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CO_2-sequestration in abandoned coal mines

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ABSTRACT

CO_2-sequestration in concealed coal mine reservoirs is inspired by storage of natural gas in coal mines. In contrast to the natural gas storage projects, sequestration of CO_2 requires that the gas would be secured for thousands of years. A specific hazard is that formation water would flood a CO_2-filled mine. The gas would then be compressed at the top of the reservoir, until possibly the pressure would be high enough to allow the gas to escape and finally be released into the atmosphere.

To prevent such a scenario, the reservoir pressure should be higher than the hydrostatic pressure of the surrounding formation water, thereby preventing the influx of formation water. Critical issues are if the top seal of the reservoir and former access shafts are able to withstand some overpressure, typically around 130 % of the hydrostatic pressure. In case of the Belgian Campine coal mines, the seal is formed by Cretaceous chalks and marls. Available tests and the documented behaviour of natural faults indicate a very low permeability, but the seal is currently insufficiently explored to allow a definite conclusion.

CO_2 stored in a coal mine may be present as pure CO_2, in solution in mine water, or adsorbed on coal. Each of these amounts can be calculated, but not all parameters are sufficiently known. Therefore they are split into an ascertained capacity, that can be calculated with a known accuracy or that is sufficiently conservative, and an additional potential capacity, an estimated surplus that can not be guaranteed or calculated accurately.

A case study for a Belgian colliery shows that sequestration in these coal mines is a viable option, even for the ascertained capacity alone. At an injection rate of 300 000 ton/y, sequestration can be guaranteed for about 25 years. This is a conservative estimate, and it may prove possible to inject at 500 000 ton/y during 25 years. This is a small, but still considerable contribution, approximately 3 to 6 % of the mitigation required to reach the Kyoto-target of Belgium, relative to the 1990 emission level.

INTRODUCTION

The effects of greenhouse gases on the earth's climate are increasingly better understood. As a result there seems to be a growing consensus among scientists that the current increase of the average temperature worldwide by 0.6 °C over the last century is at least partly related to human activity [1]. The combustion of fossil fuels causes most concern because of the large quantities of CO_2 that are produced as a waste or by-product [2].

This has encouraged many countries, among which the EU and its member states, to ratify the Kyoto protocol, which aims at reducing human induced greenhouse gas emission world-wide with 5 % by 2008-2012 relative to 1990. Although Kyoto represents just a fraction of the total effort required to stabilise the temperature increase, many countries are facing problems already to realise this objective. Technical and social evolution towards a less energy dependent society, based on sustainable energy sources, proves to take much more time than envisaged by the Kyoto-protocol. In these perspectives, CO_2-sequestration is a viable and safe alternative, and some techniques, such as aquifer storage or sequestration in coal mines, may be ready for use on short term. However, sequestration may only become truly important when deep reductions have to be achieved over the next decades.

GAS STORAGE IN COAL MINES

CO_2-sequestration in coal mines is inspired by storage projects for natural gas in abandoned coal mines. These reservoirs can be considered as extremely long deviated boreholes with a large contact surface with the coal (fig. 1), and have been used successfully since 1961 to store natural gas consisting mainly of CH_4. The oldest reservoir is located near Denver (Colorado, U.S.A.) and is still in operation today. The Leyden coal mine was in operation from 1903 until 1950, during which 5.4 Mton of sub-bituminous coal was extracted from two horizontal seams, located at depths of 210 and 225 m in the upper Cretaceous Laramie formation. Four shafts provided access to the underground workings. The cap rocks consist of 20 m of claystones, but the leak-off pressure of the reservoir is determined by shafts. These can withstand 1.8 MPa or about 75 % of the hydrostatic pressure.

Two other reservoirs, the abandoned coal mines of Anderlues and Péronnes, are located in the gassy Hainaut coalfield in southern Belgium. Both are characterised by a very low reservoir pressure. Anderlues will be discussed here because most publicly released data refer to this site. 25 Mton of coal was exploited at the Anderlues colliery between 1857 and 1969, there is evidence of small-underground mining activity before 1857. After closure, the gas drainage facility was maintained. The site was used for commercial gas storage from 1980 on, but gas injection was stopped in 2000 because of the need for costly maintenance work at the shafts. Modern exploitation levels were located between 660 and 1100 m depth, but there exist connections with flooded shallow workings and the overburden is locally only 10 m thick. The base of this overburden consists of clay, marl and chalk. This determines the reservoir pressure of 0.35 MPa. At these very low pressures adsorption of CH_4 on coal is relatively important. At Anderlues it is estimated that about 160 Mm³ is adsorbed on coal, or about 8 times more than the 20 Mm³ of free gas.

FROM STORAGE OF NATURAL GAS TOWARDS SEQUESTRATION OF CO_2

The storage of natural gas in coal mines has demonstrated the technical feasibility to use abandoned coal mines as reservoirs. Due to the explosive nature of natural gas, safety regulations need to be very strict. Keeping the maximum pressure below hydrostatic will, in case of disperse leakage across the top seal, result in a downward infiltration of water in the reservoir instead of an uncontrolled escape of gas to the surface. An under-pressured reservoir usually requires that the reservoir is kept dry by pumping. Since suited sites are also selected on their reservoir tightness, the rates of flooding will be very low. For Anderlues for example, water evacuation from the main part of the reservoir, located between 600 and 1000 m, was not necessary during the time span of over 30 years after termination of mining activities, this in spite of the extremely low reservoir pressure.

Whereas the leeway of water infiltration provides additional safety for natural gas storage sites, it could jeopardise a sequestration project. This is because reservoirs used for sequestration cannot be pumped dry for the duration of the targeted sequestration term, which may well be a thousand years or more. If a natural storage site would simply be converted for CO_2-sequestration, then the reservoir conditions can initially indeed be set according to safety regulations. With time however, the level of mine water would rise due to infiltration of formation water. This will compress CO_2 at the top of the reservoir. Eventually, pressure will exceed hydrostatic levels and CO_2 may leak towards the biosphere in case the top seal fails to withstand such pressure differences. The importance of this can be demonstrated with the Beringen mine, which will be discussed later on. 1.7 Mton of CO_2 can be stored in this mine without exceeding 80 % of hydrostatic pressure at the top of the reservoir, as long as the mine is kept dry. When pumping would be stopped, then reservoir pressure would run up to 120 % of the hydrostatic pressure due to flooding of the larger part of the mine (simulation with CO_2-VR, [3]). Fortunately, the Beringen mine should be able to handle these pressures.

It is therefore critical to a sequestration project that the leak-off pressure of a coal mine can be estimated prior to the start of the project, and that it proves to be well above hydrostatic. Safety risks are freezing and asphyxiation. The former results from the vaporisation, and concurrent cooling, of liquid CO_2 and is associated with leakage from pipelines and tanks located close to the surface. More relevant is asphyxiation that may occur when large amounts of CO_2 would suddenly escape into the atmosphere. High concentrations of CO_2 may temporarily build up close to the surface because it is denser than air. A slow leakage of CO_2 does not pose a direct threat to humans or nature because it is not toxic. However, it should be limited in order to not violate the whole concept of CO_2-sequestration (< 0.1 % per year for sequestration times over 1000 year, [4]).

STORAGE MODES FOR CO_2

Coal mine reservoirs can be regarded as very permeable and therefore isolated structures that are largely dry when injection starts. CO_2 will mainly be stored as pure CO_2 in the dry residual space, in solution in the formation water or adsorbed to coal.

Residual space, additional residual space and accessible coal

Residual space is the mined-out volume that remains after collapse and subsidence. The mined-out volume can easily be estimated from the amount of exploited coal and its density, augmented with the volume of extracted host rock. The volume lost due to collapse and subsidence is more difficult to estimate. It can be calculated from the total subsidence, if accurate data are available, or with a site-specific ratio between mined-out volume and residual space, depending of geology, mining and back-filling techniques. The latter approach requires data from drill cores or other in situ measurements.

Additional residual space is reservoir volume that is created due to an elevated reservoir pressure. In most cases this is due to the restoration of the hydrostatic pressure when an abandoned mine becomes flooded. The surface expression of this process is ground-level uplift. Ground-level recovery is slow and uniform compared to subsidence and therefore does not result in damage. Although the surface effect is usually very small, a few centimetres or decimetres, it may correspond to a considerable increase of the reservoir size. The residual and additional residual spaces add up to the total reservoir volume that will be occupied either by pure CO_2 or by CO_2 in solution in mine water.

CO_2 will also be readily adsorbed on coal. The reserves of coal remaining when the mine was closed are usually known, but it is largely a matter of common-sense judgement which part of these reserves will actively attribute to the storage capacity. CO_2 will, for example, hardly or not migrate to coal reserves below the lowest exploitation level, whereas thin seams left as non-mined gob or in highly fractured parts will be readily accessible. It is however difficult to formulate general rules. At the Leyden facility, for example, adsorption on coal is relatively unimportant, whereas at Anderlues it attributes largely to the storage capacity, even to seasonal movements of gas.

Pure CO_2

The part of a coal mine reservoir that is not flooded can be occupied by nearly pure CO_2. How much can be sequestrated in this way depends on the density of CO_2 and the non-flooded residual volume. The density is determined by reservoir temperature and pressure, and can be calculated from a suited equation of state (e.g. [5]). When a reservoir is not maintained by pumping, as is the case for reservoirs that will be used for CO_2-sequestration, then only those parts of the reservoir where the pressure is higher than hydrostatic will remain dry. These conditions can be calculated with use of a reservoir simulator [3]. In general, a reservoir needs to be deeper than 500 m to obtain sufficient high densities at reasonable reservoir pressures (not exceeding 130 % of hydrostatic). The sequestration capacity of the shallow Leyden coal mines, for example, is no more than 126 000 ton of CO_2, which is far too low to be of any significance for CO_2-mitigation.

CO_2 in solution

CO_2 has a relatively high solubility in water, and therefore the flooded parts of a mine may contribute to the total sequestration capacity. However, solution is much less efficient than storing pure CO_2. At normal reservoir conditions, less than 10 % of the CO_2 that can be stored in dry space, can be stored in the same volume occupied by mine water. It are therefore only the deepest parts of very voluminous mines that may significantly contribute to the sequestration capacity. The solution capacity of the whole Anderlues coal mine, for example, is about 0.5 Mton of CO_2.

Adsorption

Adsorption is the preferential partitioning of substances from the gaseous or liquid phase onto the surface of a solid substrate. It is the result of van der Waals forces, acting between an adsorbate, such as CH_4 or CO_2, and an adsorbent, such as coal. Since the total internal surface of coal is high due to its micro-porous structure, adsorbed amounts are generally significant.

The equations of state for pure CO_2 and the solubility of CO_2 in water are sufficiently accurate and reliable. Those describing the adsorption of CO_2 on natural coal however are still under construction, and therefore the simulations of CO_2-VR are based on the adsorption behaviour of active coal [3]. This introduces significant errors when estimating the total amount that can be sequestrated in a coal mine, as the adsorption capacity may exceed 50 % of the total sequestration capacity.

Assessing the storage capacity

The total sequestration capacity is the sum of CO_2 as a pure fluid, in solution in mine water, and adsorbed to coal. Calculation of these sub-results is not always evident when accurate data is lacking. Most recent mines, for example, will have detailed mine reports that may allow estimating the current residual volume, but when such a mine is abandoned no measurements will be available to estimate the additional residual volume that will be created by pressurising the reservoir. This uncertainty can be dealt with by splitting the estimates into an ascertained capacity that relates to the residual volume, and a potential, not yet verified capacity that would result from the additional residual volume (fig. 2).

Another significant uncertainty in the calculation of the adsorption storage is usually the amount of coal that is available for adsorption. The remaining reserves are generally well known, and are often considerable. However, not all of this coal will be sufficiently accessible to adsorb the injected CO_2. A rough, but robust approach is splitting of coal that is likely to be inaccessible to CO_2, for example, coal seams located below the deepest exploitation levels. The remainder can then again be divided into coal that is present in fractured zones close to mined levels, in gob or in pillars. Migration of CO_2 from the reservoir to these parts of the coal reserves is likely, and thus they constitute an ascertained adsorption capacity. Migration to less affected coal seams is also possible, especially when given enough time, but not certain. They therefore provide a potential adsorption capacity.

This approach splits each of the three modes of sequestration into an ascertained and a potential sequestration capacity. Assessment studies can be based on the ascertained part. If this is large enough to set up a meaningful CO_2-sequestration project, then it can be designed for the ascertained capacity, but flexible enough to include the potential capacity.

CASE STUDY: CO_2-SEQUESTRATION AT THE BERINGEN COLLIERY (BELGIUM)

Environmental concerns

An estimated 500 000 l of leaked gasoil, a few thousand transmission boxes containing PCB's and important quantities of organic waste were left underground in all 7 Campine collieries after closure. This anthropogenic pollution adds to the salinity of the water and to the CH_4 dominated gases compressed above the water table. Analyses at gas-pipes indicate that the composition of the mine gas must be above 80 % methane, the remainder being mainly nitrogen and CO_2. As pressure builds up further, this gas may migrate towards the overlying aquifers, and as the mines become completely flooded, other dissolved contaminants may migrate upwards as well.

Belgium, as a member of the European Union, has ratified the Kyoto protocol in 2002, meaning that it has to reduce anthropogenic CO_2 emissions by 7.5 % with respect to 1990 in 2008-2012. Although Kyoto represents just a fraction of the total effort required to stabilise the atmospheric CO_2, many countries, including Belgium, are facing problems in trying to meet their mitigation targets. In Belgium, CO_2-emissions have only increased since 1990. Policy measures, such as the rational use of energy and a shift towards renewable energy and decrease of the carbon intensity, are not sufficiently to meet the Kyoto targets. The not yet accepted policy of geological sequestration remains therefore a necessary option. In Belgium storage space is limited to saline aquifers, coal deposits and abandoned coal mines. Most abandoned coal mines, including those that are used for the storage of natural gas, do not meet the strict specifications for CO_2-sequestration. However, suitable coal mines may be regarded as candidates for early opportunity demonstration projects. Such a mine is the Belgian Beringen colliery, located in the Campine basin.

The Campine basin

The Campine coal basin is situated in the North of Belgium, continuing into the Netherlands and Germany. It is completely concealed, with mined Carboniferous coal deposits between depths of 500 and 1000 m. The first Campine coal mine opened in 1917, the last of 7 mines was closed in 1992. After closure, the mine shafts were sealed with concrete and filled to the surface. No leakage along the former shafts has been observed. No monitoring facilities were installed other than two gas-pipes at one colliery that were left open between 1988 and 1990 [6]. This results in uncertainty concerning the current condition of the mines. Based on comparison with Dutch mines and the hydrogeology of the Campine basin, it is assumed that only the deepest parts of the mines are currently flooded (average rise of water levels estimated at less than 20 m/y).

The temperature gradient in the Campine basin is 0.035 °C/m for the upper 1000 m [7]. The average reference temperature in Belgium has been set at 9.8 °C, but may have raised by 0.5 °C during the last 50 years. Groundwater temperatures of 48 °C have been measured at the deepest mine levels [8].

Sealing of the shafts

Access to the coal deposit was limited to two vertical shafts for each colliery (average internal diameter 4,80 – 6,20 m). Shafts were lined with metal tubing in the overburden (place with the freezing method) and concrete rings anchored in the rock mass in the coal measures, all protected against deformation and water seepage. Nevertheless all shafts experienced some leakage from shallow aquifers in the overburden, for the Beringen I shaft this even amounted to 52,5 m^3/day.

In the period 1988-1994, shafts were sealed following a programme identical for all mines [9, 10]. First, all constructions, pipes and cables left inside the shafts to be filled were removed. The bottom of the seal was at the base of the highest working level, eventually the base of the shaft. Above an existing workfloor at this level or the base of the shafts, concrete was poured in 10 metre steps (resistance factor to corrosion B15 – DIN 1045). Connections to underground workings or side-galleries were dammed at some metres or decametres from the shaft circumference, wherever a suitable place could be found for anchoring the dam. Concrete filling extended over a vertical elevation of 100-200 m, close to the top of the coal measures. Friction induced by the shaft's concrete wall rugosity and extensions into the side-galleries kept the concrete in place (load factor 335-500 MN). From the transition with the overburden and the lining with metal tubing onwards the shaft was filled with stabilised sand mixed with ash particles finer than 250 µ (resistance to compression β_{W28}>2 MN/m^2). In the overburden, 30 m thick clay barriers ($k<10^{-9}$ m/s) were placed at the level of natural aquicludes : the Vaals marly sand near the base of the Cretaceous, covering the saline Upper Carboniferous aquifer, the Waterschei clay near the base of the Tertiary, covering the major confined Maastrichtian calcarenite aquifer, and the Boom clay towards the middle of the Tertiary, separating the semi-phreatic aquifer in the Neogene sands from local confined aquifers in Lower Tertiary sands. The claystops were protected by 10 m thick concrete slabs (resistance factor B02 – DIN 1045). The top 6 m of the shaft filling was completed by a slab of concrete covered by clay. The shaft's onset is covered by a plate of armoured concrete. A chimney through this cover allows for extra filling in case of compaction or displacement of the fill.

The Beringen colliery

The Beringen colliery is a typical Campine coal mine. 79 Mton of medium to high volatile A bituminous coal was mined. The thickness of the overburden varies from 570 m to 665 m. The main stone drifts are located between 660 and 842 m depth [11]. During exploitation two connecting galleries were driven to the neighbouring colliery, and probably limited communication still exists. At each mine, communication with the surface was limited to two shafts that are completely sealed.

The permeability of the main connecting galleries is set at 1000 darcy. The porosity and permeability of the shales unaffected by mining or mining subsidence is considered to be zero [11, 12]. The marls from the overlying Cretaceous have a low to very low permeability, and form the primary seal for the reservoir succeeded by several clay units functioning as secondary seals. The sealing quality of the marl is proven by a century-long mining activity when coal measures were completely dewatered and the marl served as a protection against overlying groundwater.

Reservoir capacity

The mined-out volume of Beringen was calculated from the data of [11] and is estimated at about 85 Mm^3. Taking into account a residual volume fraction of 7 %, which was derived form published data on mines in the neighbouring Limburg, Campine and Ruhr areas (tab. 1), this results in a residual volume of about 5.9 Mm^3. To take maximum benefit of this volume, the pressure should be high enough to prevent water infiltration into the main part of the reservoir. For the mine of Beringen, a reservoir pressure of close to 130 % of hydrostatic pressure at the top of the reservoir would prevent flooding of the main stone drifts (fig. 3). These pressure conditions are realistic for aquifer storage, but it remains to be verified if even the very well sealed shafts and sedimentary cover of the Campine coal mines will hold. At these conditions about 1.7 Mton of CO_2 can be stored in the dry parts, and an extra 0.15 Mton in solution in the mine water.

Ground movements in the adjacent coal field of South Limburg (The Netherlands) have been monitored in relation to rising mine water. Currently, rises of the surface of 22.5 cm have been reported. The maximum surface recovery is estimated at between 25 and 30 cm, which is between 3 and 5 % of the original subsidence. This effect is attributed to the restoration of hydrostatic pressures in the subsurface, which decompacts the fractured zones around mined out levels [13]. The recovery of the overburden is neglected in the models. This may be a valid assumption, since there is no noticeable difference in the uplift curve before and after rise of the mine water level through the top of the Carboniferous. Starting from the profile of [13, their figure 5] an average value of 15 cm was calculated for final recovery of the subsided zone. Subsidence values of the South Limburg coal field are comparable to values in the Campine Basin. Since subsidence and recovery are closely related, similar recovery values may be expected in both areas. The recovery height is proportional to the pore-fluid pressure (reservoir pressure). A reservoir pressure 30 % higher than the hydrostatic pressure will therefore result in 30 % more additional residual space than resulting from ground water level restoration. An average uplift of only around 20 cm will create additional space that is equal to the current residual volume. The size of the reservoir, and its storage capacity for pure CO_2 and CO_2 in solution, may therefore be doubled and attain 12 Mm^3 (3.7 Mton of CO_2, see fig. 3).

As larger part of the reserves are located deeper than the deepest mined levels, or are located in parts of the concession where no coals have been extracted, only 50 % of the 80 10^6 ton of the proven reserves is considered to form part of the reservoir. These are tentatively divided into 30 % ascertained capacity and an additional potential of 20 %. This would ascertain the storage of about 0.8 Mton of CO_2 by adsorption. In an optimistic scenario this capacity could be increased to 1.3 Mton (fig. 3).

SEQUESTRATION CAPACITY

Summarising the different modes of sequestration, the ascertained sequestration capacity is around 2.7 Mton CO_2, with a potential to sequestrate a total of 5 Mton. When combined with the neighbouring and probably interconnected collieries, the combined ascertained capacity would be about 7 Mton CO_2, extendable to possibly 13 Mton. This is sufficient to sequestrate between 300 000 and 500 000 ton/y for 25 years. Such rates are relatively moderate when compared to e.g. the Sleipner project where about 1 Mton of CO_2 is injected on yearly basis, but still significant compared to the 9 Mton of CO_2-emissions that has to be avoided in order to meet the Belgian Kyoto target.

CONCLUSION

Three plants for the storage of natural gas in abandoned coal mines have proven that these reservoirs are safe as temporary gas reservoirs. CO_2-sequestration, however, poses farther-reaching demands. In order to obtain a safe and stable reservoir with sufficient capacity, the top of the mine should be at least 500 m deep, possess well-sealed shafts, be overlain by a tight cap rock and be mostly dry. Only few mines will currently meet these requirements, although in future new mines may be designed to allow CO_2-sequestration after abandonment.

Coal mine sequestration may become important since there may be additional economic and environmental gains. These may come from the extraction of methane prior to sequestration, the enhanced coal bed methane recovery, the development of a CO_2-geothermal project [14], and the proximity of industrial CO_2 producers to many coal basins.

The technical set-up of a coal mine sequestration project is relatively simple and CO_2 injection rates can be high. The main uncertainty to verify is the sealing property of shaft-fills and cap rocks. Suitable coal mines may be prime candidates for early opportunity projects, involving injection of process CO_2, captured at low cost, into nearby collieries.

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TABLES

	Residual volume fraction (%)	***Remarks***	***Author***
Limburg-Campine-Ruhr coal mines (7 ± 3) %	*5-10 %*	Ruhr basin. Collapsed panels and infrastructure. Depth > 800 m.	Kunz, 2000
	5 %	(Campine) Belgian coal mines. Back-filled areas.	Labasse, 1965
	4.5 %	The Netherlands, Limburg, abandoned panel from Maurits mine. 40 % back-filled, 60 % collapsed. Depth ~500 m. Value recalculated from original data.	Berding, 1952
	8 %	Campine coal mines. Collapsed panels.	van Tongeren & Laenen, 2001
	5.5 %	Campine coal mines. Back-filled panels.	van Tongeren & Laenen, 2001

Tab. 1:

Estimates of the residual volume fraction of coal mines from different authors, and the proposed value used in further calculations (modified from [11]).

FIGURES

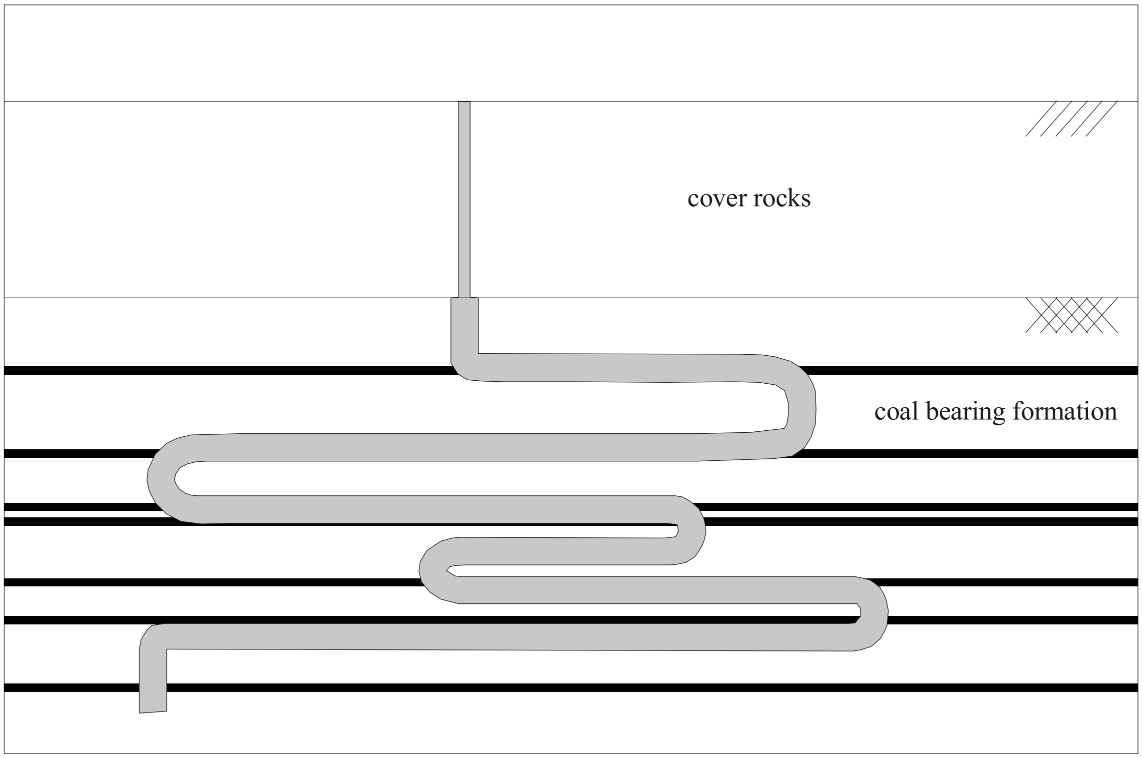

Figure 1:

A coal mine reservoir can be considered as a very long deviated borehole in a coal seam as shown by this cartoon. Uncollapsed galleries provide high-permeable pathways for rapid injection of CO_2, a major difference with CO_2 injection in unmined coal. The large contact surface favours additional storage space due to adsorption on coal.

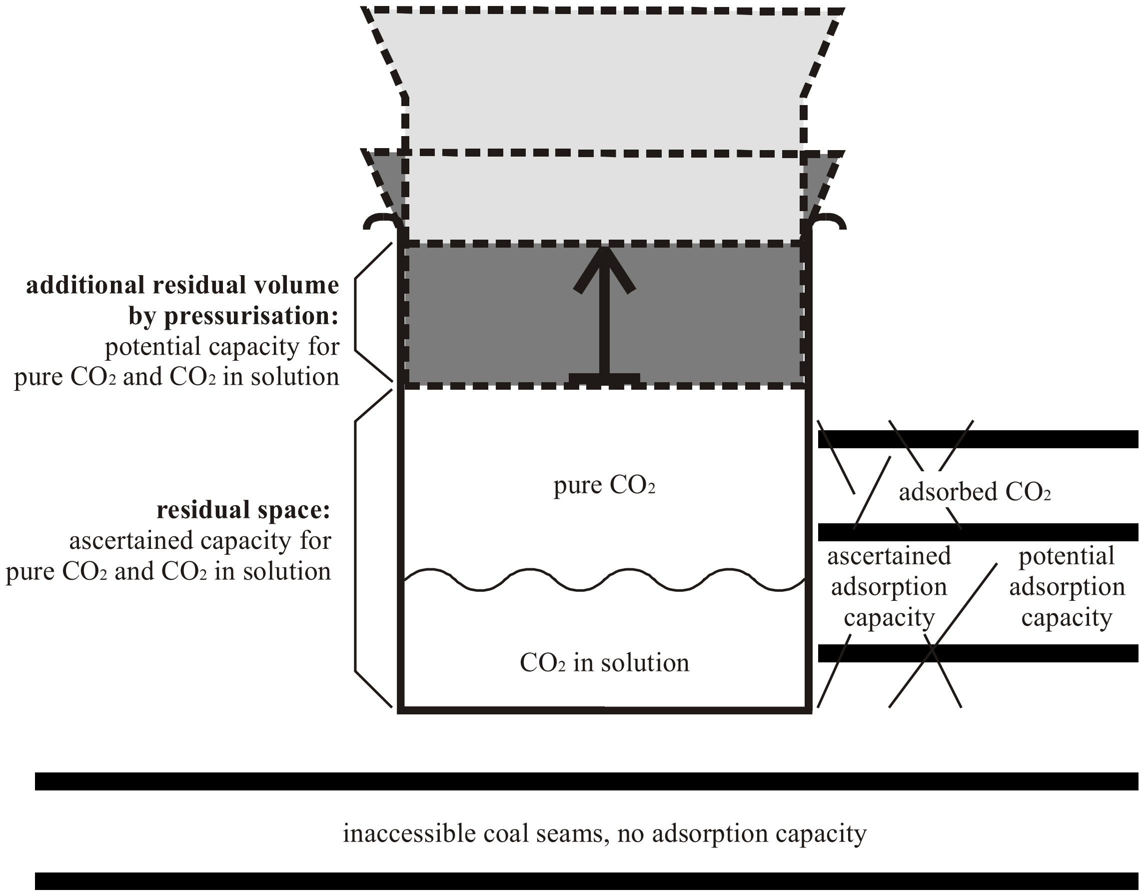

Figure 2:

Schematic overview of the three different storage modes of CO_2 in a coal mine reservoir, and the subdivision into additional and ascertained reservoir space.

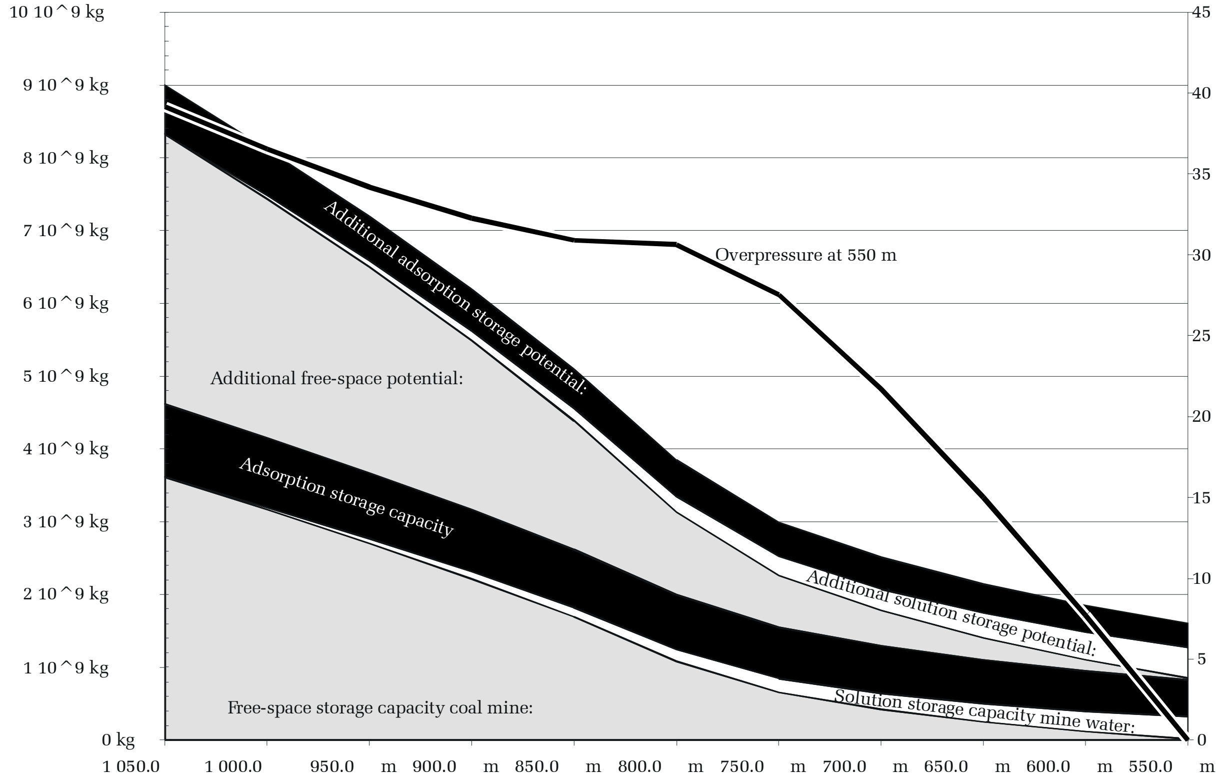

Figure 3:

The sequestration capacity of the Beringen colliery for different amounts of overpressure on the top seal. Calculation with CO_2-VR. X-axis: depth of the water table; left Y-axis: amount of sequestrated CO_2; right Y-axis: amount of overpressure at the top of the reservoir (550 m), in percentage relative to the hydrostatic pressure (30 % is 130 % of hydrostatic pressure).