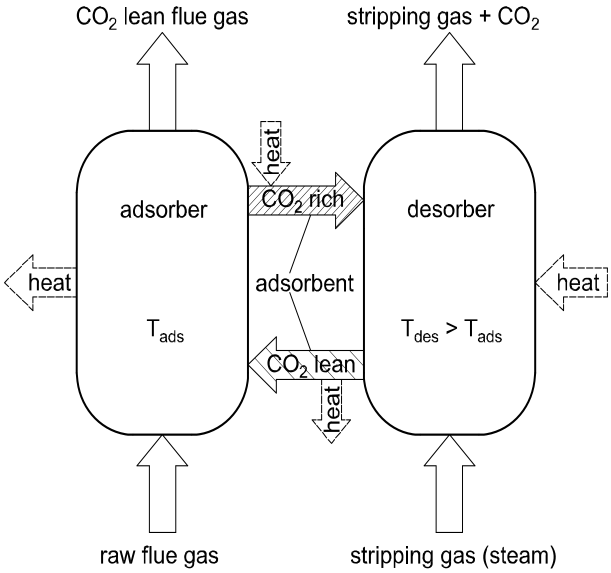

Figure 1: Simplified process setup

An innovative process, that is able to separate CO_2 from flue gas streams in a continuous temperature swing adsorption (TSA) process, was developed at TU Wien in collaboration with Shell. It was first described by Pröll et al. (Pröll et al., 2016), a bench scale unit was designed and manufactured (Schöny et al., 2016, Schöny et al., 2017) and first experiments were conducted. A simplified scheme of the process is shown in Figure 1. At low temperature (50°C) CO_2 is first adsorbed in one vessel (adsorber) where heat is released due to the exothermicity of the adsorption reaction. Subsequently, the adsorbent is transported to a second vessel (desorber), where the previously bound CO_2 is released at elevated temperature (105-120°C). In this way, the adsorbent is regenerated and can be returned to the first vessel (adsorber) where it is again available for CO_2 capture. In this way, the process can be operated in a continuous, closed loop. Heat must be withdrawn continuously from the adsorber in order to maintain the desired operating temperature. The inverse reaction (sorbent regeneration) takes place in the desorber and heat has to be provided continuously to maintain high operating temperatures throughout the entire vessel. A solid sorbent material is used, typically from the group of amine functionalized materials, where CO_2 is selectively bound (adsorbed) to active amine compounds on the surface of the sorbent. The lower the temperature is and the higher the amount of CO_2 in the flue gas, the more CO_2 can be adsorbed on the surface of the material. In the TSA process, this property is exploited. The CO_2 adsorption capacity of the adsorbent depends not only on the temperature but also on the CO_2 content in the gas phase. In the desorber, therefore, steam is used as purge gas in order to reduce the CO_2 content in the gas and thus to efficiently regenerate the adsorbent. In order to enable efficient CO_2 separation and heat integration as well as easy transport of the solid adsorbent within and between adsorber and desorber, the process is carried out as a fluidized bed system.

2.1.2 Problem definition

The goal of the ViennaGreenCO_2 project was to further develop the temperature swing adsorption (TSA) technology for continuous CO_2 capture, which had previously been studied at bench scale, and to demonstrate the process at pilot scale at the power plant site of Wien Energie in Simmering (Vienna, Austria). The goal was to develop a TSA pilot unit that is able to separate CO_2 from a partial flow extracted from the flue gas stream of the biomass CHP in Simmering and to utilize a part of the separated CO_2 as fertilizer in a test greenhouse operated by LGV Frischgemüse and lk-projekt. Results obtained from the project, especially from long-term operation of the TSA pilot unit, was incorporated in a concluding techno-economic assessment of the TSA CO_2 capture process itself, and in a study for a possible utilization of the CO_2 supply for greenhouse cultivation in the vicinity of the pilot plant in Wien, Simmering.

2.2 Relevance of the project to the call

By demonstrating an example for a whole CO_2 utilization chain, the ViennaGreenCO_2 project directly contributed to several areas of the topic (Topic 2 – Energy efficiency and energy savings) and the sub-topic (Topic 2.1 Energy efficiency in industry and trade) of the fourth call of the e!mission funding program, coordinated by the Austrian climate and energy fund (KLIEN). By investigating the innovative solid sorbent-based CO_2 capture technology at pilot scale, an energy efficient and economic industrial process was studied. If CO_2 capture from exhaust gas comes with higher efficiency and at lower cost, economics of carbon capture and storage solutions can be improved, resulting in more units and CO_2 emission avoidance. If biofuels are used, CCS has the potential to eliminate CO_2 from the carbon cycle (bio-CCS) and thus represents a promising and long-term climate repair technology. Another interesting application in the field of sustainable biofuels is efficient biogas upgrading to gas grid quality through separation of CO_2 from the residual stream of combustibles. On the other hand, CO_2 may be made available as high quality commodity using the innovative solid sorbent-based CO_2 capture technology. This route was investigated within the ViennaGreenCO_2 project for the case of greenhouse gas fertilization using highly concentrated CO_2. The technology may allow for decentralized supply with CO_2 along with reduced effort and emissions from CO_2 transport. It was the goal of the ViennaGreenCO2 initiative to investigate a highly efficient post combustion CO_2 capture technology at pilot scale and to investigate the potential of a utilization scenario for renewable CO_2 in a production process. The ViennaGreenCO_2 project therefore contributed to all three goals of the call:

- Development of the efficient CO_2 capture technology increases the energy efficiency of a post combustion carbon capture compared to the state-of-the-art and contributes to goals according to European and Austrian energy, climate and technology policies (“Energy Research Strategy for Austria”1, SET-Plan2) representing Goal 1 of the call. Furthermore, by realizing a local CO_2 utilization scenario, both, the carbon footprint and the energy demand of the CO_2 supply for vegetable production can be reduced, increasing the energy efficiency of the production process.

- The projected efficiency increase for the carbon capture process leads to potential cost reductions, compared to state of the art technologies on the market. In combination with the local scenario for CO_2 utilization resulting in reduced transportation costs, using renewable CO_2 for enhanced vegetable production will be more affordable. These two points represent Goal 2 of the call enhancing the affordability of sustainable energy and innovative energy technology.
- Since the project is built around investigating the innovative, highly efficient solid sorbent post combustion CO_2 capture technology with a potentially large international market, it is guaranteed that involved project partners will either establish or ensure technology leadership in this field or enhance their international competiveness (Goal 3). Participating universities (TUV and BOKU) can enhance their scientific excellence in the field of carbon capture technologies, gas separation and heat integration. Technology providers will be able to enhance or establish technology leadership in the fields of post combustion CO_2 capture (Bertsch) and heat recovery solutions (M-TEC) resulting in increased competiveness in international markets. The results of the project will also strengthen the competiveness of the participating end-users of the technologies by having an energy efficient CO_2 capture technology at hand creating a new business segment (WE), by opening alternative sources for renewable CO_2 and by increasing efficiency and thus competiveness of local vegetable production (LGV).

2.3 Project consortium

All project partners collaborated closely to reach the goals defined for the ViennaGreenCO_2 project. The project consortium consisted of universities, technology providers and end-users that show complementary experience, expertise and infrastructure and were thus capable to cover the complete project plan. The participating universities **TU Wien** and **BOKU**, which are widely renowned for their research activities in the fields of fluidization technology and carbon capture process development, represented the scientific partners in the project consortium, with TU Wien as the project coordinating entity. The pilot plant was developed in close cooperation with and manufactured and erected by the Austrian manufacturing company Josef Bertsch GesmbH & Co KG (**Bertsch**), who are experienced in development of commercial fluidized-bed boilers, heat exchanger solutions as well as in the delivery of customized special designs. Industry partner Shell Global Solutions International B.V. (**Shell**) was closely involved in all workstreams, especially with project management, engineering and with all efforts regarding health, safety, security, environment and social performance (HSSE&SP). The partner provided valuable guidance and experience with industry standards, the operation of chemical processes and plants at laboratory scale, pilot scale and industrial scale, process evaluation and gas treatment. Furthermore, the partner supported the project consortium financially, and brought in further competences in the field of catalyst, respectively, solid sorbent material development and the required sorbent material was supplied by them. The Austrian heat pump supplier **MTEC**, who represents a highly innovative company that supplies and develops heat pump systems for private as well as industrial customers, delivered the high temperature heat pump which was used in an experimental campaign to evaluate the potential for advanced heat integration potential via high temperature heat pumps in the TSA process. The partners lk-projekt niederösterreich | wien GmbH (**lk-projekt**) and LGV-Frischgemüse Wien reg. Ges.m.b.H. (**LGV**), who have great experience in utilisation of CO_2 for fertilization purposes in greenhouse cultivation, were closely involved in the assessment of the potential for regional utilization of renewably produced CO_2. The public utility company Wien Energie GmbH (**Wien Energie**) provided significant experience regarding plant integration, erection and operation to the consortium and provided the necessary flue gas, extracted from the biomass CHP owned by Wien Energie. All partners proved their capability for reliable collaboration, excellent teamwork and high-level research activities.

3 Project summary („Inhaltliche Darstellung“)

3.1 Key aspects

In the first project phase (“Up-scaling of TSA technology”), a “up-scale-ready” design proposal of the TSA reactor system was developed. In the second project phase, “Operation of TSA pilot unit together with complete CCU chain and performance evaluation”, the developed design solutions and conclusions from the first project phase were used to generate the detail design of the TSA pilot unit and of all peripheral equipment. Then, the facilities were constructed and finally delivered to and commissioned at the power plant site in Simmering. The final phase of the project was dedicated to the operation of the TSA pilot unit and, after the end of the experimental campaign, clear techno-economic figures for the developed CO_2 capture technology as well as for a regional and sustainable CO_2 supply chain in Simmering were produced.

Some major project goals were:

- Determine the relevant technical principles for up-scaling of the double-loop solid sorbent CO_2 capture process from bench-scale to pilot scale by targeted experiments on the TSA bench scale unit and on specific experimental configurations for indirect heat transfer to fluidized beds.
- Propose a scale-up-ready pilot plant design for the double-loop solid sorbent CO_2 capture process and investigate fluid dynamics of the proposed design in a cold flow model device.
- Investigate the possibilities for energy-efficiency increase of the process by heat integration including an active adsorber heat recovery using an environmentally sustainable heat pump system for desorber heating.
- Perform basic and detailed engineering of the pilot plant and of the CO_2 supply infrastructure.
- Conduct pilot tests at the site of Wien Energie Simmering using real combustion exhaust gas featuring process monitoring and control systems.
- Operate the pilot plant for at least 500 hours within the project.
- Evaluate the results from pilot plant testing on a techno-economic basis and in view of a possible follow-up demonstration project.

3.2 Methods

With the knowledge gained during previous research projects and dedicated preliminary studies described above, a pilot unit was designed, manufactured and delivered to the host site at Wien Energie in Simmering, Vienna, Austria. Throughout the development of the TSA process, it was necessary to gain a deeper understanding of the process and certain aspects were studied in dedicated workflows:

- Dedicated tests with a **bench-scale unit** were conduct to study the performance of different sorbent materials and the emissions to expect in the adsorber and desorber exhaust gas.
- Regarding the fluidized bed system, it was necessary to take the step from a bench-scale unit towards a pilot unit, and a robust, flexible and efficient design was required. A **cold flow model** was designed and manufactured, and experiments were conducted to study solids transport within the adsorber and desorber column (downcomers), gas sealing between the columns (standpipes), heat exchanger surface areas (bed expansion and bed density), gas distributor design, pressure drop and many other aspects of the fluidized bed system.
- To help with the design of the heat exchanger surfaces, which are necessary to provide for the continuous cooling and heating demand of the process, **a heat exchanger test rig** was designed and manufactured, and an experimental campaign was conducted.
- To evaluate the potential for advanced heat integration within the TSA process via high temperature heat pumps, experiments were conducted with a **high temperature heat pump prototype**, together with at a **heat pump testing station**.
- Mass- and energy balances were provided throughout the design process with a **process modelling** tool, which was continuously adapted and improved throughout the project. This modelling tool had already been in use to elaborate the basic thermodynamics of the process, for the design of the bench scale unit and it was now used to provide for the mass and energy balance for gross design and dimensioning of the piping and instrumentation for the pilot unit.

4 Results and conclusions

4.1 Preliminary studies

4.1.1 BSU Tests

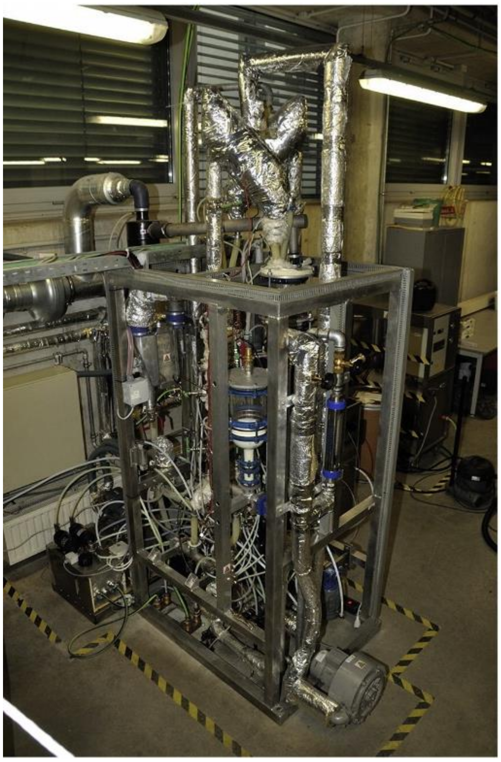

Figure 2: The TSA bench scale unit for continuous CO_2 capture from gas streams (Schöny et al., 2017)

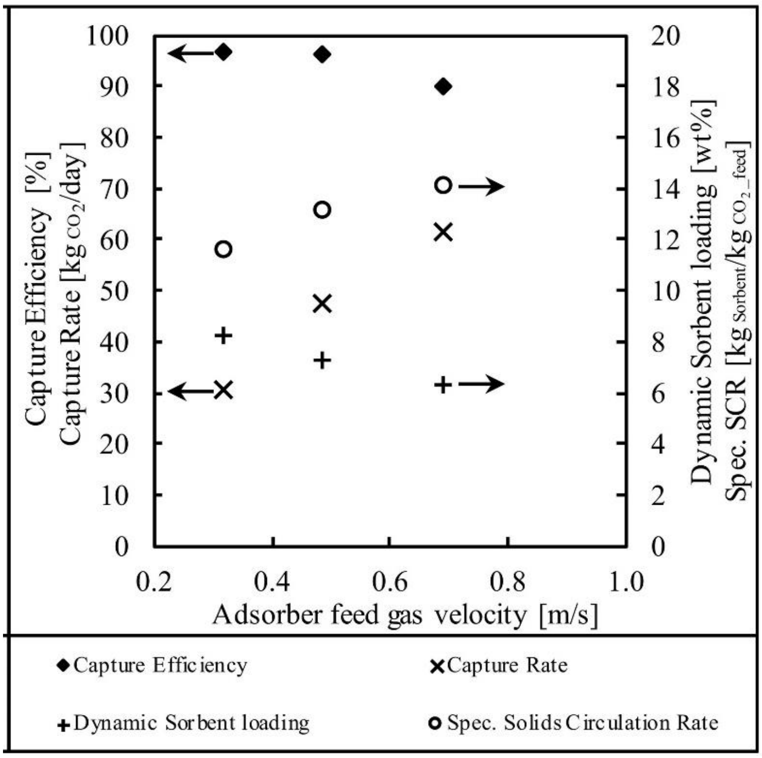

Figure 3: Influence of the adsorber feed gas flow on the performance of the TSA process, measured in a bench-scale unit (Dietrich et al., 2018)

The TSA bench scale unit (BSU) displayed in Figure 2, which had already been used for previous projects, was used during the course of the ViennaGreenCO_2 research project. Experiments with the BSU helped to gain valuable understanding of the process at small scale under laboratory conditions (synthetic flue gas), as well as in field tests with live-gas from a biomass gasification CHP station in Güssing, Austria. The BSU setup consists of two multi- tage fluidized bed (FB) columns operated in the bubbling fluidized bed regime and two sorbent transport loops.

Process parameter variations: One of the important requirements for the design TSA pilot plant, was flexibility and compactness. This means that the fluidized bed system should be able to treat large amounts of flue-gas, which in turn means that the operating flue-gas velocity in the adsorber column of the CO_2 capture plant needs to be as high as possible in order to minimize the plant footprint. Moreover, a load change of the CO2 emitting process can manifest in a change of the flue gas flow rate, which means that it should be able to handle a wide range of flow rates (high turndown ratio). However, in stationary fluidized beds, high superficial gas velocities in the fluidized beds lead to both the formation of large bubbles and reduce gas solids contact time. Hence, it is important to identify limitations of the TSA process when

operated at high flue gas velocities in the adsorber. Variations of the flue gas velocity in the adsorber were conducted to reveal the flexibility of the TSA process in this regard (see Figure 3). The defined target during these variations was to increase the superficial gas velocity in the adsorber whilst increasing the SCR proportionally so that a constant specific SCR is achieved. By operating the BSU in this way, limitations regarding mass transfer and reaction kinetics become particularly pronounced and the influence of the fluidization velocity on the process performance was studied. The observations of this study were helpful in the design of the adsorber column, especially with regards to the bubble breaking, column cross section, downcomers and gas distributors.
In the course of two different series of parameter variations, the impact of varying operating temperatures in the adsorber and varying CO_2 concentrations in the treated flue gas on the CO_2 capture performance of the TSA BSU were investigated. Variations of the adsorber temperature have a direct influence on the cooling duty and the maximum working capacity of the sorbent. Variations of the inlet CO_2 concentration were conducted to show the applicability of the process for flue gas from different sources, where the CO_2 concentration in the available flue gas may differ significantly from each other (e.g. NGCC, biomass fired power stations, coal-fired power stations).

Sorbent degradation: A loss or permanent deactivation of active amine groups on the sorbent particles may occur during operation and lead to a reduced capture performance of a TSA plant. In case the sorbent material tends to degrade or get deactivated over time, a fraction of the total sorbent inventory of a TSA plant may have to be continuously replaced by fresh sorbent at commercial scale to maintain a constant CO_2 capture performance during operation. This means that the mechanisms of sorbent degradation have to be understood because they will directly affect the cost of CO_2 capture. Trace gases that are specific to combustion processes i.e. SO_2, NO and NO_2 are of special interest for developing a post combustion CO_2 capture process, since they can induce degradation of amine functionalized sorbents, and thus reduce the CO_2 capture performance of a CO_2 capture process over time. The BSU was transported to a combined heat and power (CHP) plant in Güssing/Austria for field experiments, where live flue gas was treated in a series of experiments to identify and quantify the expectable sorbent degradation under process conditions which are as close to a commercial application as possible.

Emissions: To conform to environmental regulations, amine emissions in the CO2 depleted flue-gas must be minimal. Typically, in aqueous amine based systems, emissions are reduced by means of a water-wash section down-stream of the absorber, whereas the resulting effluent is returned to the process. However, considerable amine emissions in form of aerosols or NH_3 emissions can persist despite a water wash section. In MEA-based scrubbers such effects may make alternative emission reduction measures, such as Brownian demister units or acid wash systems necessary. Emission reduction measures such as the ones mentioned above, not only increase investment cost but can also increase the operating cost of Post-Combustion Capture processes. This is because end-of-pipe emission reduction measures typically add a pressure drop to the flue gas path and thus lead to an increased power requirement of the flue-gas blower. It is therefore key during the development of any PCCC system to identify emissions and assess whether auxiliary systems are required for the removal of critical components. For this reason, two 120-hour experiments were carried out in the TSA BSU. During these experiments, comprehensive sampling of various gas and liquid streams took place to determine the emissions of the BSU throughout the experimental runtime. The goal was to gather experience in long-term experiments with a selection of two different sorbent materials, and the results made it possible to clearly identify a sorbent of choice for reasons of easy handling, emissions and sorbent long-term-stability.

Water transport: The combustion of most fuels leads to the formation of a flue gas that contains considerable amounts of water vapour. As shown by Figure 24 in Section 2.4.1, the water adsorption capacity of Sorbent B increases with relative humidity and can be many times higher than the adsorption capacity of CO2. This may lead to a significant amount of water that is transported between the adsorber and desorber side of the process, which may lead to significant changes in the heat duty of the process, depending on the operating conditions. Although a cooler will be required upstream of the adsorber to cool the treated flue gas to adsorber operating temperature, a further reduction of the flue gas dew point represents an added cooling demand and potentially also higher CO2 capture costs. To keep the cooling requirement of the flue gas low, the humidity of the treated gas will be near saturation when entering the adsorber. The low heat demand of the TSA process is an anticipated advantage over conventional aqueous liquid amine scrubbing. Since water adsorption can influence the process heat demand, it is key to gain an understanding of the interaction between the utilized sorbent material and water vapor throughout the TSA process. An investigation was carried out in the TSA BSU to evaluate the effect of water co-adsorption at various humidity levels of the desorber feed gas. The results helped with the understanding of water transport in the TSA system, which is relevant for the design of the desorber unit, heat exchangers immersed in the fluidized bed system and the OPEX of such TSA plants.

Lean/rich heat exchange: Heat integration between the lean and rich sorbent material is crucial to energy efficiency of a TSA process, and it is necessary to find a suitable design for the scaleup of the TSA process. For liquid amine scrubber systems, highly efficient lean/rich heat exchange can be implemented via counter-current via shell-and-tube, plate or plate-and-frame heat exchangers because of the favourable hydrodynamic properties of the liquid solvents. In systems where heat is exchanged with a solid sorbent, a fluid must act as an intermediate heat carrier medium for heat exchange. A suitable design for liquid/solid heat exchangers, which promise optimized size and manufacturing cost, are moving-bed heat exchangers. Two different types of moving bed heat exchangers were designed, manufactured and tested in dedicated experiments to gather experience with this innovative type of heat exchanger, and to understand what the expectable performance is (lean/rich heat recovery ratio).

4.1.2 Cold flow model (CFM) tests

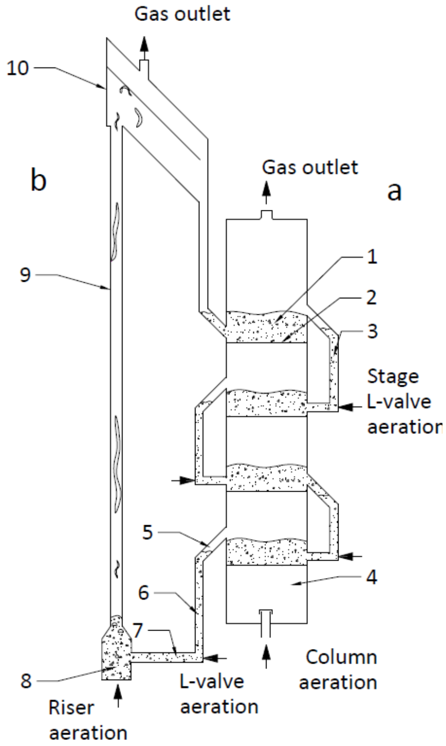

Figure 4: CFM setup used for external downcomer tests

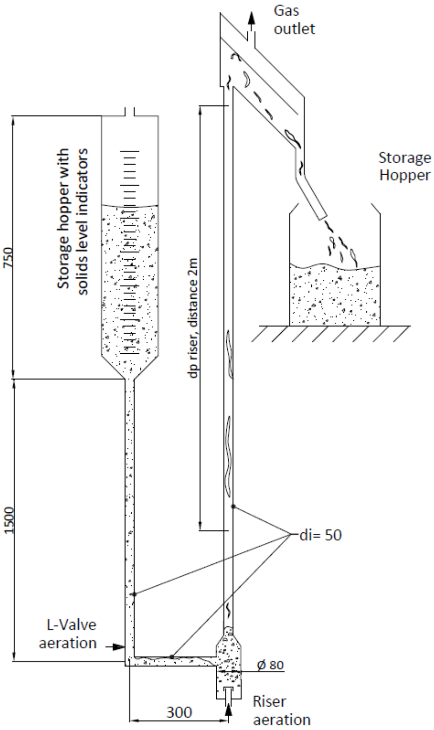

Figure 5: CFM setup used for riser tests

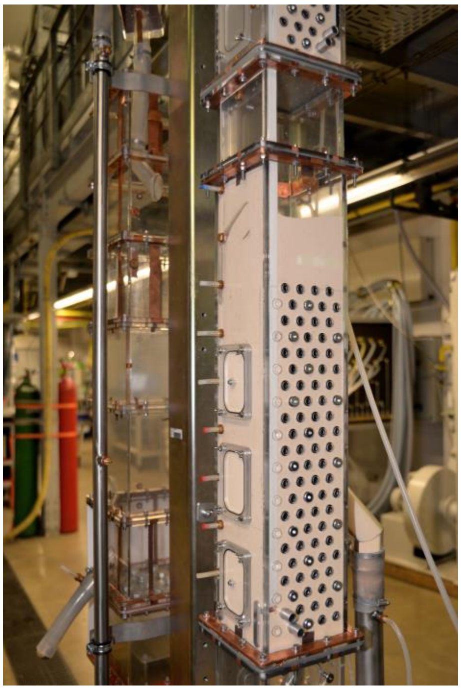

Figure 6: CFM setup for desorber tests with internal downcomers

Bed expansion and bed density: Due to the vast amounts of flue gas that have to be treated in post combustion CO_2 capture units, the pressure drop of the adsorber column is limited to 50 mbar in order to reduce the energy demand of the flue gas blower. While the average pressure drop per stage can be determined, the calculation of the available bed height and, therefore, the available space for internals is challenging. Thus, an experimental cold flow model campaign was conducted to investigate the development of the bed height for different gas velocities and different heat exchanger geometries.

Solids transport between the columns: The solid circulation rate within the TSA pilot unit is measured from the pressure drop measured in the lean transport riser. Furthermore, the solids pressure drop in the transport sections of the risers determines the pressure drop that has to be balanced by the standpipes below the fluidized bed columns. The transport riser were replicated and tested under ambient conditions. The results of this campaign allowed an estimation of the solids pressure drop within the transport risers in the pilot unit.

Downcomer design: The fluid dynamic operating behaviour of a multistage fluidized bed column was experimentally studied with the aid of the cold flow model. The focus of this work was to study the operational boundaries for different downcomer geometries with the aim to maximise the achievable solids flux in the adsorber. The maximum solids circulation rates were measured for several superficial gas velocities and the operating regimes resulting within the downcomers were monitored. External and internal downcomers were tested successfully within the adsorber cold flow model. It was shown, that both types are capable of similar solids fluxes. Similar to the experimental campaign for the adsorber column, the downcomer system for the desorber column was investigated and different designs of external and internal downcomers were tested. This was necessary due to the very low fluidization numbers required within the pilot desorber column in order to minimize the consumption of stripping steam. Experience gained from the adsorber downcomer tests, like the importance of the inlet height and effects of the gas distributor design were implemented in these investigations. Internal and external downcomer designs were tested within the desorber cold flow model. Both designs were capable of supporting the required solids circulation rate.

Gas distributors: For fluidized beds, the pressure drop across the gas distributor is usually recommended to be in the range of 10 % - 40 % of the bed pressure drop in order to ensure even gas distribution across the complete gas distributor area. However, high gas distributor pressure drops are critical when it comes to operating costs. This is especially true for a multistage unit, since the pressure drop of each gas distributor adds to the blower power demand. Hence, a CFM study was conducted, with the aim to test different gas distributor designs and to optimize the adsorber columns overall pressure drop

Gas leakage: One of the main challenges for the TSA pilot unit is to provide the CO_2 at the exit of the desorber with high purity, whereby especially the oxygen content due to gas leakage from the adsorber column should be in the ppm range. To investigate if these requirements can be met, a cold flow model study was conducted. It comprises a stationary fluidized bed and a solids recirculation system. The recirculation system itself consists of an L-Valve that extracts bed material from the bottom of the fluidized bed at a controlled rate and a pneumatic transport riser that redirects the material back onto the top of the fluidized bed. The aim of this work was to investigate the L-Valve transport behaviour and operating range under different pressure conditions. Furthermore, the leakage of fluidization gas from the main fluidized bed into the recirculation system was measured by using propane as tracer gas.

4.1.3 Heat transfer tests

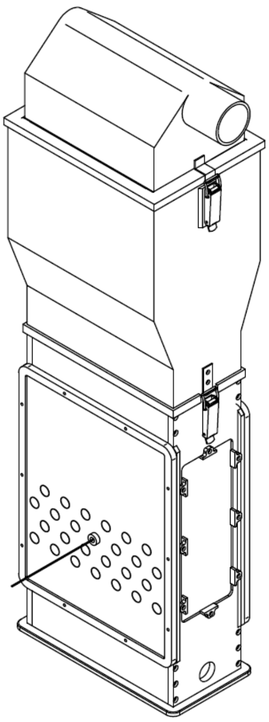

Figure 7: 3D-View of the heat exchanger test rig at BOKU (Hofer et al., 2017)

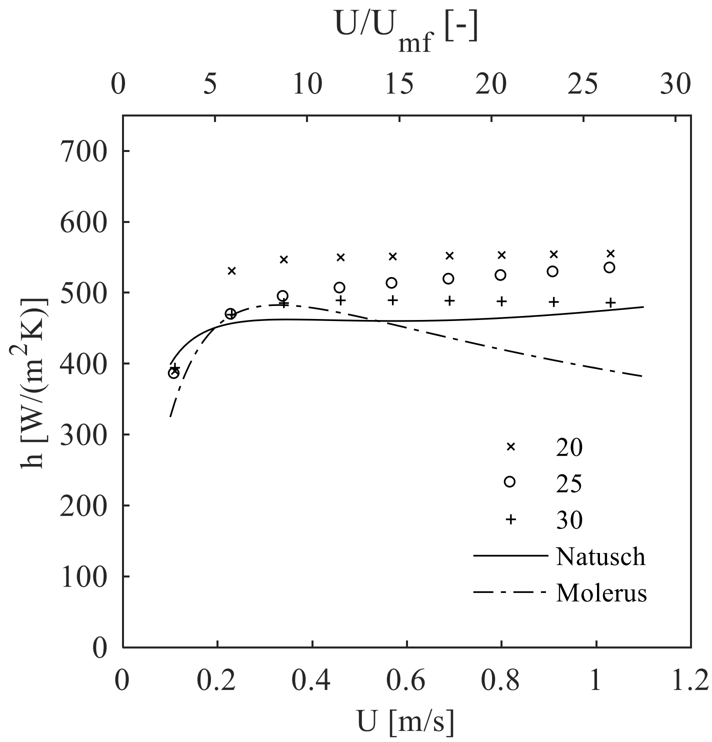

Figure 8: Single tube heat transfer, compared to selected models from literature for 200µm glass beads (Hofer et al., 2017)

For proper operation of the TSA pilot unit, it is essential to provide sufficiently large heat transfer surface area within the individual stages of both columns as well as within the pre-desorber. Dimensioning of the corresponding heat exchangers requires the availability of valid values for the prevailing heat transfer coefficients between the fluidized adsorbent material and immersed heat transfer surfaces. Since literature data and modelling tools for the determination of heat transfer coefficients of densely packed heat exchanger bundles immersed in stationary fluidized beds are lacking, it was decided to assess the prevailing heat transfer coefficients experimentally. For this purpose a heat transfer measurement test rig (HTMT) was designed, constructed and put into operation. The HTMT was then equipped with tube bundles of the same geometry as those that are foreseen for the adsorber and desorber/pre-desorber, respectively. It was shown that the achievable heat transfer coefficients are in the range of about 40 to 220W/(m²K). The results clearly indicate that heat transfer strongly depends on the tube location within the fluidized bed. While relatively low heat transfer rates were measured close to the wall of the HTMT, the heat transfer tends to increase when moving towards the center of the fluidized bed. Results were presented at a scientific conference (Hofer et al., 2017), and published in scientific journals (Hofer et al., 2018) and in a PHD Thesis (Hofer, 2019).

4.1.4 Heat pump tests

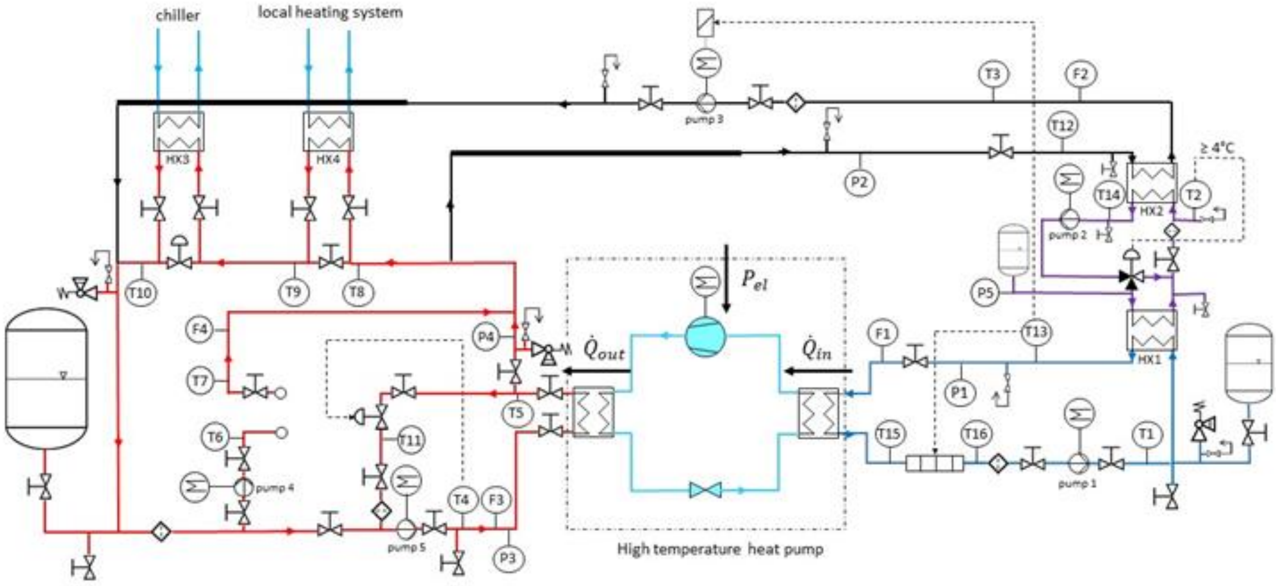

Figure 9: Instrumentation scheme of the heat pump test rig at BOKU

To study the possibilities for heat integration between the adsorbed (heat source) and the desorber (heat sink), the decision was taken to design and erect a heat pump test rig where the performance of innovative high temperature heat pumps can be studied. A high temperature heat pump prototype was designed together with and manufactured by the Austrian project partner company MTEC and delivered to BOKU for testing. The tested heat pump prototype was based on the M-TEC WPS618 heat pump. The utilized compressor was a semi-hermetic reciprocating compressor with cylinder head cooling from HKT Huber-Kälte-Technik GmbH. In experiments conducted with the heat pump testing rig at BOKU it could be shown that the constructed heat pump setup can provide the required design temperatures. Tests were conducted under stationary operating conditions, at pre-defined inlet-temperature levels, and the heat pump's coefficient of performance (COP) was measured experimentally. Furthermore, these results were compared to theoretical performance indicators obtained by simulation tools and conclusions were drawn for the performance of a high-temperature heat pump at large scale. The efficiency of a heat pump depends on the temperature levels of evaporation and condensation, and the temperature difference between the adsorber and desorber column proved to be a challenge for high temperature heat pumps. The measured COP with the used refrigerant R365mfc (1,1,1,3,3-Pentafluorobutane) was limited (<2,11) because of the large temperature span caused by between the adsorber and desorber column, where a temperature swing is essential for achieving a high process efficiency and efficient regeneration of the sorbent. Thermodynamic calculations showed that a COP of up to 3,17 is possible at larger scale with the used refrigerant. In order to end up with a performant heat pump process for the specific temperature level requirements of such a TSA system, the refrigerant R245fa is proposed which promises a higher volumetric heating capacity and higher evaporation pressure levels.

4.1.5 Process modelling

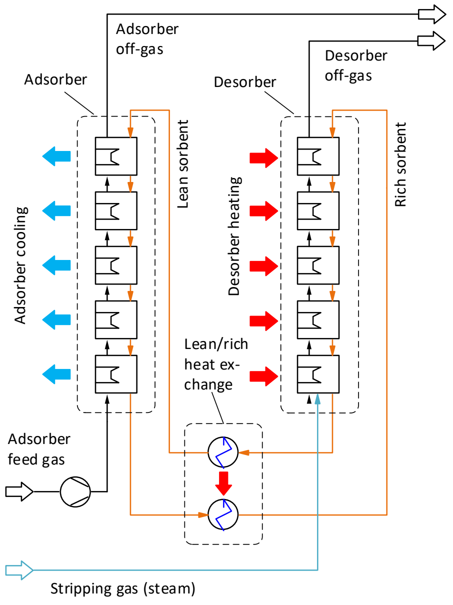

Figure 10: Simplified flowsheet of the thermodynamic process modelling tool, developed in IPSEpro

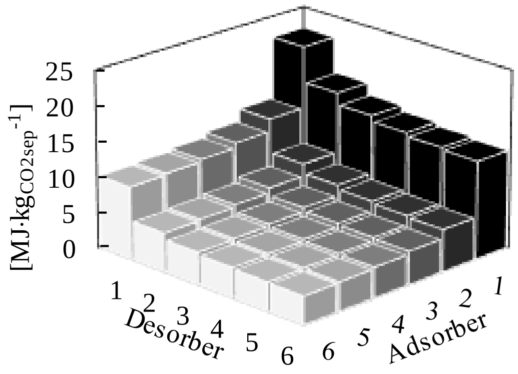

Figure 11: Investigations regarding the optimal stage configuration in the adsorber and desorber column (Pirklbauer et al., 2018)

A thermodynamic modelling tool was used throughout the course of the ViennaGreenCO_2 research project to calculate mass-and energy balances for the TSA process. The TSA process model was created in the commercial software IPSEpro, which is a flowsheet modelling tool, that can handle large and complex thermodynamic models and calculate solutions accurately and conveniently. It has been used at TU Wien and BOKU for the modelling of fluidized bed processes (CLC, CLR, gasification, combustion, TSA), steam cycles and gas turbine processes, as well as other fields of application (Pyrolysis, smart heat grids, heat pumps and refrigeration). The library used for modelling of TSA processes contains comprehensive databases to describe the thermodynamic states of different gaseous, liquid and solid species, and it provides the user with the necessary tools to describe gas-solids contact and adsorption/desorption mechanisms. All process units strictly fulfil mass and energy conservation requirements. In order to develop a functioning process modelling tool, it was necessary to develop an understanding about the properties of the sorbent material (heat capacity, equilibrium isotherms) and about the heat and mass flows occurring in a solid sorbent TSA unit. For material characterization, the uptake of CO_2 under dry and wet conditions, as well as the uptake of H_2O were studied in lab experiments by different project partners (TU Wien, BOKU, Shell). These results were used to develop a mathematical description of the adsorption equilibrium with suitable model function fits. The models were used during the design and dimensioning of the pilot unit, as well as for troubleshooting during the experimental campaign and for data validation, to verify whether the model is actually able to predict the behavior of a solid sorbent TSA separation process satisfactorily. Experimental results gathered with the bench scale unit (BSU) and the pilot unit were compared to values predicted by the modelling tool. **Fehler! Verweisquelle konnte nicht gefunden werden.** and **Fehler! Verweisquelle konnte nicht gefunden werden.** show the measured heat demand of the pilot unit, compared to the predictions of the model.

4.2 Pilot unit campaign

4.2.1 Description of the pilot plant

Figure 12 shows a simplified process scheme of the TSA pilot unit. Both, the adsorber and the desorber consist of a series of five bubbling fluidized beds. Within both columns, the net solid flow is downwards and gas is passing the contactor from the bottom to the top. The adsorber is fluidized with the flue gas to be treated in the TSA unit, whereas the desorber is fluidized with stripping steam. On the way through the adsorber column, the CO_2 concentration within the flue gas stream gradually decreases as CO_2 is being selectively adsorbed by the passing adsorbent material. Ideally, the cleaned flue gas leaves the adsorber with the desired CO_2 concentration at the top of the column while the CO_2 loaded, rich adsorbent material is extracted from the lowermost stage of the column. In the desorber, the rich adsorbent material gets regenerated, thereby, desorbing CO_2 again so that finally a mixture of CO_2 and steam is obtained in the offgas of the desorber. Each of the individual fluidized bed stages in both columns features a gas distributor, a downcomer that enables controlled particle flow downwards between connected stages as well as a heat exchanger for heat supply and cooling, respectively. The downcomers of the adsorber column are placed outside of the column and designed and operated as L-Valves. The level in the individual adsorber stages can be controlled with the feeding rate of aeration gas introduced into the standpipe of the external downcomers. The downcomers within the desorber column are placed inside the column (internal downcomers). Weir plates extend the downcomer area in each stage and define the solids level in the individual desorber stages.

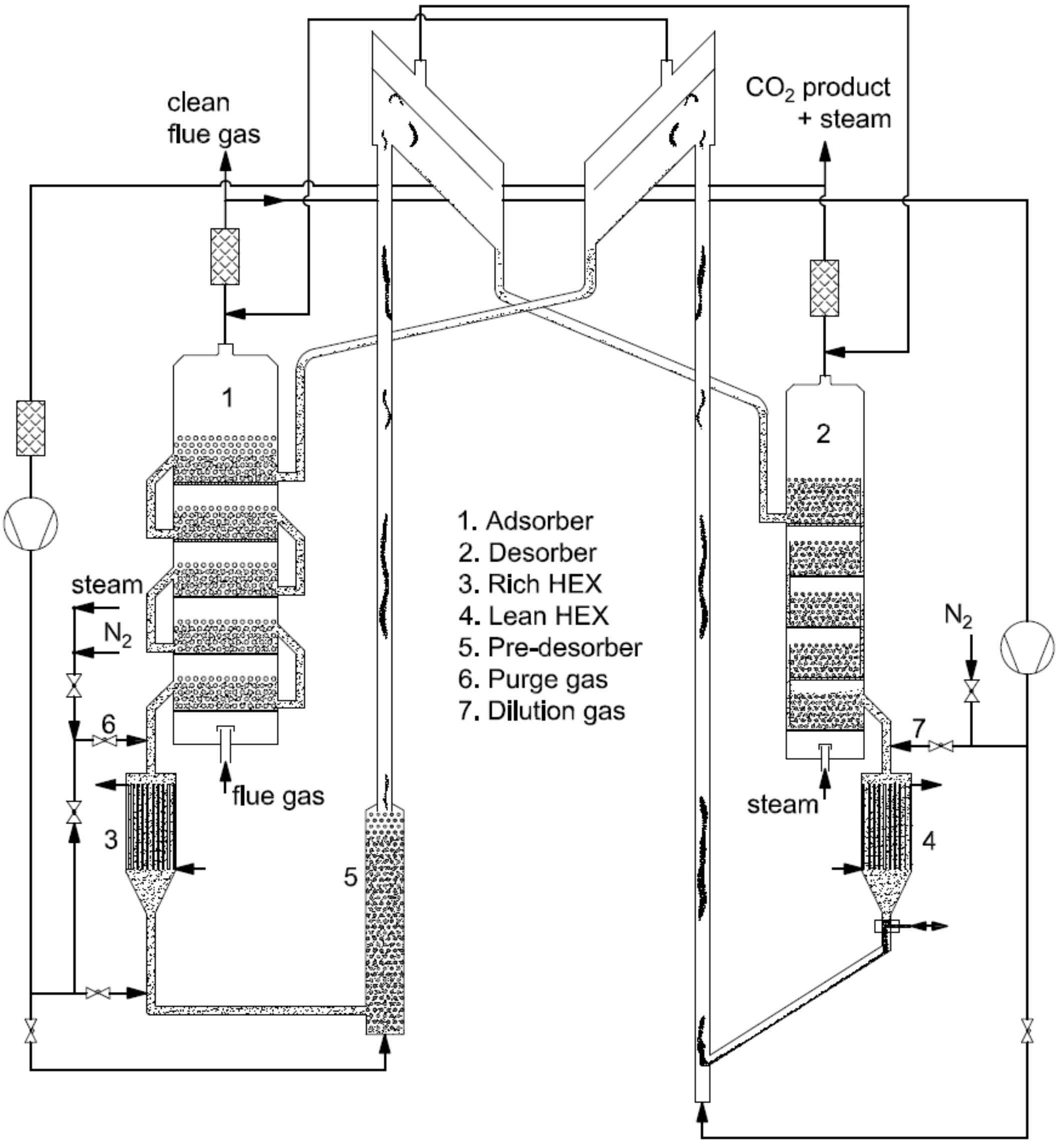

Figure 12: Simplified scheme of the design of the ViennaGreenCO2 pilot unit

Perforated plate type gas distributors are used in both columns to sufficiently distribute the gas flow across the cross section of the fluidized beds. In the desorber column, the gas distributor perforations below the internal downcomers are modified to allow for stable transport of adsorbent material from stage to stage. The perforation size in the gas distributors is big enough to allow the sorbent material to drain through them as soon as the column fluidization is switched off. However, in the lowermost stages of both columns, non-weeping gas distributors need to prevent solids from draining into the windbox. The top stages of both columns exhibit an extended freeboard that provides that the transport disengaging height for the utilized adsorbent material is exceeded under design operating conditions.

Table 1: Design basis for the ViennaGreenCO2 pilot unit

Load cases	LL	DL	HL	Unit
CO_2 capture target				

CO_2 capture capacity	29.6 (0.7)	41.7 (1.0)	59.5 (1.4)	kg/h (t/day)
Target CO_2 capture efficiency	90	90	90	%
CO_2 feed concentration	4.0	4.0	4.0	%-Vol.
TSA operation				
Adsorber operating temperature	50	50	50	°C
Desorber operating temperature	115	115	115	°C
Pre-Desorber operating temperature	115	115	115	°C

Sorbent material is continuously discharged from the lowermost stages of both contactors under controlled conditions. Below the adsorber column a L-Valve is used to solids flow control, whereas in the desorber, a mechanical slide valve is used. Before passing the solids flow control valves, the adsorbent material enters a counter-current moving bed heat exchanger that allows for pre-heating or pre-cooling of the circulating adsorbent material streams (i.e. simulated lean/rich heat exchangers). After the adsorbent material passed the L/R HEXs, it is further directed into transport risers that pneumatically lift the particles up to above the top stage of the respective other column. The riser that transports adsorbent material into the desorber column (adsorber riser) exhibits a heat exchanger in the bottom section. This heat exchanger heats up the rich adsorbent material to the desorber operating temperature. Consequently, part of the adsorbed CO_2 is already desorbed in this section, called pre-desorber. At the top of both risers, the lifted particles enter a gas-solid separating device. Simple gravity separators are applied here in combination with filter separators to remove any adsorbent dust that can be formed during operation of the pilot unit. The separated solids are directed into the top stage of the respective other column to complete the process cycle. The lift gas used in the adsorber riser (adsorbent transport from ADS->DES) is desorber off-gas that is continuously extracted downstream to the off-gas filter by means of a gas blower. During normal operation of the TSA pilot unit, part of the extracted gas flow is also used as aeration gas in the L-Valve below the adsorber (solids flow control) and as purge gas above the rich heat exchanger (gas sealing). After solids separation, the gas flow from the adsorber riser is fed back into the desorber off-gas, upstream to the desorber off-gas filter. In the desorber riser (DES->ADS), recycled clean flue gas is used as lifting gas. The opening of the mechanical slide valve downstream to the lean heat exchanger will be controlled to achieve the desired sorbent circulation rate in the system. The actual sorbent circulation during operation of the unit can be calculated from the pressure drop measured over the transport section of the desorber riser. The aeration rate of the L-Valve below the adsorber column is adjusted to obtain a constant solids inventory within the desorber column (i.e. a constant distribution of sorbent inventory between the adsorber and the desorber during operation). The overall pressure drop over the columns gives a good indication for the amount of sorbent material that is currently present in the column. Hence, the measured pressure drops over the columns will be used to control the aeration gas flow of the L-Valve downstream to the rich heat exchanger. For several reasons, it is important that the purity of the CO_2 product that is extracted from the desorber is high.

BERTSCH

BERTSCHenergy
Kraftwerksanlagen
Prozessapparate

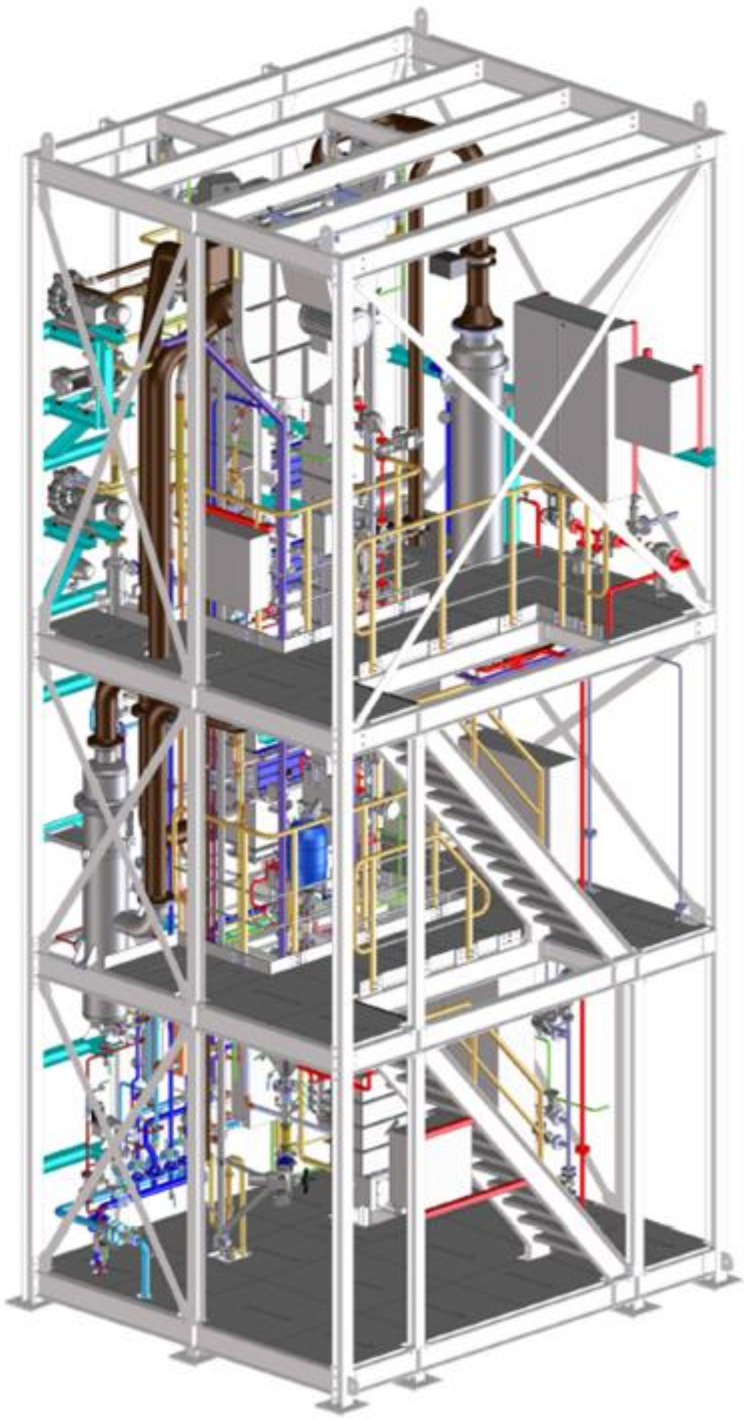

Figure 13: 3D-View of the ViennaGreenCO_2 pilot plant

In carbon capture and storage (CCS) applications, it is of special importance to limit O_2 or N_2 contaminations in order to reduce equipment corrosion, power consumption for CO_2 compression or storage space demands. It is also undesirable to have oxygen slip into the desorber, since at higher operating temperature oxygen could significantly increase the sorbent degradation rate. Consequently, the TSA process needs to provide a selective transport of CO_2 from the adsorber to the desorber whilst any slip of other gaseous species needs to be prevented by proper gas sealing measures. On the desorber side, no purge gas is used for gas sealing. However, a small gas stream is used to build up a pressure that is just enough to minimize steam slip from the desorber and to dilute this stream in order to prevent steam condensation within the lean heat exchanger.

4.2.2 Design and manufacturing

Highlights and milestones during the design and manufacturing of the ViennaGreenCO_2 pilot plant were:

Start of the ViennaGreenCO2 project:	Jan 1st 2015
Preliminary studies on hydrodynamics, heat exchanger, thermodynamics, process performance at bench scale, etc.	2015-2017
HAZOP study, together with TÜV	January 2017
Basic design and engineering package (BDEP) completed:	June 2017
Manufacturing at Bertsch factory:	Q4,2017-Q1,2018
Permit granted by authorities:	March 1st 2018
Plant delivery to Wien Energie:	April 2018
Commissioning:	May-July 2018
Pilot plant opening ceremony:	June 21st, 2018
Pre-Startup safety review:	July 2018
First hot operation:	Oct 2018
First testing campaign:	Q4 2018
Plant modifications:	Q1 2019
Pilot plant experimental campaign:	May-July 2019
Data post-production, technoeconomic assessment, feasibility study for regional utilization of renewable CO_2, project finalization and reporting:	Q3+Q4 2019

Figure 14: View of the steel structure of the TSA pilot unit container before installation of the façade (December 2017). Main pieces of equipment (adsorber and desorber column, rich and lean heat exchangers as well as flue-gas cooler and condenser) were already installed.

Figure 15: Delivery of the pre-manufactured pilot plant from the factory (Bludenz, Austria) to the host site at the Wien Energie (Simmering, Vienna, Austria) in April 2018.

After intensive preparations conducted by all project partners in the first two years of the project, the design basis (BDEP) for the TSA pilot unit was finished. This included the design of the process, equipment (columns, vessels, heat exchangers, pumps, blowers, etc.), the process control and safeguarding systems (control narrative, safeguarding, cause and effects matrix, tie-ins and interfaces, fire and gas protection systems, system architecture, power supply, etc.), civil and structural design (container design, HVAC, statics and foundations, structural steel, piping, drainage, access roads, site preparation, etc.), the electrical scope, material selection, health, safety, security, environment and social performance (HSSE & SP), piping and layout or the process and operating guidelines (startup and shutdown, operating procedures). Regarding HSSE & SP, a comprehensive approach was followed to ensure the safety of personnel and equipment throughout the entire pilot plant campaign. HAZOP and HAZID studies were carefully conducted and continuously implemented during the entire design and planning process of the pilot unit, the surrounding work environments and the process control system. Operating procedures and a detailed operating manual was written and the operators were given intense theoretical and practical training on safety-related issues and on pilot plant operation. The manufacturing at the Bertsch factory in Vorarlberg, Austria was commenced in the end of 2017 and finished in April 2018, when the pre-manufactured pilot plant was delivered in two containers to the power plant host site at Wien Energie in Vienna, Austria. After successful delivery to Vienna (Figure 15), connections at the tie-in points were established (flue-gas feed and flue gas return, steam, water) piping and electrical installations were finished and the plant was commissioned.

4.2.3 Summary of the pilot unit experimental campaign

After first tests between October and December 2018, modifications for improved stability and productivity of the process were successfully implemented, and finally, between March and July 2019, the pilot campaign was executed until the combined heat and power plant, which delivered raw flue gas for the pilot unit, was shut down permanently on August 1st 2019.

Table 2: Overview of the performance of the pilot unit during the experimental campaign

Parameter	Value	Unit
Time in hot operation	900+	hours
Time in cold operation	250+	hours
Temperature adsorber	40-60	°C
Temperature desorber	105-120	°C
Capture efficiency	variable	%
Capture rate	200-700	kg/day
Flue gas flow	500-700	Nm^3/h
Flue gas CO_2 content	0-10	Vol%
Solids circulation rate	0-400	(kg/h)

The operations team was able to gather more than 1200 hours of operating experience in hot operation in with a capture performance between 200 and 700kg of capture CO_2 per day, and more than 250 hours in cold operation (>1450 hours combined). Figure 16 and Figure 17 show the view of the graphic user interface (GUI) of the process control system during a steady-state operating point, where critical operating parameters were kept constant. During stable operation it was possible to keep stable desorber and adsorber temperatures, as well as a constant solids circulation rate (SCR), capture efficiency and capture rate for multiple hours per datapoint. The achieved performance parameters are summed up in Table 2.

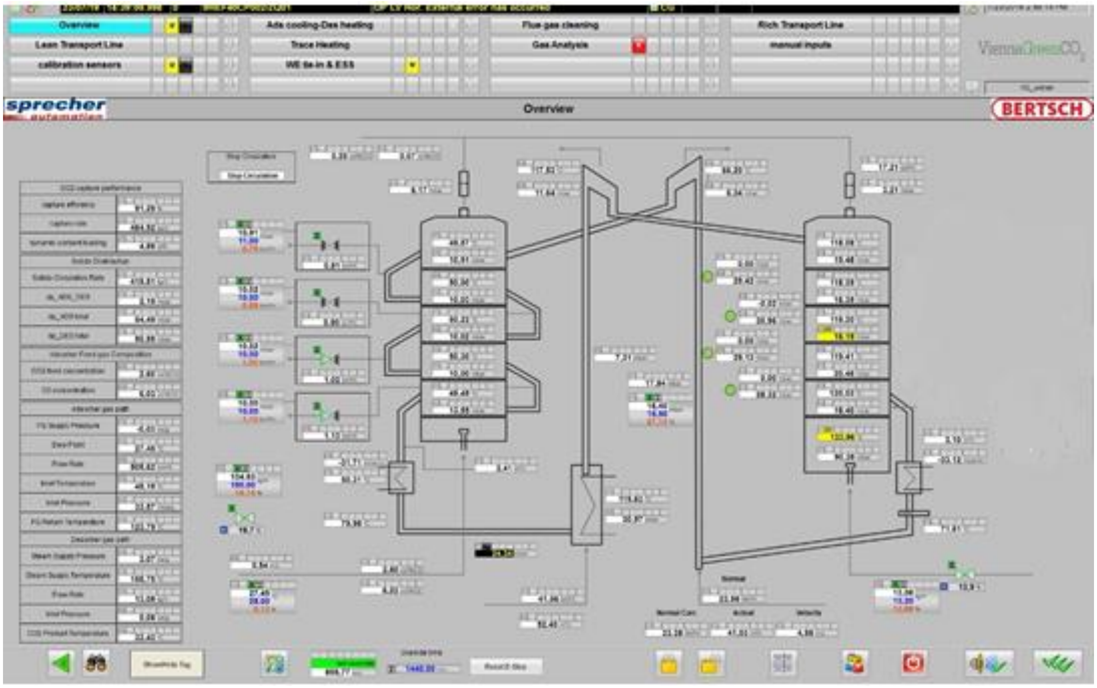

Figure 16: View of the process control system during a steady-state operating point (Screen#1)

Figure 17: View of the process control system during a steady-state operating point (Screen#2)

Figure 18: View of the predesorber and the lean heat exchanger unit (courtesy of Shell Austria GmbH)

Figure 19: Group photo at the pilot plant opening ceremony (courtesy of Shell Austria GmbH)

4.3 Greenhouse fertilization study

One goal of the ViennaGreenCO_2 project was to investigate whether it is possible to produce renewable CO_2 locally for use as a fertilizing agent in greenhouses farming. In this way local CO_2 capture and utilization (CCU) scenario could be established in the vicinity of the Wien Energie combined heat and power (CHP) plant in Wien, Simmering. A study was conducted together with the Research Institute of Organic Agriculture Austria (Forschungsinstitut für biologischen Landbau Österreich, FIBL Austria) to evaluate the quantity that would be needed to provide CO_2 for the horticultural operations in Simmering, to judge whether the TSA product gas would be suitable for this purpose, and to evaluate the costs of production and distribution of the CO_2 product. From the total area of 100 hectares area under protected greenhouse production in Vienna Simmering, gardeners apply CO_2 on 50 hectares. Of these around 30 hectares are still using the district heating, which lead to a potential annual demand of 9.000 t of CO_2 per year considering a CO_2 application rate of 30kg CO_2/m²/year.
To understand whether the CO_2 product has the desired quality, it was necessary to monitor the gas stream during the pilot plant experiments for trace components, which may originate from the biomass CHP flue gas or from the TSA process itself. The CO_2 product gas stream was continuously monitored online with an FTIR gas analyzer during the experimental campaign as well as sampled and analyzed by GC-MS offsite. In the CO_2 product, traces of ammonia and hydrocarbons were identified, but both were within the threshold limits reported in literature. From a horticultural point of view, it would therefore be possible to use the raw TSA product gas as a CO_2 rich greenhouse fertilizer.

4.4 Technoeconomic assessment

By operating the pilot plant, operational experience and data were gathered, and these learnings were also used for a techno-economic assessment of the technology. Based on the design of the pilot unit, and the learnings throughout the course of the ViennaGreenCO_2 project, a design for a next-scale TSA plant for CO_2 capture at commercial scale was developed by a project partner (Van Paasen et al., 2018).

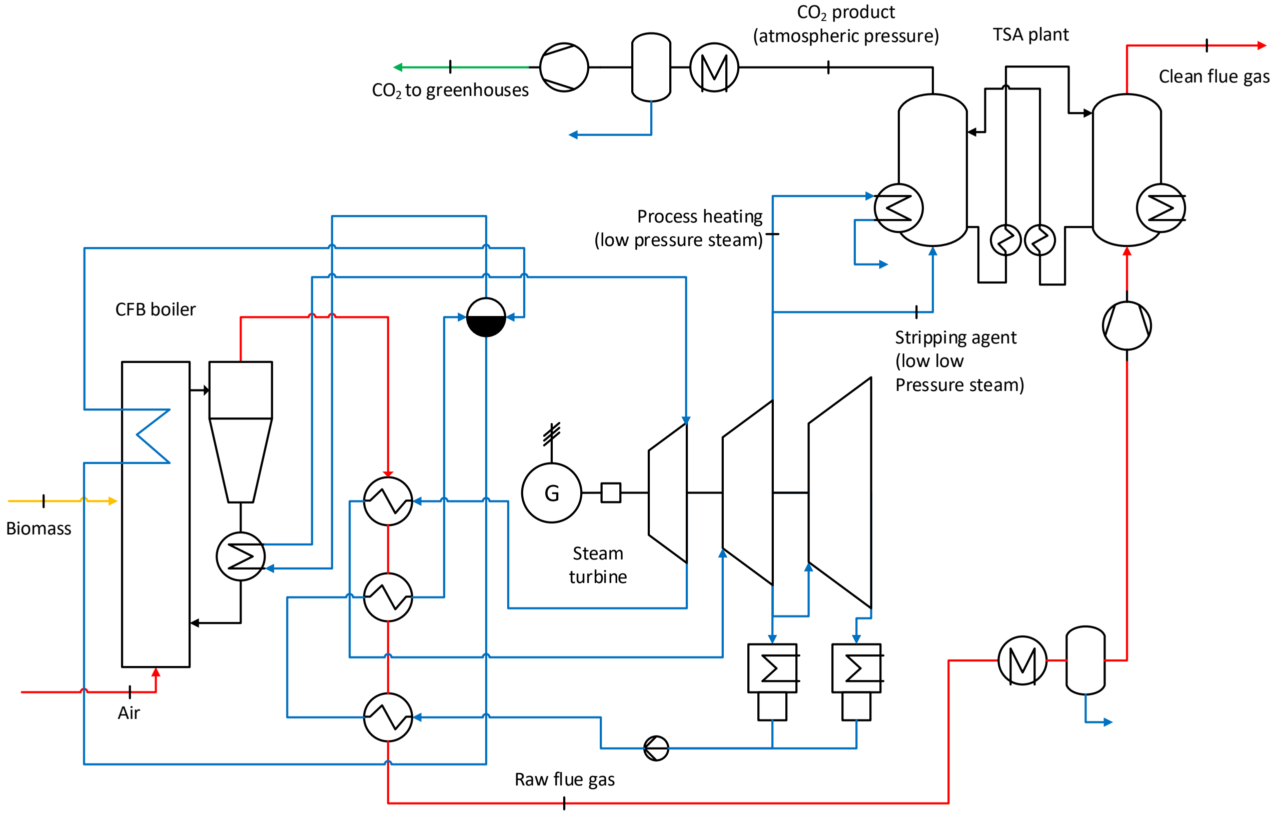

Figure 20: Process flow scheme of a TSA plant for local production of green CO_2 for greenhouse farming in Wien Simmering

The solid sorbent TSA concept published by Van Paasen et al. however was designed for industrial scale CCS applications from natural gas fired power stations. To understand whether the TSA technology is also suitable for the local production of renewable CO_2 for greenhouse farming in the vicinity of Wien Simmering, the capture cost was assessed for a TSA plant with a capture capacity of 9.000 tonnes of CO_2 per year, as mentioned in chapter 4.3. A simplified process arrangement is displayed in Figure 20. The capital expenditure (CAPEX) for the erection of such a TSA plant was estimated by Bertsch with roughly 12.57 Million Euros. This estimation includes the manufacturing of large vessels (columns, heat exchangers), piping, instrumentation and control engineering, electrical work, pumps and blowers, assembly, engineering, and steel construction. A total steam demand of roughly 3.73 GJ/t_{CO2} and a power consumption of 0.19 GJ/t_{CO2} for the operation of boosters and pumps were calculated with the process modelling tool developed in WP4 for steady state operation at design load. This estimation is based on energy consumption only (steam and electricity), other cost factors such as sorbent makeup or labour are not included. Assuming a depreciation time of 20 years and a plant availability of the Wien Energie biomass CHP plant of 8000 hours per year, the specific capture cost per mass of captured CO_2 is estimated to be in the range of 100-125 €/t_{CO2}. In case new ways to sell or utilize the produced CO_2 can be found and more CO_2 is produced via solid sorbent CO_2 capture from the biomass CHP, the capture cost per mass of CO_2 could be reduced to values as low as 50-60 €/tCO_2 (assumption: 90% of the CO_2 emitted by the biomass CHP is captured).

5 Outlook and recommendations

A pilot unit was designed, manufactured, erected, commissioned and an experimental campaign was conducted in order to assess the performance of the solid sorbent TSA process. Results of the final assessment of the technology indicate that this technology has great potential for efficiently capturing CO_2 from power plant flue gases. Furthermore, it was shown that it is possible to provide renewable CO_2 for utilization as a fertilizer in greenhouse farming, which is produced regionally from a biomass CHP in Wien, Simmering. The pilot plant was operated for more than 1200 hours by the operations team and a lot of experience was gathered by all project partners during the course of the project.

6 Literature

Esmeijer, M.H. (1999). CO2 in Greenhouse Horticulture. Applied Plant Research. Wageningen

Hofer, G., Fuchs, J., Schöny, G, Pröll, T., 2017. Dimensioning tube bundle heat exchangers for a continuous temperature swing adsorption process. Presented at the 12th international conference on fluidized bed technology (CFB-12), May 23rd-26th 2017, Cracow, Poland

Hofer, G., Schöny, G., Fuchs, J., Pröll, T., 2018. Investigating wall-to-bed heat transfer in view of a continuous temperature swing adsorption process. Fuel Processing Technology 169, 157-169.

Hofer, G., 2019. Fluidized bed heat transfer and solids mixing in view of a post-combustion carbon capture process. PHD Thesis. University of Natural Resources and Life Sciences, Vienna, Austria.

Pröll, T., Schöny, G., Sprachmann, G., Hofbauer, H., 2016. Introduction and evaluation of a double loop staged fluidized bed system for post-combustion CO_2 capture using solid sorbents in a continuous temperature swing adsorption process. Chemical Engineering Science 141, 166-174.

Schöny, G., Zehetner, E., Fuchs, J., Pröll, T., Sprachmann, G., Hofbauer, H., 2016. Design of a bench scale unit for continuous CO_2 capture via temperature swing adsorption—Fluid-dynamic feasibility study. Chemical Engineering Research and Design 106, 155-167.

Schöny, G., 2017. The ViennaGreenCO_2 project - Demonstration of low temperature solid sorbent CO_2 capture at pilot scale. Presented at the 74th IEA-FBC Meeting, Kraków, Poland.

Schöny, G., Dietrich, F., Fuchs, J., Pröll, T., Hofbauer, H., 2017. A multi-stage fluidized bed system for continuous CO_2 capture by means of temperature swing adsorption – First results from bench scale experiments. Powder Technology 316, 519-527.

Zehetner, E., Schöny, G., Fuchs, J., Pröll, T. Hofbauer, H., 2017. Fluid-dynamic study on a multistage fluidized bed column for continuous CO_2 capture via temperature swing adsorption, Powder Technology 316, 528-534.

Dietrich, F., Schöny, G., Fuchs, J., Hofbauer, H., 2018. Experimental study of the adsorber performance in a multi-stage fluidized bed system for continuous CO_2 capture by means of temperature swing adsorption. Fuel Processing Technology 173, 103-111.

Pirklbauer, J., Schöny, G., Pröll, T., Hofbauer, H., 2018. Impact of stage configurations, lean-rich heat exchange and regeneration agents on the energy demand of a multistage fluidized bed TSA CO_2 capture process. International Journal of Greenhouse Gas Control 72, 82-91.

Van Paasen, S., Bhalodi, A., Grigoriadou, K., Infantino, M., Van de Graf, J., 2018. Process development for large scale solid sorbent post combustion CO2 capture technology for application to natural gas fired power stations. Presented at the 14th International Conference on Greenhouse Gas Control Technologies, GHGT-14, 21st -25th October 2018, Melbourne, Australia.

7 Contact

TU Wien
Institut für Verfahrenstechnik, Umwelttechnik und Technische Biowissenschaften
Getreidemarkt 9/166, 1060 Wien
http://www.vt.tuwien.ac.at

University of Natural Resources and Life Sciences, Vienna (BOKU)
Institute of Chemical and Energy Engineering
Peter Jordan Straße 82, 1190 Wien
https://boku.ac.at/

Josef Bertsch GesmbH & Co KG (Bertsch)
Herrengasse 23, 6700 Bludenz
https://www.bertsch.at/

Shell Global Solutions International B.V. (Shell)
Grasweg 31, NLD 1031HW Amsterdam
https://www.shell.com/

Wien Energie GmbH (WE)
Klestil-Platz 14, 1030 Wien,
https://www.wienenergie.at

M-TEC Energie.Innovativ GmbH (M-TEC)
Arnreit 51, 4122 Arnreit, Österreich
https://m-tec.at/

lk-projekt niederösterreich | wien GmbH (lk-projekt)
Wiener Straße 64, 3100 St Pölten
http://www.lk-projekt.at/

LGV-Frischgemüse Wien reg.Ges.m.b.H. (LGV)
Haidestraße 22, 1110 Wien
https://lgv.at/