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Modelling of the CO_2 process- and transport chain in CCS systems—Examination of transport and storage processes

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A B S T R A C T

Given the development of power plants with integrated capture of carbon dioxide and the subsequent storage of this captured CO_2 (CCS—carbon dioxide capture and storage) the future fossil fuel-based energy system will most likely consist of very different types of processes and units. Past studies mostly focused on a very specific part of this CCS system. The effects and reactions on bordering processes were often only rudimentally considered. Due to these complex interactions between individual parts of a CCS system it is necessary to examine the behaviour of the whole system in order to achieve secure and efficient operation. This article intends to highlight problems that will occur when such interactions are not considered and examined properly.

This work presents an approach for the examination of a system consisting of a power plant, capture unit, transport system and CO_2 storage facility. A number of typical problems are shown, with focus on the examination of thermodynamic behaviour of the captured CO_2 in a pipeline followed by a well in a saline aquifer storage site. It is shown that, under special conditions, the combination of a CO_2 pipeline and a well down to a saline aquifer will not work due to phase changes and pressure conditions, which would lead to operational problems or at the very least to partial destruction of either the pipes or the sedimentary storage rock.

1. Introduction

In many countries there has been a change in opinions regarding the current usage of fossil fuels. This for example is expressed in obligations to reduce greenhouse gas emissions and ratification of the Kyoto Protocol by many countries. This rethinking also leads to an increase in research concerning the reduction of CO_2 emissions from power plants. Besides the increase in efficiency the so-called CCS technology is expected to help reduce emissions to an acceptable level (IPCC, 2005).

Carbon dioxide capture and storage (CCS) is a synonym for a variety of operations with the goal to separate and store CO_2, which is produced when using fossil fuels. In the medium term the separation of CO_2 from the earth's carbon cycle is expected to decrease the concentration in the atmosphere or at least maintain current levels. This is expected to counteract climate change until world wide energy supply is primarily supported by renewable, low-emission energy resources. Therefore CCS technology is often referred to as a bridge between the fossil fuel period and the future solar/renewable energy period.

CCS consists of many elements (concepts, processes, units, mass flows, etc.), which are partly interconnected. In Germany, there is a tendency to examine and test only a few possible concepts (Göttlicher, 2004). These include oxyfuel combustion and the integrated gasification combined cycle (IGCC) as well as amine scrubbing of the flue gas followed by pipeline transport and storage of the captured CO_2 in a saline aquifer in northern Germany (WI, 2007).

In this work an approach to analyse the behaviour of the whole CCS system will be presented. This system consists of a power plant with an integrated CO_2 capture system, a pipeline for transportation and a saline aquifer for storage of the captured CO_2 (this will represent the actual possibilities in Germany).

Special consideration is given to reactions generated as a result of the pipeline connection affecting power plants and storage units.

In order to reduce the variables in the first step when designing the general procedural method, a CCS-reference scenario will be developed and examined.

2. Methods

2.1. Simulation programs

The current software market has very good products available for the calculation and simulation of individual applications (power plants, chemical plants, pipelines, storage). But the possibility of using a single commercial software package is restricted by the multidisciplinary approach of the presented research project

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(Nimtz et al., 2009). The modifications that would be necessary in order to use a single existing program for all of the questions described above are too complex. Even if the required interfaces were available in the programs, the number of modifications is nearly equal to the design of a completely new program.

Therefore it was decided to use the well established programming and calculation environment MATLAB®/Simulink® to develop the algorithms and to simulate the CCS system. In the first step, when considering simple systems it is also possible to use spreadsheet calculation programs or similar software to perform the calculations.

2.2. CCS scenario

The aim of this paper is also to present the need for the overall examination of the described systems. Therefore a very simple scenario consisting of a system with a power plant, CO_2 transport and a storage facility will be presented and a number of problems will be discussed.

This scenario is generated based on the following conditions:

- fuel—lignite;
- emission of pure CO_2;
- electrical net power—600 MW;
- electrical gross efficiency—50%;
- electrical net efficiency—40%;
- pipeline, subsurface type
  - length—150km;
  - horizontal ground laying;
- storage—saline aquifer
  - Depth of aquifer—1000 m.

Further simplification leads to the treatment of the power plant and the capture unit as black boxes. Therefore it is assumed that the power generation and capture units are able to supply the pipeline with pure pressurised carbon dioxide. The kind of power plant or capture unit that is represented here is not stated as this is not significant here.

The storage site is represented by a model from Wiese et al. (submitted for publication), see this special issue of Chemie der Erde / Geochemistry. All processes are assumed to be under static conditions without considering any transient behaviour.

2.3. Boundary conditions

For model generation, a number of boundary conditions are assumed. The power plant is described under section 3.2. Additional parameters can be seen in Table 1.

The transport system consists of a subsurface pipeline, properties can be seen in Table 2 (IPCC, 2005). The length of the pipeline represents a typical distance between power plants and storage facilities in east Germany. The diameter is chosen so that flow velocity is not too high (0.5,…,2 m/s) and was to ensure that the pressure losses along the pipeline are minimal (overall 15,…,25 bar). No additional valves or other units are considered, which would increase the frictional losses.

Table 1
Power plant/CO_2 mass flow.

Property	Value	Unit
Full load hours	7000	[h/a]
Capture rate[a]	90	[%]
Captured CO_2[b]	3.7	[mil. t/a]
CO_2 mass flow[c]	422.2	[t/h]

[a] To account for losses e.g. in a purification unit.
[b] Pure CO_2 assumed.
[c] Average value.

Table 2
Pipeline parameters.

Property	Value	Unit
Length	150	[km]
Internal diameter	0.5	[m]
Heat transfer coefficient	4[a]	[W/(m^2 K)]
Pipe roughness	0.0005	[m]

[a] Estimated, constant over the whole length.

Table 3
Boundary conditions of CO_2 flow.

Property	Inlet pipeline	Min.	Max.	Outlet bottom of the well	Unit
Pressure	< 200	p_c+10	< 200	< 130[a]	[bar]
Temperature	50	> 10[b]	50	> 10[b]	[°C]

[a] Defined by rock stability of the storage site.
[b] Possibility of hydrate formation if CO_2 flow contains water.

Table 4
Storage conditions.

Property	Value	Unit
Hydrostatic pressure gradient	0.11	[bar/m]
Maximum excess pressure	20	[bar]
Temperature gradient	0.035	[K/m]
Temperature at 0 m	10	[°C]

The pressure and temperature of the CO_2 flow are restricted due to material, technical and economical factors. This is also true for the conditions in the injection.

The maximum pressure is restricted by the thickness of the pipe walls (economically restricted) and also by the existing compressors for large amounts of CO_2. It is set to 200 bar. The minimum pressure is restricted by the phase behaviour of CO_2. Under normal circumstances, carbon dioxide is transported under supercritical conditions (Barrie et al., 2005), at pressures above p_c=73.773 bar (Span and Wagner, 1996) so that no phase transition occurs when temperature changes. This is required so as to avoid two-phase flow in the pipeline, which could cause cavitation and pressure peaks and would most likely damage the pipeline. The transport pressure should be at least 10 bar above the critical pressure p_c to ensure that supercritical conditions are maintained even under transient flow conditions (Zhang et al., 2006). The minimum pressure is expected to appear at the end of the pipeline or at the highest points of the pipeline route.

The temperature must not exceed 50 °C in order to ensure that any outer anti-corrosion coating of the steel pipe is not destroyed. The discharge temperature of the compressor is assumed to be 40 °C. With decrease in temperature, density increases, which will reduce pressure losses along the pipeline due to lower flow velocity. If there is water in the CO_2 flow and temperature decreases hydrates could be formed (De Visser et al., 2008). Therefore the minimum temperature is set to 10 °C (Ullrich and Eggers, 2004). A list of the allowable pressures and temperatures can be found in Table 3.

The storage site is taken from a model by Wiese et al. (Wiese et al., submitted for publication), see this special issue. It represents some results from the examination in *Ketzin*, which is the first CO_2 storage site in Germany (Schilling et al., 2009). Storage is in an underground saline aquifer. Its depth is 1000 m and its basic conditions can be seen in Table 4. It was calculated

that about 60 wells would be required in order to store the amount of CO_2 described above (using flow rate in single well from Wiese et al., submitted for publication, see this special issue) if full injection rate is assumed.

The wells are represented by a single well, which transports 1/60 of the overall mass flow. The arrangement of the 60 wells on the surface is not considered. A well is modelled as a vertical pipe without any valves or other flow barriers, see Table 5.

The calculation of the thermodynamic behaviour of CO_2 in this CCS system consists of the stepwise calculation of pressure losses and the heat transfer from or to the fluid. All properties of CO_2 are calculated with equations for pure carbon dioxide (Span and Wagner, 1996; Vesovic et al., 1990).

Heat transfer is calculated with a static heat transfer coefficient for the pipeline and the well. The pressure loss is calculated with the equations for turbulent pipe flow from the VDI Heat Atlas (VDI, 2006, Chapter Lab 1).

3. Results

For the CCS-reference scenario, a few typical problems will be presented. These problems include:

- transport and injection; maintaining minimum pressure at the end of the pipeline above 85 bar;
- transport and injection; ensuring the pressure at the bottom of the well is less than the maximum pressure;
- transport, pre-conditioning and injection; avoiding two-phase flow in pipeline and well;
- transport and injection, alternative conditioning measures;
- influence of impurities on transport and storage.

Table 5
Well parameters.

		Unit
Length	1000	[m]
Tubing diameter	0.12	[m]
Heat transfer coefficient	10[a]	[W/(m^2 K)]
Pipe roughness	0.0005	[m]
Number of wells	60	[–]

[a] Estimated, constant.

3.1. Case 1: injection of CO_2 without pre-conditioning, minimum pressure at the end of the pipeline

This is a scenario whereby carbon dioxide is transported to the storage facility and the minimum pressure (at the end of the pipeline, just before entering the well) should not fall below the critical pressure. As a safety factor it is assumed that the pressure should not fall below 85 bar.

The results for pipeline transport are shown in Fig. 1. The discharge pressure from the compressor has to be 98 bar. Along the pipeline CO_2 cools down and density increases. This results in a decrease in flow velocity.

When entering the well, the CO_2 pressure increases due to hydrostatic pressure. At the bottom of the borehole at a depth of 1000 m, the pressure would be above 174 bar. This is a result of the high density of the CO_2 along the whole well (see Figs. 2 and 3). The temperature and flow velocity in the well are shown in Table 6.

3.2. Case 2: injection of CO_2 without pre-conditioning, safe pressure at the bottom of the well

In this scenario, the maximum excess pressure at the bottom of the well is maintained and does not exceed 20 bar. This leads to a maximum pressure of 130 bar at the end of the well. The pressures upstream are dependent on this pressure. The resulting CO_2 properties along the pipeline and well are presented in Figs. 4 and 5.

As can be seen in Fig. 4, the inlet pressure is about 90 bar. Due to friction, the pressure decreases along the pipeline and the CO_2

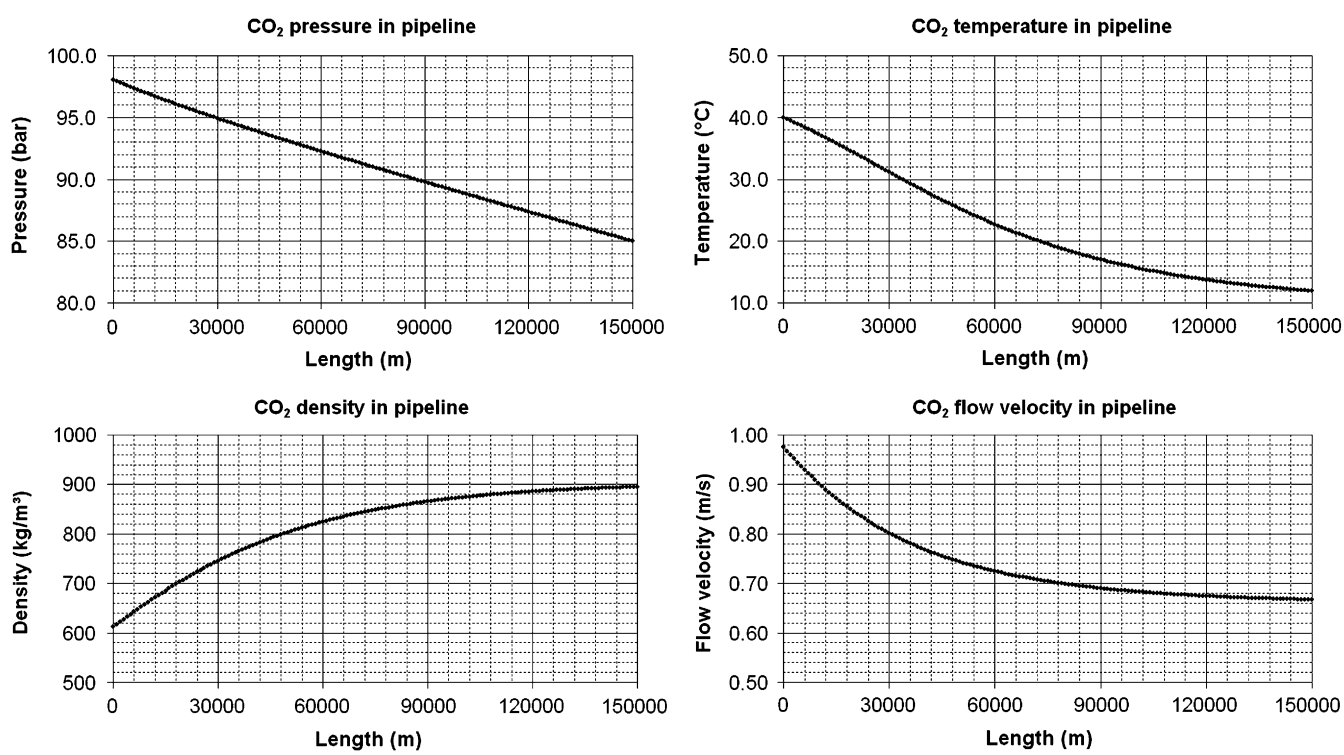

Fig. 1. Behaviour of the transported carbon dioxide from pipeline inlet to pipeline outlet (case 1).

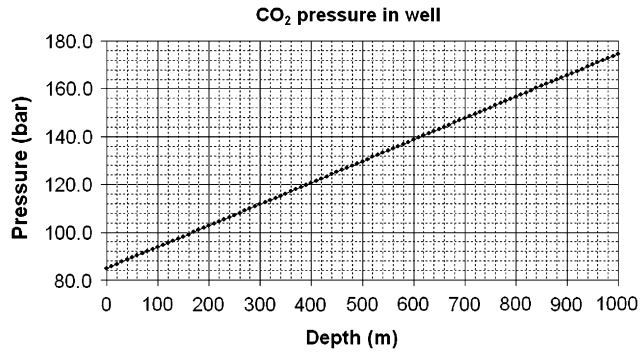

Fig. 2. Pressure of CO_2 in the well (case 1).

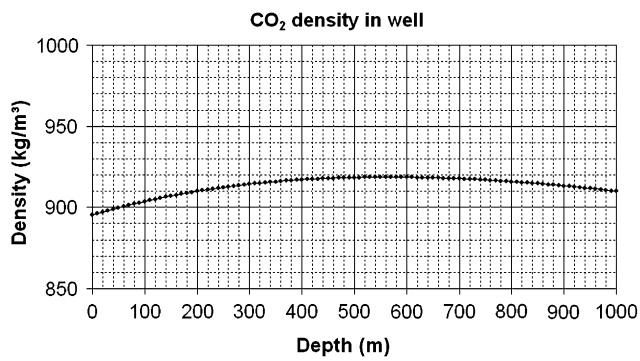

Fig. 3. Density of CO_2 in the well (case 1).

Table 6
CO_2 behaviour in well (case 1).

Property	Value	Unit
Inlet temperature	12	[°C]
Outlet temperature	22	[°C]
Inlet flow velocity	0.193	[m/s]
Outlet flow velocity	0.190	[m/s]

phase changes from supercritical state to gaseous state at the pipeline outlet. This leads to a decrease in density from 468 kg/m^3 at the inlet to about 181 kg/m^3 at the end of the pipeline. The flow velocity triples from 1.3 to 3.3 m/s. This leads to an increase in pressure losses due to friction.

When entering the well, the pressure of CO_2 increases and a subcritical phase change from gaseous state to liquid state occurs at a depth of 130 m. The complete transport and storage process is presented in Fig. 6, where all changes in pressure, temperature and density are plotted in a pressure–density diagram.

3.3. *Case 3: injection of CO_2 with pre-conditioning, safe pressure at the bottom of the well and minimum pressure at the end of the pipeline*

This case represents a simple solution to ensure that the pressure at the bottom of the well is below 130 bar, ensure that the minimum pressure in the pipeline is above 85 bar and to avoid two-phase flow within the well.

As will be shown, a further unit is needed consisting of a heater and a throttle to avoid a phase change when entering the well. Table 7 shows the results for the pipeline transport.

To ensure that the pressure at the bottom of the well is below 130 bar, it is necessary to throttle the CO_2 before entering the well. Due to the decrease in temperature when throttling, a phase change would usually occur. This is avoided by heating the CO_2 before throttling. To achieve a temperature of 40 °C at the wellhead, it is necessary to warm up the CO_2 to about 48 °C. The process states are listed in Table 8. Fig. 7 shows the behaviour of the CO_2 in the well.

3.4. *Case 4: injection of CO_2 with alternative conditioning, safe pressure at the bottom of the well and minimum pressure at the end of the pipeline*

If reheating and throttling of CO_2 before injection is not possible, another option is to install a throttle at the end of the well. Pipeline transport and the behaviour in the well are the same as in case 1. The difference is the throttling at the end of the well. Table 9 shows the properties of CO_2 before and after the throttle.

4. Discussion

4.1. *Injection without pre-conditioning (case 1)*

As can be seen in Fig. 2, the pressure at the bottom would be about 174.6 bar. If a maximum excess pressure of 20 bar is permitted to avoid rock fractures in the sediment layer, this case cannot not be realised without further measures. It is reported that the maximum injection pressure could probably be higher than the hydrostatic pressure of brine+20 bar (needs to be permitted by Administrative Offices of Mining/Geology). If the excess pressure was allowed to rise up to 70,…,80 bar, then case 1 would be preferable. Otherwise a way needs to be found to decrease the pressure at the bottom.

In principle there are two ways to lower the pressure at the bottom:

- decrease the density of the CO_2 column to decrease the weight of the column and therefore decrease the hydrostatic pressure at the bottom;
- Throttling of the flow before entering the aquifer.

Both solutions are presented in cases 2–4, and will be discussed in the following sections.

4.2. *Injection without pre-conditioning (case 2)*

This case represents the simplest way to keep the pressure under the reported maximum bottom pressure of 130 bar. The goal is to decrease the average density of the CO_2 column in the well. Therefore the pressure has to be lowered at the wellhead if we assume the temperature as a given value. The results are presented in Figs. 4 and 5. The CO_2 stream enters the wellhead under subcritical conditions (23.3 °C, 57.7 bar) in the gaseous phase. While flowing down the well, the pressure rises and a phase change from gaseous to liquid will take place at approximately 60.86 bar and 22.6 °C at a depth of 135 m.

This phase change may result in instability in the flow and cavitation in the pipe. It would also lead to high and low pressure peaks, which oscillate within the pipe and are undesirable (Ramamurthi and Sunil Kumar, 2003).

Within the pipeline, there is a supercritical phase change. When CO_2 flows through the pipe, its pressure decreases due to friction but the temperature remains above the critical temperature of 30.98 °C (Span and Wagner, 1996). Therefore there is no

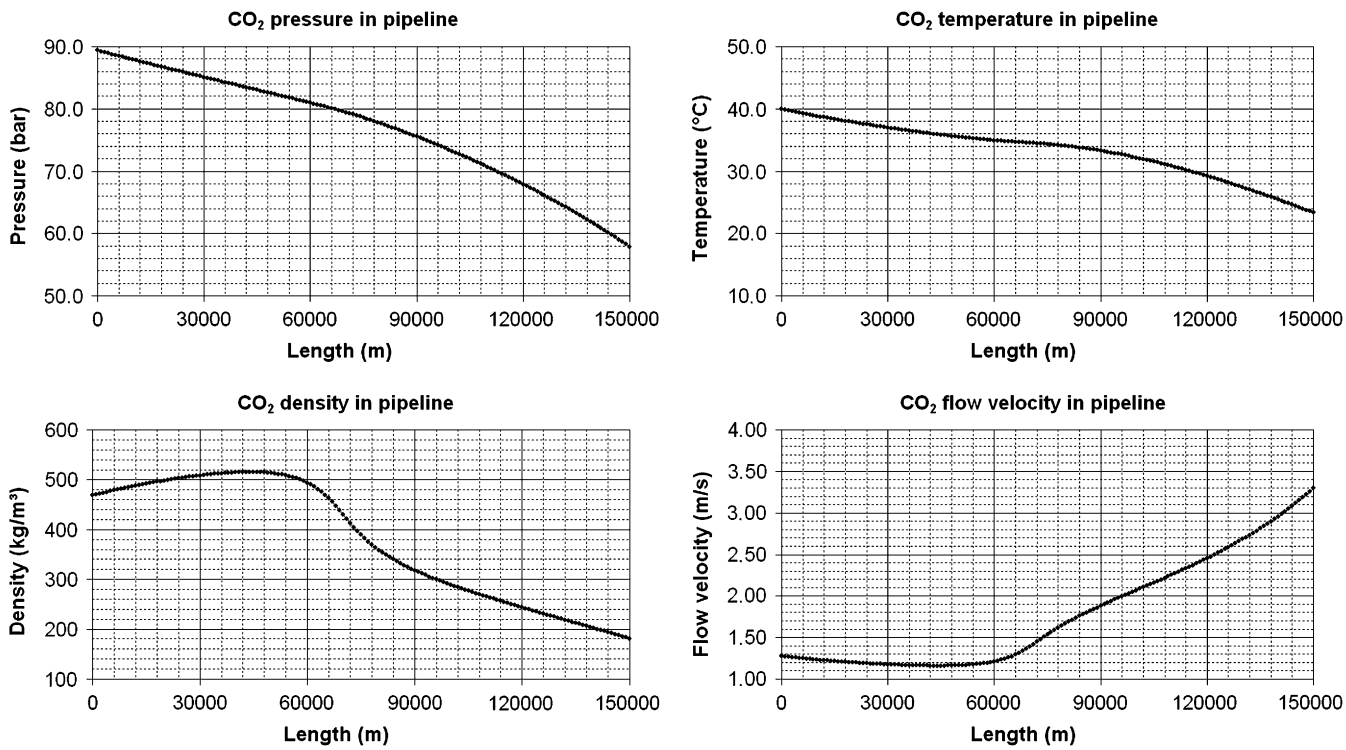

Fig. 4. Behaviour of the transported carbon dioxide from pipeline inlet to pipeline outlet (case 2).

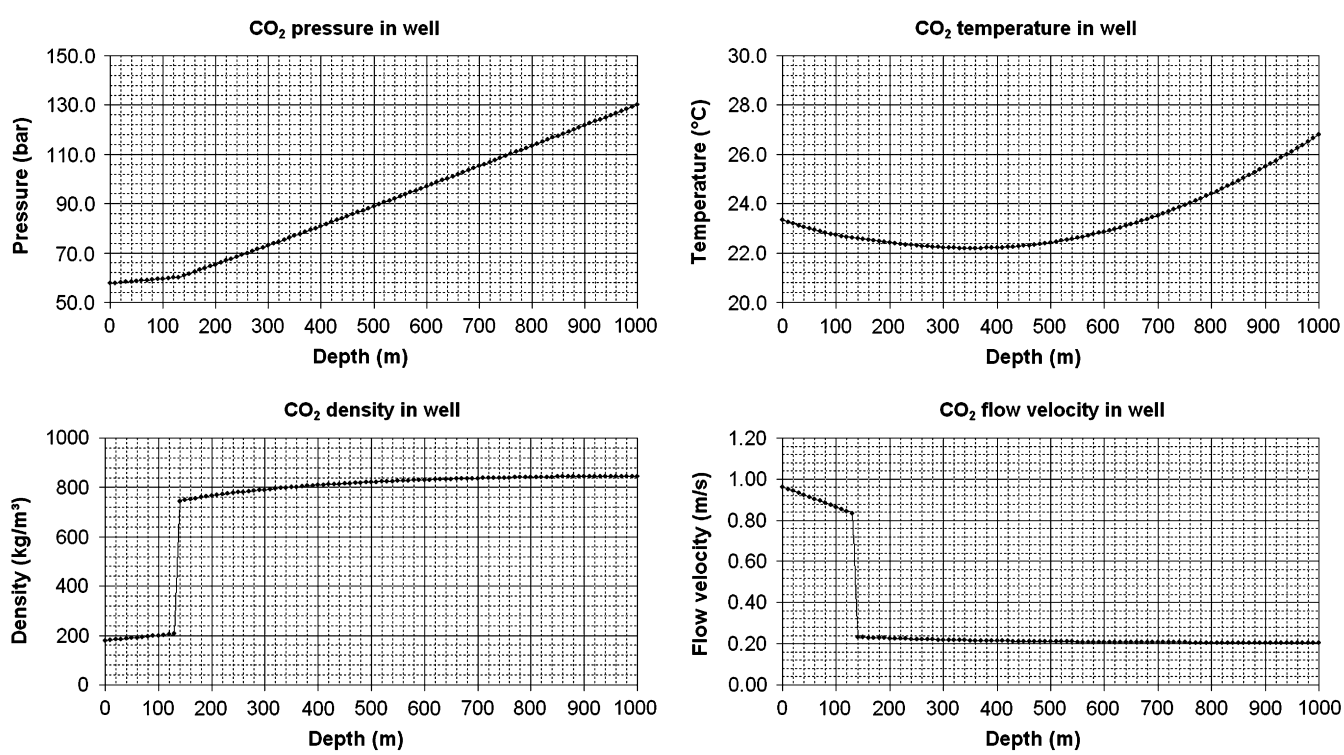

Fig. 5. Behaviour of the transported carbon dioxide from well inlet to well bottom (case 2).

real phase change such as condensation or evaporation; instead the density changes continuously so that CO_2 is in the gaseous phase at the end of the pipeline (see Fig. 6). A phase change like this is not critical.

4.3. Injection with pre-conditioning (case 3)

To avoid two-phase flow in the well (and also in the pipeline), another possibility to decrease the density within the well exists.

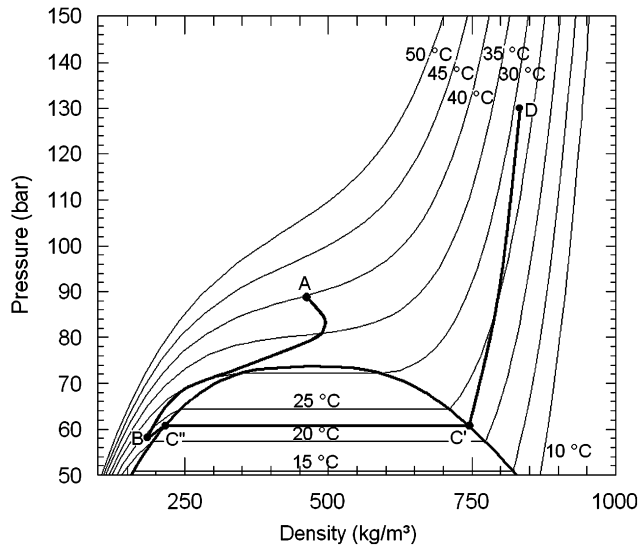

Fig. 6. Transport and storage process in pressure–density diagram (case 2). Marked points: A, pipeline inlet; B, pipeline outlet/wellhead; C″, beginning of phase change; C′, end of phase change; D, bottom/reservoir.

Table 7
CO_2 behaviour in pipeline (case 3).

Property	Value	Unit
Inlet pressure	98.4	[bar]
Outlet pressure	85	[bar]
Inlet temperature	40	[°C]
Outlet temperature	12	[°C]
Inlet density	616	[kg/m^3]
Outlet density	895	[kg/m^3]
Inlet flow velocity	0.97	[m/s]
Outlet flow velocity	0.67	[m/s]

Table 8
CO_2 behaviour in heater and throttle (case 3).

Property	Value	Unit
Inlet pressure (heater)	85	[bar]
Outlet pressure (heater)	85	[bar]
Inlet temperature (heater)	12	[°C]
Outlet temperature (heater)	48	[°C]
Inlet pressure (throttle)	85	[bar]
Outlet pressure (throttle)	74.8	[bar]
Inlet temperature (throttle)	48	[°C]
Outlet temperature (throttle)	40	[°C]
Outlet density (throttle)	228	[kg/m^3]
CO_2 mass flow	117.3	[kg/s]
Heater power	22.96	[MW]

For this scenario the flow has to be throttled and heated. To avoid the CO_2 cooling down too much, it is first heated and then throttled to a pressure that is a result of the maximum bottom pressure and the hydrostatic pressure from the CO_2 column in the well (and frictional losses). Fig. 8 shows the processes in the pressure–density diagram for CO_2. This figure was created with the help of the NIST Standard Reference Database 23 (REFPROP) (NIST, 2007). It represents the dependency of density on pressure and temperature in the typical range for the transport in pipelines and wells. The thick curve represents the liquid and vapour saturation lines. The thick line with the arrow marks the regime of heating and throttling process (starting at the right side). It can be seen that there is a strong dependency between density and temperature in the region near the critical point. Little changes in temperature cause high changes in density. This fact is used to identify the proposed solution (case 3).

The stream is heated (isobaric process) to approximately 48 °C and then throttled (isenthalpic process) to the pressure resulting from reservoir and well conditions. The upper temperature is calculated so that the temperature after throttling (and cooling due to the Joule–Thompson effect) is about 40 °C. This is necessary to ensure a safe distance to the critical temperature and therefore to stay in the supercritical phase.

Within the well the pressure rises and due to the supercritical temperature there is no phase change and so density increases continuously from 228 kg/m^3 at the wellhead to 770 kg/m^3 at the bottom.

Of course the heating requires a lot of energy. In this case, an additional 22.96 MW_{th} is needed for warming up the CO_2 stream. Normally, this energy will be provided by oil or natural gas; this has an impact on the amount of captured/avoided CO_2 and of course increases the costs of operation.

4.4. Injection with alternative pre-conditioning (case 4)

There is another possibility to control the pressure at the bottom of the well. This would be throttling of the flow down in the well, just before the bottom. It is not clear whether it is possible to design such a throttle, which should be able to throttle very different amounts of CO_2, which due to this must be a kind of controllable nozzle/diffuser combination. If it was possible to construct such a throttle, the next problem would be the maintenance, as it is usually not desirable (or possible) to open the well once it is sealed. The process is presented in Table 9.

4.5. Influence of impurities

All presented cases were calculated using the properties of pure CO_2. This simplification is applicable only if general statements are required. If other components are included in the CO_2 stream, the thermodynamic properties will change and will dramatically influence the transport and storage processes (De Visser et al., 2008; Anheden et al., 2005).

Possible impurities from the power plant and the capture process can include:

- Ar, CH_4, CO, H_2O, H_2S, N_2, SO_2 and others.

Typical ranges for mass percent impurities vary between 0.2 and 10 mass percent (Anheden et al., 2005).

To get an understanding of the influence of impurities a mixture of 95 mass-percent CO_2, 3 mass-percent N_2 and 2 mass-percent O_2 was examined. This mixture represents the composition of a simplified flue gas from an oxyfuel combustion process (dried flue gas, other possible impurities like Ar, SO_2, H_2O are represented by O_2 and N_2). A real flue gas would of course have a more complex composition (Holling et al., 2009), but in principle the behaviour can be seen with this mixture.

Most impurities are low-boiling compared to CO_2. When supercritical CO_2 is mixed with small amounts of these impurities, a homogeneous mixture is formed, but its thermodynamic behaviour is strongly influenced by these impurities.

This can be seen for example in the change in density. Fig. 9 shows the density of pure CO_2 and the described mixture over temperature. It is calculated using the GERG 2004 equation for natural gas mixtures (Kunz et al., 2007).

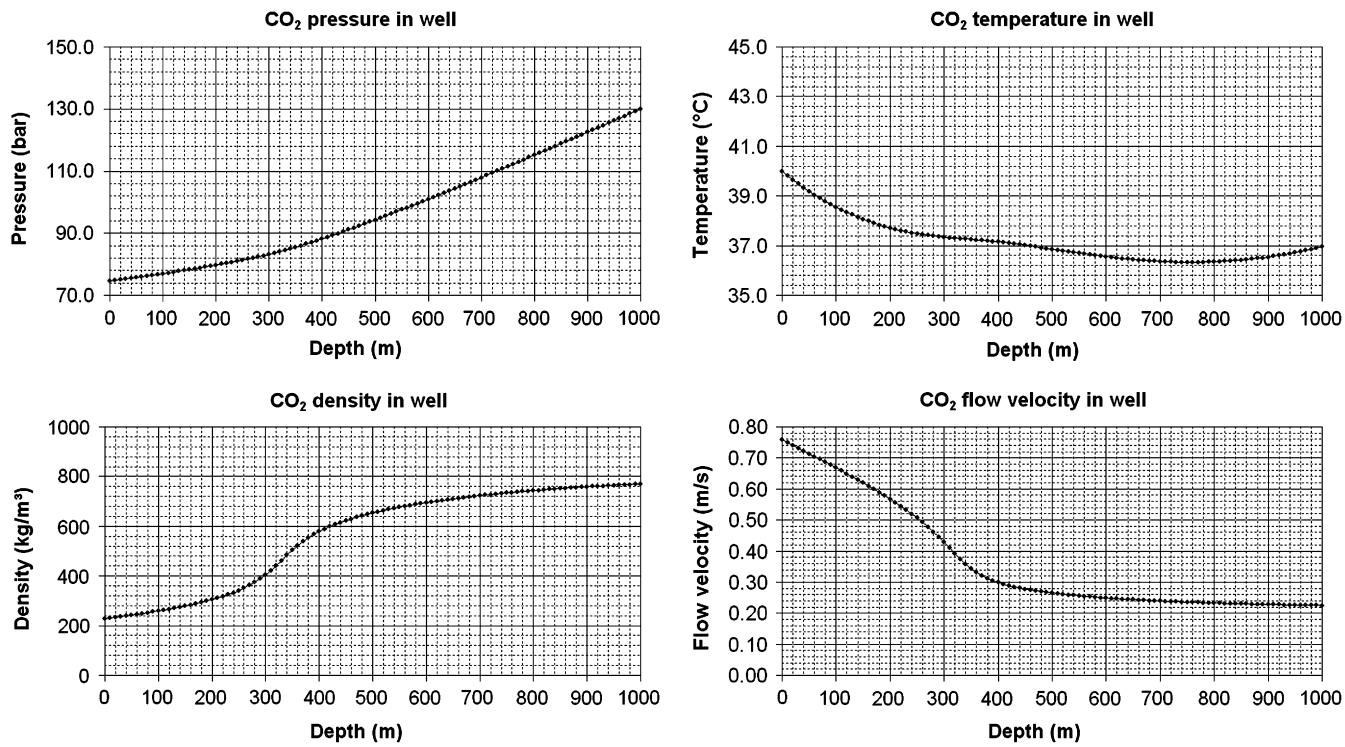

Fig. 7. Behaviour of the transported carbon dioxide from well inlet to well bottom (case 3).

Table 9
CO_2 behaviour in throttle (case 4).

Property	Value	Unit
Inlet pressure (throttle)	174.6	[bar]
Outlet pressure (throttle)	130	[bar]
Inlet temperature (throttle)	22.3	[°C]
Outlet temperature (throttle)	20.7	[°C]
Outlet density (throttle)	884	[kg/m^3]

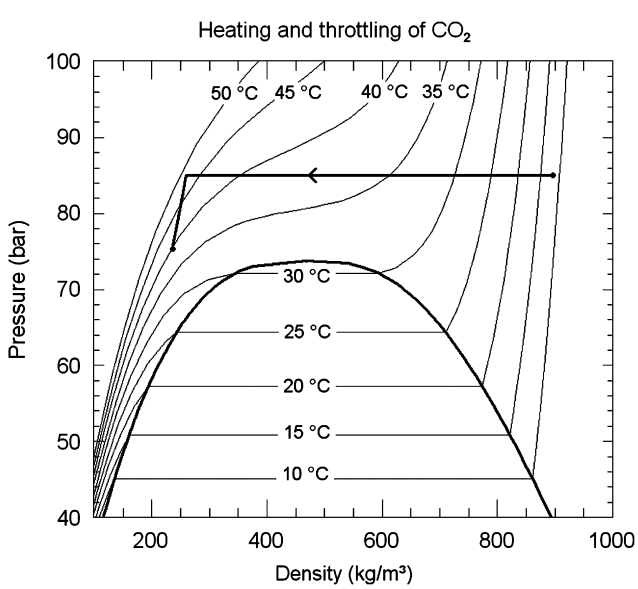

Fig. 8. Heating and throttling of the CO_2 stream (case 3).

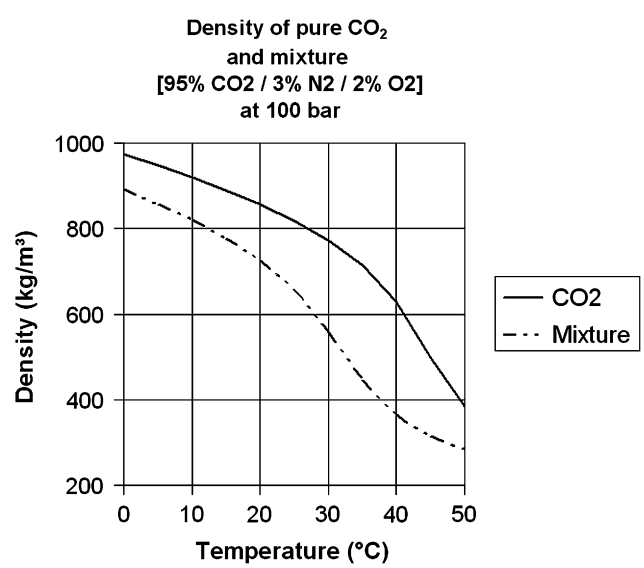

Fig. 9. Differences in densities of pure CO_2 and a CO_2 rich mixture at 100 bar.

The pressure of 100 bar is in the typical range of pipeline transport and storage conditions. It can be seen that the density is lowered due to the impurities. At 40 °C the density of the mixture is only about 60% of the density of pure CO_2. This would lead to a lower storage potential in the aquifer and to higher flow velocities (pressure drops).

Another problem is the phase behaviour of the mixture. The impurities lead to the formation of a phase envelope (bubble point curve and dew point curve) in the pressure–temperature diagram. This is the region where the mixture consists of two phases (liquid and gaseous phase); in Fig. 10 it is marked in grey. As explained above, such thermodynamic states have to be avoided during transport and injection.

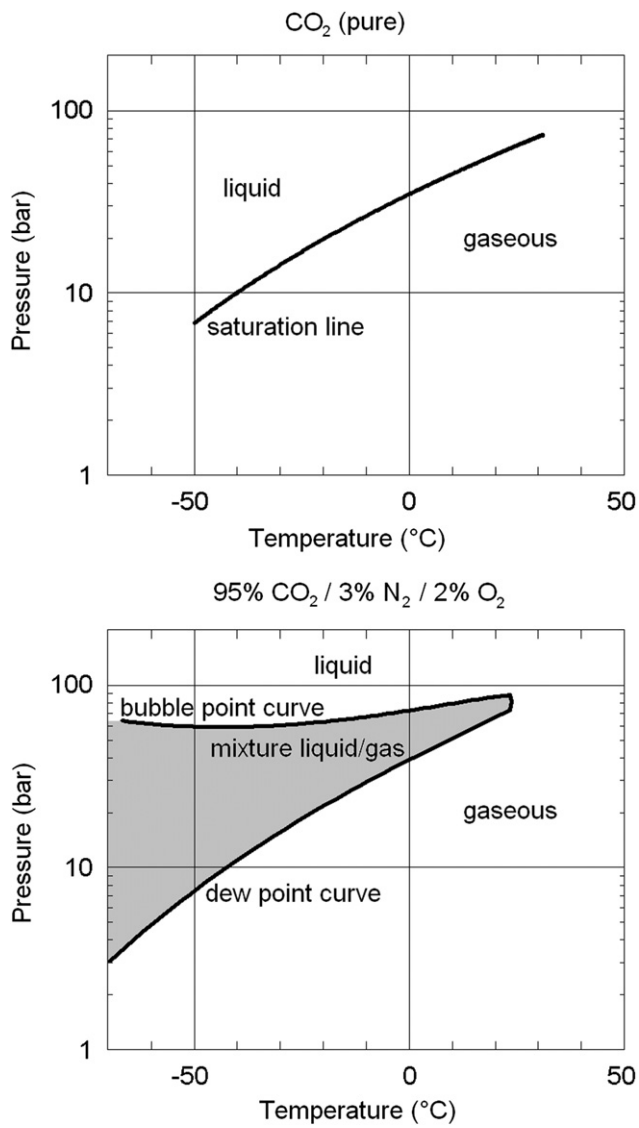

Fig. 10. Phase behaviour of pure CO_2 and a CO_2 rich mixture.

It can also be seen that the maximum pressure, where two phases occur (maxcondenbar point), is higher for the mixture. This point is important because it influences the whole system. Due to the possibility of phase changes, the pressure must be kept above the maxcondenbar point. This leads to higher pipeline pressures and higher energy demand from compressors.

If the captured CO_2 is not pure, it should be examined if a purification unit is affordable (Li et al., 2009). If the costs are not too high, this extra process would lead to easier operation of the pipeline and well and it would also increase storage volume.

5. Conclusions

The simple processes and problems presented in this article are only a few of a number of issues that need to be discussed if a safe, efficient and stable CCS system is to be established in the next 10–20 years. It was shown that a small change in the boundary conditions of such a system will lead to unexpected behaviour and sometimes technical feasibility is uncertain. It is imperative that research on CCS focuses on the connections between capture, transport and storage in the next few years.

To realise the long-term goal of the work the following questions will be addressed:

- What are the general conditions for operation of the CCS system?
- How much auxiliary energy is required by the additional units?
- What is the overall efficiency of the system?
- What differs when operating in partial load?
- What happens when applying load changes?
- What are the effects of malfunctions and disasters?
- How high are the avoided CO_2 emissions into the atmosphere?

Acknowledgements

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