

FEASIBILITY OF CO₂ SEQUESTRATION IN ABANDONED COAL MINES IN BELGIUM

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(9 figures and 3 tables)

ABSTRACT. The concept of storing gas in abandoned coal mines is proven technology (Anderlues, Péronnes). However, this was done for seasonal storage of natural gas in underpressured reservoirs, whereas CO₂ sequestration preferably requires overpressured reservoir conditions for long-term storage. Marl and chalk apparently form a sufficiently tight seal in a number of coal mines, but this will need verification. Safety risks involved with the storage of CO₂ in mines are low. Natural faults and induced fractures need detailed evaluation, especially with respect to changes in the stress-regime.

Standard techniques to inject CO₂ into reservoirs involve the injection of liquid CO₂. Excessive cooling of the reservoir and injection equipment can be avoided by depressurising CO₂ and injecting it as a gas or, when the reservoir becomes partly fluid-filled, to apply custom-designed injection schemes for injection of liquid CO₂.

When CO₂ is injected into a coal mine, it may be stored in free space, in solution in the formation water, or adsorbed in coal. These amounts can be calculated, but not all parameters are sufficiently known to produce reliable results. Therefore, the amounts are split into an ascertained capacity that can be calculated with a known accuracy, and an additional capacity, which is an estimated surplus that cannot be calculated accurately.

CO₂ sequestration in coal mines seems to be technically feasible and may be a useful option for Belgium to reduce the industrial emission of greenhouse gases into the atmosphere. A case study for the Beringen-Zolder-Houthalen collieries shows that sequestration is a viable option, even for the ascertained capacity alone. At an injection rate of 300 000 tons/y, sequestration can be operational for about 25 years. This is a conservative estimate, and it may prove possible to inject at 500 000 tons/y for 25 years. This could be a considerable contribution, approximately 3 to 6% of the mitigation required to reach the Kyoto target for Belgium. Petrochemical plants that produce nearly pure CO₂ streams are prime candidates for early opportunity projects in abandoned coal mines.

Keywords: CO₂ sequestration, abandoned coal mines, Belgium, Campine basin, Hainaut coal basin.

1. Introduction

Predictions indicate that the technical and social evolution towards a less energy-dependent society, based on sustainable energy sources, will take more time than envisaged by the Kyoto-protocol. According to these perspectives, CO₂ sequestration is a viable and safe alternative, that is ready for use on short term. Currently, storage in depleted oil or gas fields, mostly in combination with enhanced oil recovery, or storage in saline aquifers, is proven technology. CO₂-ECBM (enhanced coal bed methane recovery with CO₂-injection) is in an early stage of development (CO₂NET, 2002), and may not be feasible in the near future outside the San Juan basin in the USA (4 wells in the Allison unit have injected 166 000 tons/year for enhanced methane recovery). The recovery of CH₄ is sufficiently known by commercial practice, but the injectivity and behaviour of CO₂ are less known. Until now, sequestration in abandoned coal mines has not been considered. Studies in the GESTCO-framework concentrated on the geometry and residual space left by

underground mining (Piessens & Dusar, 2003a-c; van Tongeren & Laenen, 2001; van Tongeren & Dreesen, this volume). These studies show that:

1. Gas storage in abandoned coal mines is proven technology.
2. Technical barriers and uncertainties for CO₂ sequestration in coal mines are limited.
3. The sequestration potential is substantial at the local scale, taken into account that the majority of coal mines are probably not suited for CO₂ storage.
4. CO₂ sequestration can be incorporated in mine closure programmes.
5. Opportunities are created for environmentally friendly energy production.

The latter point may be of specific interest regarding the expected public reservation towards underground storage of CO₂. A key factor is the possibly widespread conviction that waste prevention is to be preferred over "dumping". If CO₂ sequestration leads to new opportunities to produce environmentally friendly energy, such

as enhanced coal mine methane, enhanced coal bed methane, or geothermal energy, then CO₂ sequestration would come down to CO₂ recycling (Piessens & Dusar, 2003a). The technological difficulties of sequestration in coal mines are no more substantial than those of sequestration in depleted oil fields and aquifers. On the other hand, public acceptance may be more readily gained for mine sequestration in view of the environmental benefits it potentially offers. It is therefore proposed to consider sequestration in coal mines as first-line projects, which may be used to assist meeting the emission reductions of 2008-2012, and can lead to a more general acceptance of underground-storage of CO₂. A third potential obstacle is the current cost of capturing CO₂, but this may be countered by using pure CO₂ that is produced by the chemical industry in sufficient quantities for 'early opportunities' demonstration projects.

2. State of the art: CH₄-storage in abandoned coal mines

Abandoned coal mines are and have been used successfully to store natural gas (mainly CH₄) since 1961. Two of these are located in Belgium (Péronnes and Anderlues, between Mons and Charleroi, Fig. 1; Cajot, 1979; Dusar & Lagneau, 1991; Dusar & Verkaeren, 1992; Houtrelle,

1999), and a third is in the U.S.A. (Leyden coal mine near Denver, Colorado; EPA, 1998; Schultz, 1998; Collings et al., 2002). These projects have been set up to meet peak demands and to avoid peak pricing. Although storage of gas is expensive and avoided where possible, the Leyden coal mine has been positively evaluated, leading to an expansion of its capacity. The site at Péronnes has been abandoned after an increase in local taxes, and the activities at Anderlues are currently being terminated instead of upgrading the underground facilities. All three projects are pilot projects and have faced some unforeseen problems (sealing of shafts and production wells, flooding, compositional and pressure changes due to adsorption and desorption). These could all be solved so that storage in coal mines has become as safe as it is in classical storage facilities (aquifers, depleted oil fields and salt caverns, cf. Association technique de l'Industrie du Gaz en France, 1990). In the few projects realised so far, the initial costs for sealing the mine workings used for storage were higher than expected. The storage volume is relatively small because of the shallow depth of the mines compared to hydrocarbon reservoirs (EPA, 1998).

Many more abandoned coal mines, especially those that also provide opportunities for gas drainage, are potential candidates for CO₂ storage. However, when considering true sequestration of CO₂, a reservoir should meet stricter specifications. The reservoirs in the existing

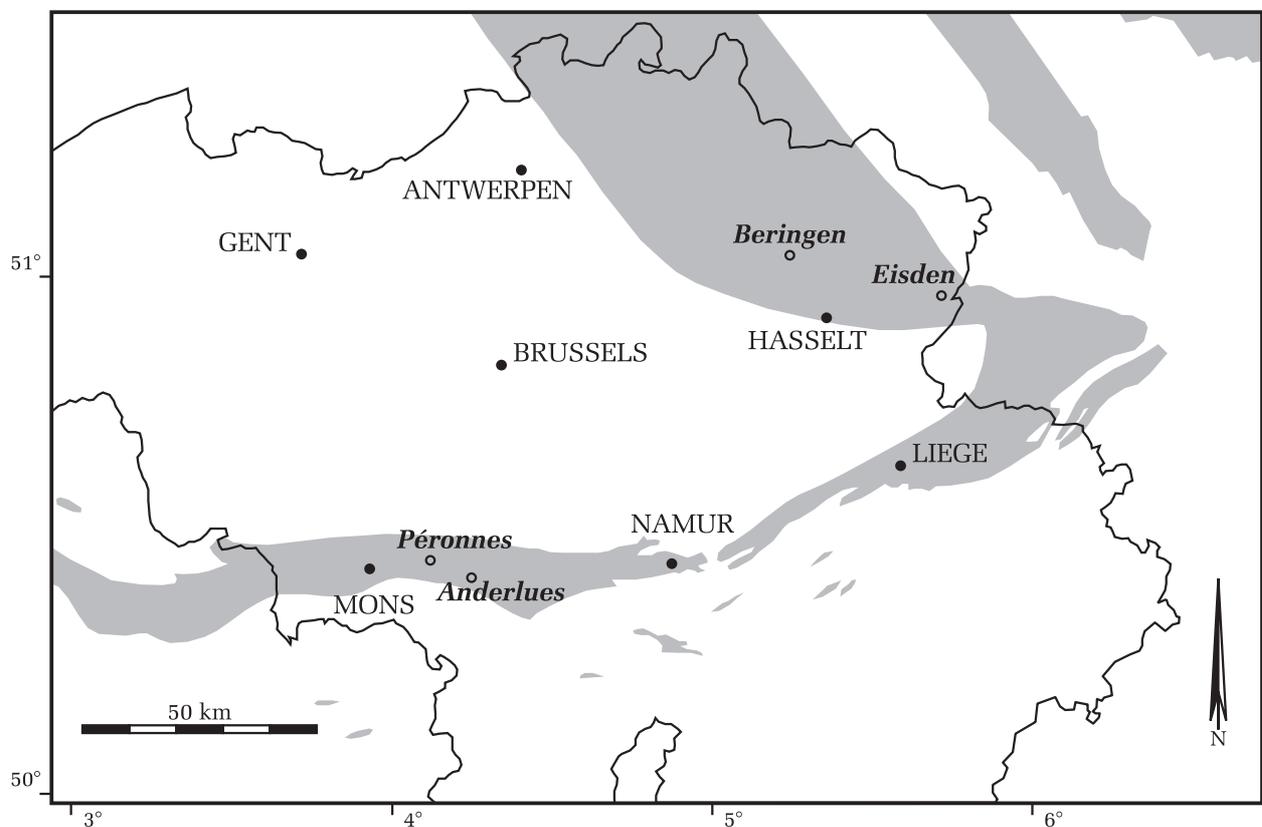


Figure 1. Map of the outcropping or shallow subsurface coal basins (shaded area) in and around Belgium. Open circles are the location of the mines that are mentioned in the text, filled circles are major cities. After Renier (1954).

storage sites for example, have only been filled to a maximum pressure of 80% of the hydrostatic pressure. This implicates that measures must be taken to prevent influx of ground water. In order to acquire a stable, long-term maintenance-free reservoir, pressure should be raised to the regionally stable pressure conditions (usually hydrostatic pressure) or higher.

2.1. The concept of storing CH₄ in coal mines

The basic conditions for storage of natural gas (mainly composed of CH₄) in coal mines are (Houtrelle, 1999):

1. There should be no lateral communication with other mines, because this would allow the gas to migrate to places where it becomes difficult to retrieve.
2. There should be no communication between the mine reservoir and the surface, as this would allow gas to escape to the atmosphere.
3. The amount of influx of water into the mine is preferably low, as the mine has to be kept dry by pumping for storage of CH₄.

2.2. CH₄-storage in Belgian coal mines

2.2.1. Anderlues coal mine

The coal mine of Anderlues or former Bois-de-la-Haye colliery in the Hainaut coal field forms part of the Beau-leusart concession, with a total surface area of 20,89 km². The colliery was in operation between 1857 and 1969 (cumulative coal production 25 Mtons). Small-scale underground mining occurred before 1857 but is not documented. After closure, the gas drainage facility was maintained (peak production rate 18 Mm³/y at 92% CH₄). Research activities were started in 1971 by Distrigaz to evaluate the possibility of developing a natural gas reservoir. Injection tests began in 1976, and commercial use began after 1980.

The average thickness of the post-Carboniferous overburden is about 50 m, but varies significantly from south to north and is locally only 10 m. The Carboniferous coal measures were mined between depths of 120 m and 1100 m. Older mine workings down to 600 m may be

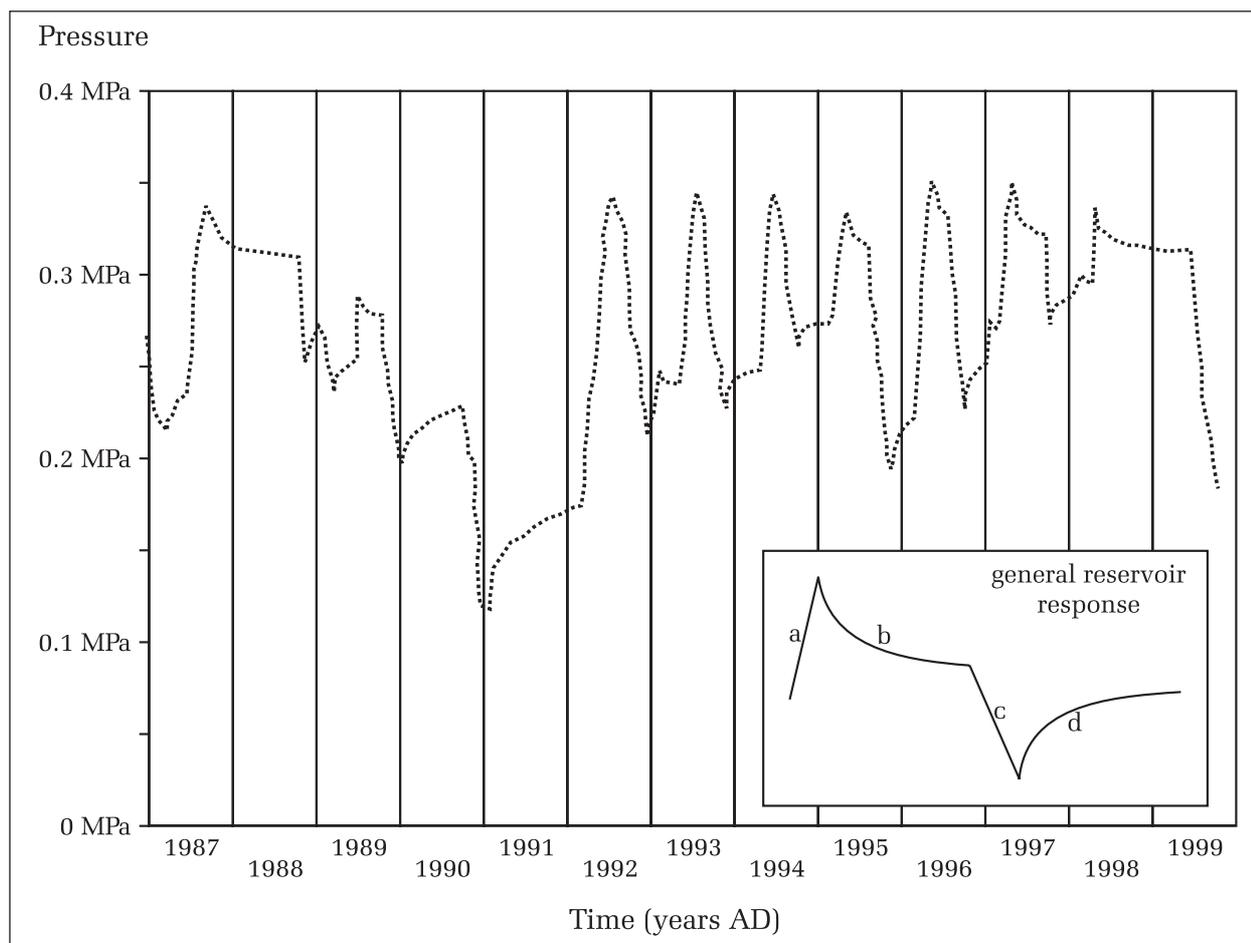


Figure 2. Reservoir pressure of the Anderlues storage facilities from 1987 to 1999 (data by Distrigaz, after Houtrelle, 1999). The inset shows the relation of reservoir pressure to injection-production. (a) Injection resulting in a steep increase of pressure. (b) After injection has stopped, the reservoir pressure decreases gradually as CH₄ becomes adsorbed in coal. (c) Extraction results in a sharp decrease of reservoir pressure. (d) After extraction, the reservoir pressure increases again as CH₄ becomes desorbed from coal. Note that the reservoir response can be predicted (simulated versus measured curve).

partly flooded, whereas the more recent, deeper exploitation levels between 600 and 1100 m are considered dry. Gas thus is mostly injected in the levels below 600 m. The shallow workings are isolated from the deeper by a thrust fault that acts as a primary hydrogeological barrier. This thrust fault is only traversed by vertical shafts that do not drain the shallower aquifers. All surface shafts are sealed at a depth of 60 m, within the impervious zone encompassing the contact between the coal measures and the overburden.

The sealing rocks covering the reservoir are water-saturated. At the reservoir pressures used (maximum 0.35 MPa), no gas would escape to the surface because leaking gas would be dissolved in the aquifer. Nine piezometric wells were drilled in the overburden aquifer, covering the presumed storage area to verify the security of gas containment. Piezometric levels do not show any relation to pressure changes due to injection or production of gas in the reservoir. No gas from the storage reservoir has ever been detected in the piezometers.

The storage facility at Anderlues operates at very low pressure. The reservoir volume below the thrust fault at 600 m depth is estimated by the operator between 6 and 10 Mm³ (mainly based on regional assessments of accessible parts of old mine workings). This allows for the storage of about 20 Mm³ of CH₄ (14 000 tons) at working reservoir pressures. This implies that almost 8 times more CH₄ is adsorbed on coal since the total storage capacity is 180 Mm³ or 130 000 tons CH₄. This is due to the fact that at low-pressure adsorption is important relative to compressibility. The effects of adsorption are evident from the injection-production diagrams (Fig. 2).

The main technical challenge for developing Anderlues and Péronnes as natural gas storage facilities was the sealing of all older shafts. This was accomplished by constructing working shafts close to each existing shaft and using these to access the old shafts at a depth of 60 m and to seal the two shafts with reinforced concrete, after freezing and removal of rubble used as shaft fill material. Initial investments were considerable because of the discovery of additional, undocumented shafts.

Injection at Anderlues ceased in 2000 because of the need for costly maintenance work at the sealed shafts. Although underground storage at Anderlues was profitable, storage cost is about 5 times higher than at Loenhout, an aquifer storage facility in the Campine basin of northern Belgium, operated by the same company. This is mainly due to the slightly different calorific content of the produced gas (heavier hydrocarbons remain preferentially adsorbed on coal) necessitating the addition of 3% propane to the outflowing gas when the gas supply used for storage changed from 'lean' Groningen gas to 'rich' Norwegian and Algerian gas. This has almost doubled the operational cost. After extraction, the gas is recompressed to ~8 bar (0.8 MPa) for delivery to the local network.

2.2.2. Péronnes coal mine

The coal mine of Péronnes is located in the same area as the Anderlues colliery and is quite comparable in setting, operation and production rates. It belongs to the Charbonnage du Centre concession grouping of the Ressaix, Mariemont and La Louvière collieries and was exploited from around 1860 until 1969. Nineteen shafts were sealed between 1978 and 1982 to allow for underground storage of CH₄. This is considerably more than was originally estimated.

As for the adjoining Anderlues coal field, the geology of the Carboniferous strata is relatively complex. It is subdivided into a shallow part, the Massif de Masse, and the underlying Massif du Poirier. Both are separated at about 500 m depth by a tectonic thrust-fault zone of about 80 m thick. The latter is water-tight, and it is sufficient to control the level of water in the shallow Massif de Masse to prevent water from entering the dry Massif du Poirier through the limited number of vertical shafts that connect the two massifs. The gas was injected into the Massif du Poirier and retracted from the Massif de Masse. The overburden at Péronnes varies between 32 and 100 m, for mining depth varying between 60 and 1070 m.

The total capacity of Péronnes was about 120 Mm³ of CH₄ (85 000 tons). Injection operations have been terminated since 1996 due to an increase of local taxes on gas storage. The cost of storage was almost 60% higher than for the comparable Anderlues facility because of the more complex design involving injection in two shafts, and because of the need for recompression as pipeline gas (50 to 60 bar, or 5 to 6 MPa).

2.3. Comparison of CH₄-storage and CO₂-sequestration

The natural gas storage projects demonstrate the technical and economical feasibility of gas storage in coal mines. The sequestration of CO₂ gas in coal mines is similar but poses additional requirements. These mainly originate from the very long time for which storage must be guaranteed. Another item is the different physical and chemical properties of CO₂ and CH₄. At the envisaged pressures and temperatures CH₄ has a near ideal behaviour, whereas CO₂ will often be near the critical point. Also, CO₂ is generally better adsorbed on coal and is much more soluble in water. The latter is caused by the formation of the weak acid H₂CO₃.

3. Principles of CO₂ sequestration in coal mines

CO₂ sequestration in a coal mine can be considered as a special case of gas storage. The conditions for successful storage in mines also apply to sequestration (see §2.1):

1. There should be no communication between the mine reservoir and the surface, because this allows CO₂ to escape to drinking water aquifers and to the atmosphere.
2. There should be no lateral communication with other mines or aquifers, not used for CO₂ storage, because this makes long-term prediction of the migration of CO₂ (and possible escape to the surface) uncertain.
3. The amount of influx of water into the mine should be low, because an initially dry mine facilitates CO₂ sequestration.

The first of these conditions is the most critical and relates directly to the maximum reservoir pressure, or leak-off pressure, of the coal-mine reservoir. This in turn strongly determines the sequestration capacity. CO₂ may be sequestered in three states:

1. Free-space; this is as a gas, liquid or supercritical fluid.
2. In solution in mine water.
3. Adsorbed to the remaining coal in the mine.

The combination of these three storage mechanisms is typical of storage in coal mines (Piessens & Dusar, 2003b). Moreover, specific conditions have to be met in order to allow the stored CO₂ to be considered as sequestered (stored for a geologically long period of time).

1. Reservoir integrity has to be assured without reservoir maintenance. Monitoring, with the exception of initial follow-up, would not be necessary.
2. Reservoir simulation should include initial conditions, critical intermediate conditions, and equilibrium conditions. The reservoir should be found suitable in all three conditions.

3.1. Free-space storage

Free space refers to all voids in a coal mine that are accessible to injected CO₂. This definition corresponds to the absolute effective porosity of a coal mine reservoir, and includes workings that are still open (e.g. shafts or main galleries), and the remaining pore space in collapsed parts of the mine, including gob, backfill and the fractured zone above the original workings. In rare cases the residual space can be estimated directly from detailed excavation and subsidence data or from pressure tests for mines that are used for storage of natural gas. For most mines, this kind of data is lacking, and estimates have to be based on the extracted volume and the residual volume fraction. The final reservoir pressure and the resulting decompaction of the fractured zone around the mined out levels may result in additional reservoir volume.

3.1.1. Residual volume

Longwall mining has been the common excavation technique in most recent European coal mines. Panels were backfilled where subsidence was a reason for concern, or

were left unsupported resulting in caved 'goaf' panels. The initial free space in these zones, produced by coal and rock extraction, is reduced by mainly two processes:

1. Backfilling of panels, either by hydraulic means, material slinging, or pneumatic methods.
2. Subsidence of the unsupported roof (up to 30 or 90% of the original height of the front zone, even where back-filling has been applied).

Therefore, the residual volume of a panel is only a fraction of the extracted volume. Data on the residual volume fraction are limited, and estimates vary significantly (van Tongeren & Laenen, 2001). For the mines in the Limburg-Campine-Ruhr area, the residual volume is tentatively estimated to be an average of 7% of the extracted volume (Berding, 1952; Labasse, 1965; Kunz, 2000; van Tongeren & Laenen, 2001). For other European mines this volume may be higher, up to 20% (Malolepszy & Ostaficzuk, 1999; van Tongeren & Laenen, 2001).

Other workings including shafts and related infrastructure, stone-drifts and blind shafts may in many cases have largely remained open (a residual porosity fraction close to 100%) and are extremely permeable connections between parts of a mine. Other workings such as panel gate roads have collapsed or were backfilled, and are thus comparable to panels.

3.1.2. Ground-level uplift

Ground movements in Limburg (the Netherlands) have been monitored in relation to rising mine water after closure of the mines. 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 pressure in the subsurface, which decompacts the fractured zones around mined out levels (Pöttgens & van Herk, 2000). Subsidence values of the Netherlands Limburg area are comparable to values in the Campine Basin. Since subsidence and recovery are closely related, similar recovery values may be expected. Although the surface effects of recovery are very small, especially in comparison to subsidence, it has a relatively large effect on the reservoir.

Like subsidence, ground-level uplift is a response to volume changes in the subsurface. In theory, the volume change measured at the surface will be equal to the volume change at depth.

$$\begin{aligned} V_{\text{surface}} &= V_{\text{reservoir}} \\ h_{\text{surface}} \cdot S_{\text{surface}} &= V_{\text{reservoir}} \end{aligned}$$

where h_{surface} is the average ground-level uplift, S_{surface} is the area of ground-level uplift, and $V_{\text{reservoir}}$ is the additional reservoir volume resulting from the recovery effects. h_{surface} can be taken as equal to the predicted average values of Dutch Limburg (0.15 m), S_{surface} is equal to the area affected by subsidence. This additional residual space consists of effective porosity, since it is induced

by an increase of pore water pressure. The current total residual space of a nonflooded coal mine may therefore be considerably smaller than the residual space at final reservoir pressure if decompaction of the overburden as a source for ground-level uplift is negligible. The recovery height is proportional to the pore-fluid pressure (reservoir pressure). A reservoir pressure 10% higher than the hydrostatic pressure will therefore result in 10% more additional residual space (Pöttgens & van Herk, 2000). The effect of pore pressure on the reservoir volume, as predicted from theoretical models, needs to be confirmed by field evidence.

3.1.3. Density of CO₂

The density of CO₂, or in fact any fluid mixture, is determined by temperature and pressure, except at saturated conditions (both liquid and gaseous CO₂) when an additional parameter (volume or mass) needs to be defined. Equations of state for many gas mixtures have been formulated, but they are usually not accurate close to critical or saturated conditions. Since conditions in coal mines are often close to the critical point for CO₂ (P=7.4 MPa and T = 31.1 °C), the equation of state of Span & Wagner (1996) for pure CO₂ can be used. Effects of addition of CH₄ and H₂O can be evaluated through the changes of critical point and saturated domain.

For accurate calculations, it is not possible to use an average density for the whole reservoir. This is due to the phase or near-phase transitions that result from pressure and temperature gradients in a reservoir with a large vertical extent (several 100 m). Reservoir properties are calculated with "CO₂-VR", a custom designed MS Excel spreadsheet that simulates the behaviour of CO₂ in reservoirs with a large vertical extent (Piessens & Dusar, 2003c).

3.2. Adsorption on coal

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 CH₄ or CO₂, and an adsorbent, such as coal. Since the total internal surface area of coal is high due to its microporous structure, adsorbed amounts are generally significant. Active coal at room temperature and high pressure can adsorb up to 50% of its own weight. For natural coal samples this is usually 4 to 10 times lower.

The adsorption capacity of coal increases with increasing pressure and decreasing temperature. Note that under normal hydrostatic and geothermal conditions, pressure and temperature will both increase with depth. At shallow depth (0 to ~300 m), the effect of pressure will dominate and increase the adsorption capacity of coal, while at greater depth the maximum adsorption will slowly decrease due to increasing temperature (Yang &

Saunders, 1985). These effects are commonly illustrated with adsorption isotherms, which are conventionally modelled using the Langmuir adsorption equation. The effect of pressure increase is considerable at lower pressures, but will be minimal at higher pressures, because the isotherm is asymptotic to a maximum (saturated) value. As a rule of thumb, the molecular adsorption capacity for CO₂ can be considered to be about twice as high as for CH₄. However, some caution is needed when translating the adsorption characteristics for pure gases to exchange rates. Certain types of coal, especially among low-rank coals, will show preferential adsorption for CH₄ at lower pressure (Krooss et al., 2002; Busch et al., 2003), contrary to what may be expected from experiments with pure gases. This may make certain types of coal unsuited for CO₂ sequestration.

The microporosity, which is directly proportional to the internal surface area (surface area available for adsorption), the ash content (inorganic components), and the maceral composition, are the most important factors that cause scattering of adsorption capacity. At present, no general quantitative relationship exists that predicts the adsorption behaviour of coal in relation to these properties. However, local or regional values may be estimated based on coal maturity that are sufficiently accurate for feasibility studies (e.g. Carroll & Pashin, 2003).

Bed moisture reduces the adsorption capacity of coal. A reduction of 65% due to total saturation with water for a coal sample from the Beringen colliery has been reported by Haan (1998), Bertheux (2000) and Hamelinck et al. (2001). Isotherms determined for dry and wet coal samples determined by Krooss et al. (2002) indicate the same trend, but the amount of reduction varies significantly. This may be due to the adsorption behaviour of the polar fluid H₂O (Coppens, 1967).

3.3. Solution of CO₂ in mine water

CO₂ is characterised by a relatively high solubility in pure water. This is due to the hydration of CO₂. If the water is sufficiently alkaline, then the solubility will be increased by the formation of hydrocarbonate ions. Basically, an increase in pressure will increase the solubility, whereas increased salinity has the opposite effect. The latter is referred to as the salting-out effect. An increase in temperature may either increase or decrease solubility, depending on the prevailing pressure (cf. Takenouchi & Kennedy, 1965). The competing effects of pressure, temperature, and salinity generally result in an increasing solubility of CO₂ in formation waters with depth.

4. Key issues for CO₂ sequestration

The main issues concerning the feasibility of CO₂ sequestration in coal mines are the technical possibility of sequestration, the safety of an overpressured coal mine

reservoir and the possibility of injection in a low-pressure reservoir.

4.1 Reservoir pressure

Unlike natural gas-storage, CO₂ sequestration requires a minimum reservoir pressure to ensure the free-space storage capacity on long-term. The minimum pressure needed to prevent the reservoir from becoming flooded is the hydrostatic pressure prevailing in the formations at the gas-water contact (Fig. 3, point B). It can be assumed that the reservoir below this depth will become flooded and will generally have a lower sequestration capacity (solution storage and adsorption on wet coal; Fig. 3 between B and C). Parts above this depth will have a reservoir pressure that is higher than the hydrostatic gradient (Fig. 3 between A and B). The reservoir will therefore be overpressured. The highest amount of overpressure is exerted at the top of the reservoir (Fig. 3 point A). It is therefore generally the topseal that will determine the maximum reservoir pressure. The latter is directly related to the free-space capacity.

The quality of the topseal is therefore a critical issue in the evaluation of the sequestration capacity of coal mines. The overburden of many deep European coal mines contains lithological units that may act as a primary seal (chalks, marls, greensands or clays). In the Campine coalfield, marls in the lower part of the Cretaceous have acted as aquicludes. It is likely that the sealing properties of reactive chalk improve when brought in contact with CO₂ or CO₂-rich solutions by mineralisation at the pore throats (Czernichowski-Lauriol et al., 1996). Overlying Cenozoic deposits usually contain several clay-rich formations, which may act as secondary seals. For abandoned mines, also the sealing properties of the refilled shafts and reactivity of the fill need to be considered.

Many recent mine shafts in Europe have been filled with a combination of reinforced concrete, clay, and gravel down to a depth of several hundred meters (Fig. 4).

4.2. Risk assessment: fault-permeability and fault-valve activity

CO₂ is a normal constituent of exhaled air and is not toxic in low concentrations, but high concentrations (starting from 5 vol% for prolonged exposure) are hazardous (IEA, 2002). An uncontrolled release from an underground storage facility will not have a long-term effect, such as would be the case for nuclear or high toxic wastes, as CO₂ will be diluted in air or ground water. Therefore, slow migration (diffusion) toward the surface, although unlikely in sequestration systems, is no direct threat to humans or nature. At the surface, high concentrations can only be reached by a sudden, temporary release of CO₂. Because CO₂ is denser than air, it could build up in depressions or confined areas close to the ground surface and cause unconsciousness and asphyxiation of humans and animal life near the venting point. This is a known risk near volcanic lakes (e.g. natural disaster at Lake Nyas, Cameroon in 1986). In extreme cases, the pressure release could also result in freezing of the immediate surroundings, although this risk is generally limited to near-surface facilities (e.g., pipelines). Leakage may occur along infrastructure, such as injection wells or along geological structures, such as faults. Injection and monitoring facilities can be designed to be sufficiently safe. The effect of faults on the sealing properties of the overburden is site-specific. The discussion will therefore be focussed on abandoned coal mines in an extensional setting, such as the mines in the Limburg-Campine-northern Ruhr area.

In the Campine mine district, faults of post-Carboniferous origin are known to cross the overburden and the roof

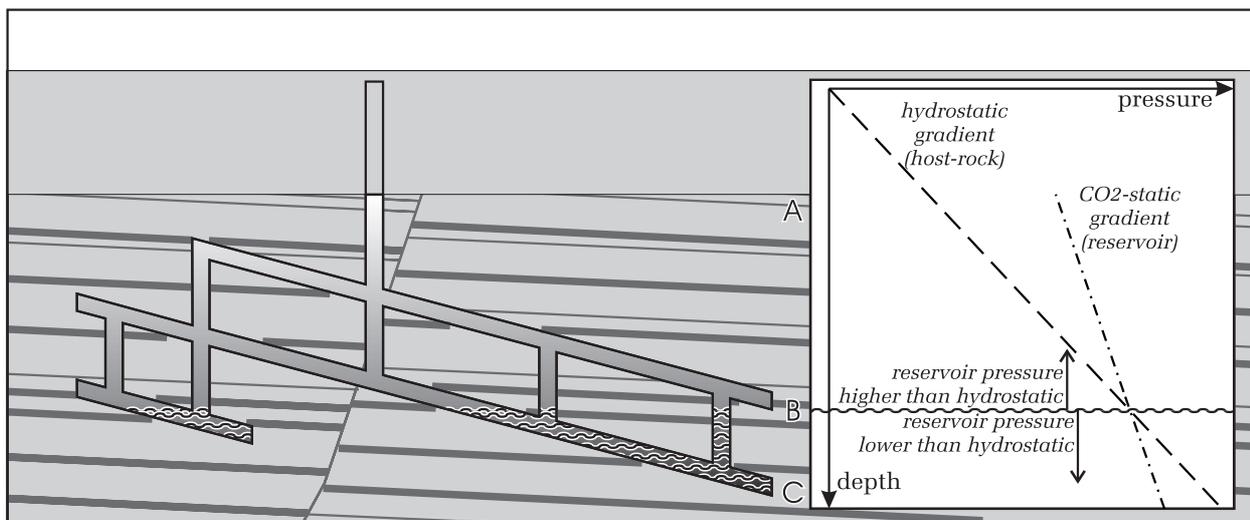


Figure 3. Schematic representation of a mine reservoir and the pressure evolution in the mine reservoir and the hosting formations. The reservoir is in overpressure relative to the hydrostatic gradient between points A and B, and in equilibrium with the hydrostatic pressure between B and C.

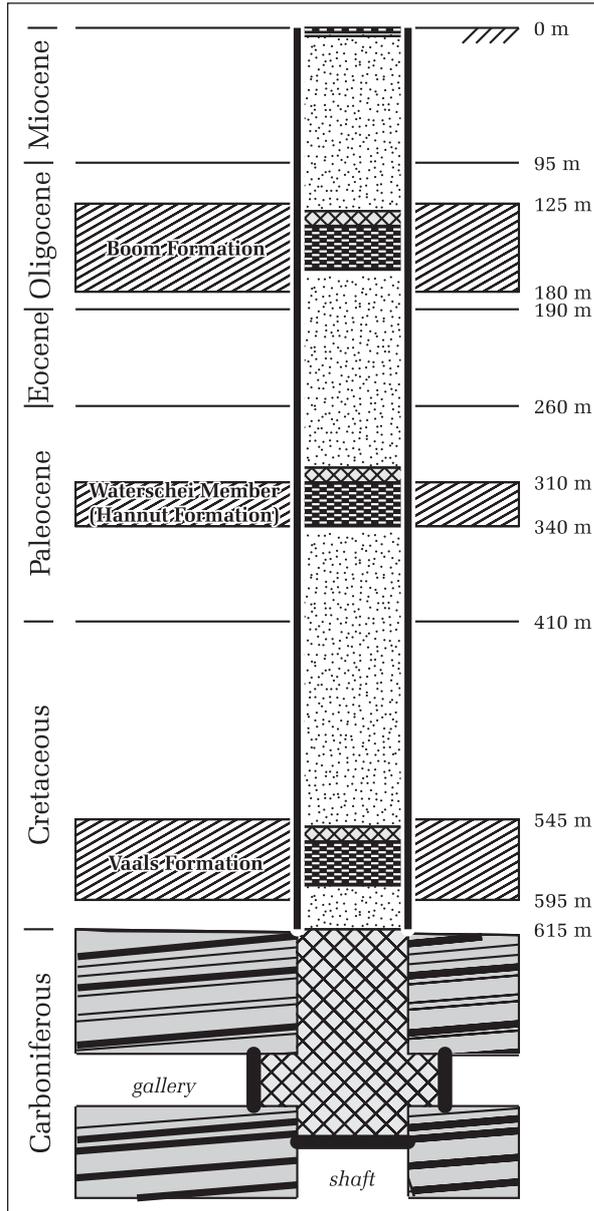


Figure 4. A schematic cross section of one of the shafts of Beringen that was meticulously sealed, as is typical for Campine coal mines. First, all constructions, pipes and cables left inside the shafts to be filled were removed. Above an existing workfloor at the highest working level or the base of the shafts, concrete was poured in 10-metre steps (resistance factor to corrosion B15 – DIN 1045). 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 $\beta_{w28} > 2$ MN/m²). In the overburden, 30 m thick clay barriers ($k < 10^{-9}$ m/s) were placed at the level of natural aquicludes. 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.

of the mines. These faults therefore may be preferential pathways connecting the mine reservoir with Cenozoic aquifers or the surface. During mining, some of these natural faults have caused extensive flooding of underground workings. Exceptionally, the composition of the water and the geologic architecture indicated Cenozoic aquifers as prime source. No data were found on the permeability of fault zones in the Campine area, but it is unlikely that faults that connect shallow aquifers with a mine, and that were considered as dry structures (after initial draining) for the lifetime of a mine, have high permeability. This is possibly due to fault gouge smearing in clay-rich formations. Faults that were crossed when constructing galleries only started to produce water, sometimes in a dramatic way, when nearby coal seams were mined by the longwall method, accompanied by roof collapse. This does not indicate primary permeability of the faults, but an opening or dilatation of the faults in response to changed stress conditions. There are no indications for strongly increased permeability in or around fault structures, but there is evidence of dilation of fault structures in response to changes in the stress regime by mining activity (Fig. 5). The stress regime will again change when the mine becomes flooded or will be used as a CO₂ reservoir. Therefore, the possibility that this would again result in dilatation of fault structures, followed by venting of CO₂ towards the surface, should be studied further.



Figure 5. Constructing a major gallery in silty claystone rock in the Waterschei colliery (Campine basin), showing decompacted natural fractures and veins (predominantly dipping right with steep angle 67°), besides some rock shattering produced by explosives. Photo Minders, in: Kolenmijnen André Dumont N.V., 1957. Waterschei 1907–1957. Edition L. Cuypers, Brussels (by permission of N.V. Mijnen).

Such an evaluation is a complex matter for at least three reasons. First of all, the stress situation close to faults is often anomalous. As a general rule, the main stress axes will to some extent be parallel or perpendicular to the fault plane. This makes it difficult to foresee how a change in the general stress state, with near horizontal and vertical axes, will affect the stress state on the fault plane. Second, the situation is not static, but dynamic, as the Campine area, close to the Rur Valley Graben, is an active extensional setting where stress states have changed through time and faults are reactivated. And third, the current stress distribution is seriously affected by the historical mining activities. This resulted in zones, mainly in a rim around a mined panel, where the vertical stress may be several times higher than the hydrostatic pressure. Inside this rim, the vertical stress may be much lower, but it will again increase towards the centre of the panel. The exact distribution of the stress does not only depend on the configuration of the workings and the rock properties, but also on the use of backfilling techniques and the direction and sequence at which a panel was excavated. Over longer periods, though, pressure distribution will return to almost normal undisturbed conditions.

In conclusion, the fault structures themselves may not be highly permeable, although their actual permeability remains to be determined. This is demonstrated by the documented examples of influx of water along faults or fault zones into the mine that indicate fault-valve behaviour (opening and subsequent closure of faults). The response of faults to changes in the stress regime that will occur upon filling the mine with CO₂ presently cannot be accurately foreseen. A case study is probably necessary to resolve this matter. It may, however, be remarked that faults cross-cutting cap rocks also occur in natural gas and oil reservoirs without affecting sealing properties, and that several aquifers and secondary seals in the overburden will hinder the escape of CO₂ toward the surface. Mining-induced fractures are not necessarily a direct threat to reservoir integrity but should be evaluated in closer detail as well, including a new fracture system that may develop in response to possible CO₂-induced coal swelling.

4.3. Injectivity - temperature control during injection

A technical obstacle for injection of CO₂ in coal mines is the low initial reservoir pressure, which will be close to atmospheric pressure just after abandonment. Other sequestration systems start injection at pressures where CO₂ is liquid or liquid-like. In coal mines, the initial pressure is often close to atmospheric. If liquid CO₂ would be injected into such a reservoir, then liquid CO₂ would evaporate and cool parts of the reservoir significantly below 0 °C. Any water present at the injection point would freeze and possibly hinder further injection. Also, the freezing may result in damage and collapse of parts of the reservoir.

Part of this problem may be overcome by adjusting the injection pressure. If CO₂ is being transported by pipeline, then it will be delivered as liquid CO₂ at high pressure (~11 MPa). As long as the reservoir conditions are such that the reservoir is gas-filled, CO₂ can be partly depressurised prior to or during injection. This will prevent evaporation of liquid CO₂ after injection and thus avoid excessive cooling of the reservoir. Injection rates of gaseous CO₂ can be sufficiently high, as is proven by the CH₄ storage facility at the Leyden coal mine, where maximum injection occurs at an average of 5 m³/s (~300 000 tons CO₂/y) per well (Schultz, 1998).

The problem arises when the reservoir contains both gas and liquid. At these conditions, only liquid CO₂ can be injected, and a considerable amount of cooling will occur around the injection point. This cannot be simply avoided by pressure reduction. Two options have been considered. Apparently, the simplest is heating CO₂ prior to injection, but this is a very expensive operation that is debatable because of the extra, be it limited, production of CO₂. Another possibility is using adapted injection equipment. A possible example of a simple custom-engineered solution is illustrated in Figure 6. The configuration consists of an outer and an inner injection pipe, mounted in an open space, like an intact vertical shaft section, and combines three principles:

1. Allowing higher injection rates and relatively low injection pressures during the injection of gaseous CO₂.
2. Spreading the heat depletion sufficiently when injecting liquid CO₂ in order to allow heat buffering by the reservoir.
3. Expansion of CO₂ in a water-free surrounding, which minimises the risk of blocking of the injection pathways by ice.

In the initial situation where the reservoir has low pressure (Fig. 6b), both the outer and inner pipe can be used for injection of gaseous CO₂ in order to allow for high flow rates. A minimum flow rate of 6.5 kg/s or 3.3 m³/s should be realised if 200 000 tons of CO₂ need to be stored per year, which is lower than the injection rates used for CH₄ storage in coal mines (e.g. average of over 5 m³/s per well at Leyden coal mine). This will limit the pressure reduction at the injection point. Any cooling here will probably be distributed efficiently by the migration of the injected CO₂ gas (rapid circulation and migration through galleries).

When the reservoir pressure is raised to the point of saturation of the reservoir, this is when both liquid and gaseous CO₂ occur, liquid CO₂ will have to be injected. This is done only through the inner tube. The higher density of liquid CO₂ will still guarantee sufficient injection capacity. Expansion and cooling will mainly occur at the exit point of the inner tube (Fig. 6c). This will not result in an immediate and localised cooling of the reservoir, since the expanded fluid is contained in the outer pipe. When the cold fluid moves further downward, it will partly equilibrate to reservoir temperature before

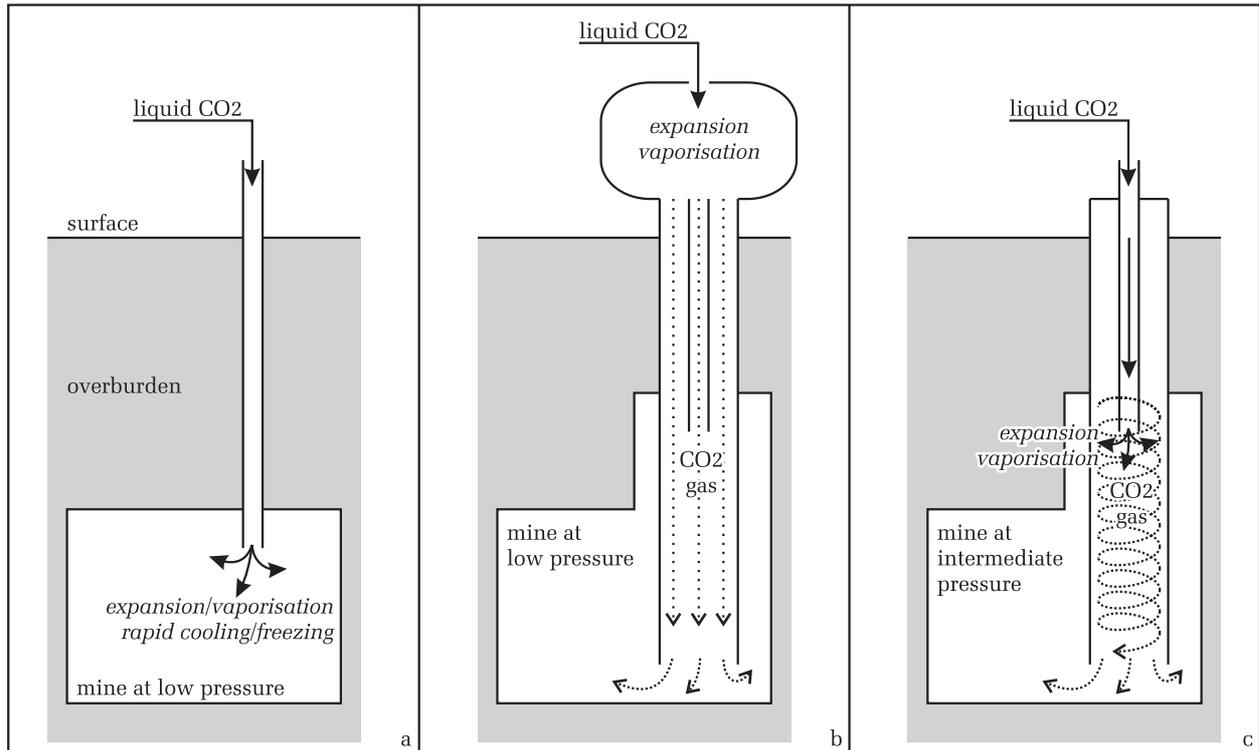


Figure 6. Schematic example of customised injection equipment to increase the injection rate of low density CO₂ and prevent freezing of the reservoir when injecting liquid CO₂. (a) If liquid or supercritical CO₂ is injected in a low pressure reservoir, then CO₂ will expand inside the reservoir. This will cause rapid cooling, with danger of blocking of the injection equipment by ice, or damaging underground infrastructure. (b) In a first phase, CO₂ can be expanded at the surface and be injected at lower pressure. This can be done using the outer and inner pipe, that become necessary when (c) the reservoir pressure is reached at which CO₂ will start to condense. This implies that injection of liquid CO₂ becomes possible. When CO₂ is injected in the inner pipe, it will be inside the outer pipe when it expands. This allows the cold CO₂ to equilibrate to reservoir temperature before exiting the outer tube. Because of the continuous flow of CO₂, no water will be present in the outer tube.

exiting the outer tube. Therefore, no drastic cooling will occur near the injection point, and the risk of freezing the reservoir will be reduced. The risk of blocking injection equipment is minimal, since dry CO₂ has been injected for some years, and no water will be available in the outer tube to form ice. If condensation or ice form, for example because continuous injection could not be maintained, slightly heated CO₂ could be injected for a limited amount of time to dry the outer tube.

Injection of CO₂ in an abandoned mine poses specific problems when compared to other sequestration systems, but these can be overcome by limiting the amount of overpressure and adjusting the design of the injection wells.

5. Assessment

As discussed in chapter 3, it is generally not realistic to obtain all data necessary to calculate the exact sequestration capacity of a coal mine. The total amount of accessible coal and the reservoir expansion in response to elevated pressures are examples of parameters that

usually can not be accurately determined. However, for most mines part of the storage capacity can be ascertained. If this conservative estimate is sufficiently large, then the project may be evaluated positively. Injection and pressure tests can be used to estimate the true sequestration capacity of the coal mine during its development as a CO₂ reservoir.

5.1. Campine coal field

Mining history (1917-1992) and geology in the Campine basin are well documented (e.g. van Tongeren & Laenen, 2001). The setting and layout of the seven mines abandoned are comparable. Coal mining occurred at depths between 350 and 1090 m. Some connecting galleries have been driven, mainly for purposes of safety. After closure all shafts were filled from a depth of ~560 m upwards, using reinforced concrete at the bottom and top, and clay where relatively impermeable strata occur. No monitoring facilities were installed. Based on comparison with Dutch mines and the geology and hydrogeology of the Campine basin, it is assumed that only the deepest parts of the mines are currently flooded (average rise of water

	Residual volume	Sequestration capacity		
		ascertained	potential	total
Beringen	(5.9 10 ⁶ +/- 2.3 10 ⁶) m ³	3.0 Mton	2.5 Mton	5.5 Mton
Zolder	(6.2 10 ⁶ +/- 2.4 10 ⁶) m ³	3.2 Mton	2.6 Mton	5.8 Mton
Houthalen	(1.8 10 ⁶ +/- 600 000) m ³	0.9 Mton	0.8 Mton	1.7 Mton
<i>subtotal</i>	<i>13.9 10⁶ m³</i>	<i>7.1 Mton</i>	<i>5.9 Mton</i>	<i>13.0 Mton</i>
Eisden	(5.5 10 ⁶ +/- 2.1 10 ⁶) m ³	2.8 Mton	2.3 Mton	5.1 Mton
Waterschei	(5.1 10 ⁶ +/- 2.0 10 ⁶) m ³	2.6 Mton	2.2 Mton	4.8 Mton
Winterslag	(4.8 10 ⁶ +/- 1.9 10 ⁶) m ³	2.4 Mton	2.0 Mton	4.5 Mton
Zwartberg	(3.0 10 ⁶ +/- 1.1 10 ⁶) m ³	1.5 Mton	1.3 Mton	2.8 Mton
<i>subtotal</i>	<i>18.4 10⁶ m³</i>	<i>9.4 Mton</i>	<i>7.8 Mton</i>	<i>17.2 Mton</i>
<i>total</i>	<i>32.3 10⁶ m³</i>	<i>16.4 Mton</i>	<i>13.7 Mton</i>	<i>30.1 Mton</i>

Table 1. Estimated residual volume and sequestration capacity for the abandoned Campine coal mines.

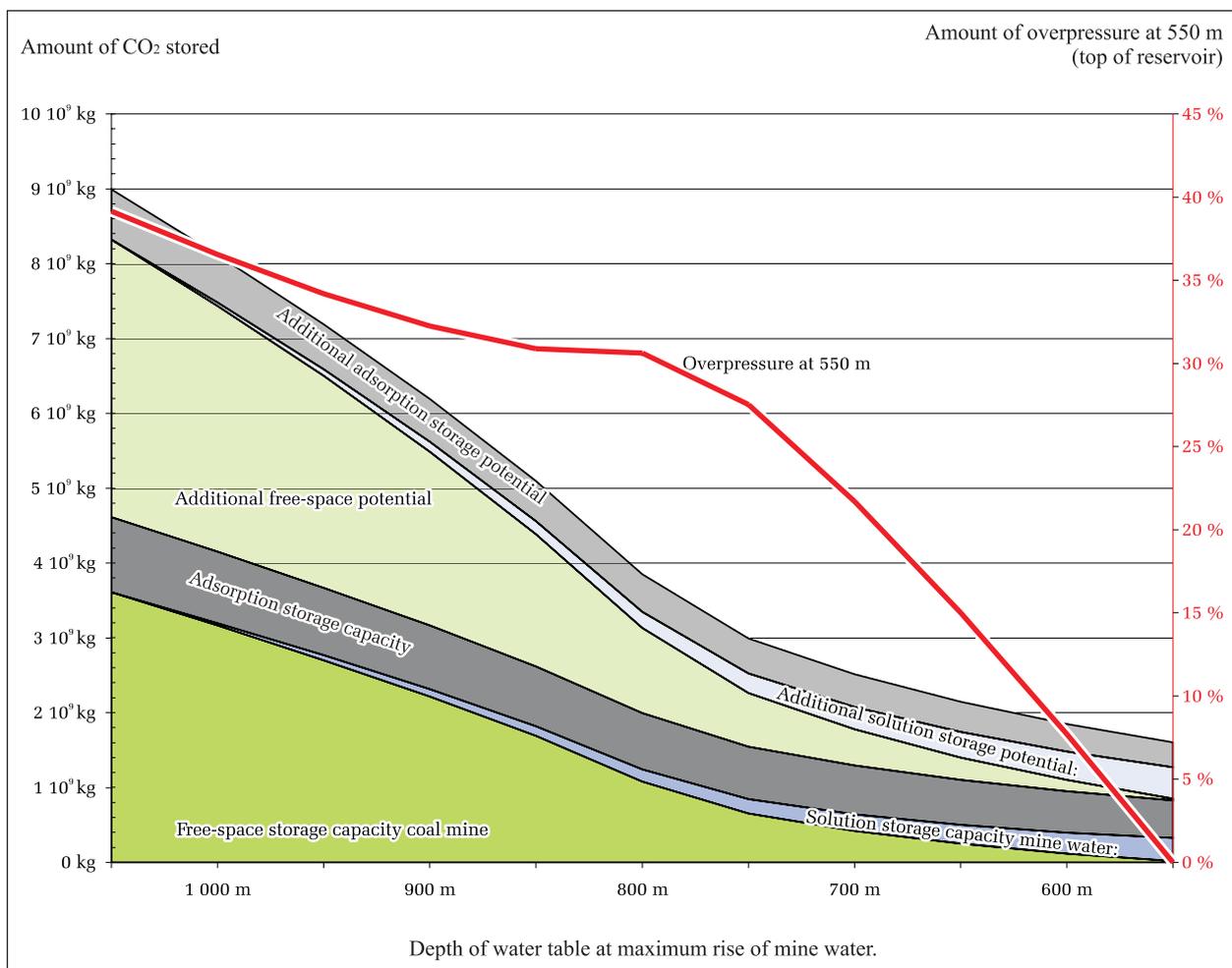


Figure 7. The sequestration capacity of the Beringen colliery for different amounts of overpressure on the top seal. Calculation with CO₂-VR (Piessens & Dusar, 2003c). X-axis: depth of the water table; leftmost Y-axis: amount of sequestered CO₂; right Y-axis: amount of overpressure at the top of the reservoir (550 m), in percentage relative to the hydrostatic pressure.

Company	City	Country	Distance (km)	CO ₂ emission (ton/y)	CO ₂ concentration (%)
DSM Limurg BV	Geleen	The Netherlands	44	4048000	100
BASF	Antwerp	Belgium	58	818000	100
Ineos	Antwerp	Belgium	58	121000	100
Esso Benelux	Antwerp	Belgium	58	115000	100
Clariant	Gendorf	Germany	67	62000	100
Shell Chemicals	Moerdijk	The Netherlands	80	91000	100
Hydro Agri Sluiskil BV	Sluiskil	The Netherlands	99	1099000	100
Kemira S.A.	Tertre	Belgium	121	696000	100

Table 2. Nearest large producers of pure CO₂ for the Beringen colliery, for 'early opportunities' projects (source Ecofys for GESTCO project).

levels estimated at less than 20 m/y). Progressive flooding will increasingly hinder sequestration as time passes.

The temperature gradient in the Campine basin is 0.035 °C/m for the upper 1000 m (Vandenberghe et al., 2000). The average reference temperature in Belgium has been set at 9.8 °C, but may have risen by 0.5 °C during the last 50 years. Groundwater temperatures of 48 °C have been measured at the deepest mine levels (Dreesen & Lagrou, 1999).

The residual space volume of the coal mines were calculated from the data of van Tongeren & Laenen (2001), taking into account a residual volume fraction of 7% (Tab. 1). The permeability of the main connecting galleries is set at 1000 darcy. The porosity of the unaffected host rock is around 3%, and porosities in the range of 15% are reported in Westphalian C sandstones, but permeability rarely reaches 1 mD. The porosity and permeability of the shale unaffected by mining or mine subsidence is considered to be zero (van Tongeren et al., 2000; van Tongeren & Laenen, 2001). The chalk from the basal Cretaceous deposits has a low to very low permeability and forms the primary seal for the reservoir. Several secondary seals are present as clay layers in the Cretaceous and Tertiary section. The sealing quality of chalk and marl is supported by century-long mining activity in which coal was completely dewatered and groundwater remained perched above the chalk. A large part of the coal reserves are located deeper than the deepest mined levels or are located in parts of the concession where no coals have been extracted. Therefore, it is assumed that only 50% of the 80·10⁶ tons of coal reserves in the Beringen colliery is forming part of the reservoir, divided into 30% ascertained capacity and an additional potential of 20%.

The sequestration capacity of the Beringen colliery has been calculated for different amounts of overpressure (Fig. 7). If an overpressure of ~30% is taken to be realistic, then an ascertained amount of about 3·10⁶ tons can be sequestered, possibly extendable to over 5.5·10⁶ tons. Since Beringen is a typical Campine coal mine, the sequestration capacity of the other collieries can be estimated, using the residual volume of each mine (Tab. 1). Beringen alone can maintain CO₂ sequestration for over

15 years at a minimum rate of 200 000 tons/y. Beringen is connected with Zolder and Houthalen. The combined capacity of these collieries is sufficient to sequester 200 000 tons/y for 35 years. The total ascertained capacity for all Campine collieries is 1 000 000 tons/y for more than 15 years, with a potential to sequester for over 30 years at this rate.

In view of the ascertained sequestration capacity, the availability of CO₂ producers (Tab. 2), the presence of primary and several secondary seals, and sufficient data on geology and mining history, all Campine coal mines can be considered as first-line projects, with a possible preference for Beringen or the combined Beringen-Zolder-Houthalen collieries. The main reservation is the insufficient characterisation of the primary chalk seal.

5.2. Hainaut coalfield

The mining history in southern Belgium started in the 12th century and lasted until 1983. Reliable mining maps are available from circa 1840 onward, especially for the larger collieries. The Basse-Sambre, Namur, Liège, Herve, Theux, and Dinant basin coalfields of South Belgium are of no particular interest for CO₂ storage. In most cases, there is an absence of overburden or of an intraformational seal. Inclined galleries and shafts form numerous connections with the surface. Exploitations are relatively small and are shallow, reaching maximal depths of 750 m in the Liège coalfield, 500 m in Herve and Basse Sambre and only 150 m in the other coalfields. As a result, there is usually direct inflow of meteoric water, and some of these collieries may even drain into surface river systems.

The most important coal basin for potential CO₂ storage sites in southern Belgium is the Hainaut basin, composed of the Borinage, Centre and Charleroi coal fields. This region is characterised by an intraformational seal, recent mining (post 1920) up to 1495 m depth, and often dry conditions for mining with only localised leakage through faults and man-made shafts and boreholes. Gas drainage was common practice and occasionally continued up to 25 years after closure of the collieries

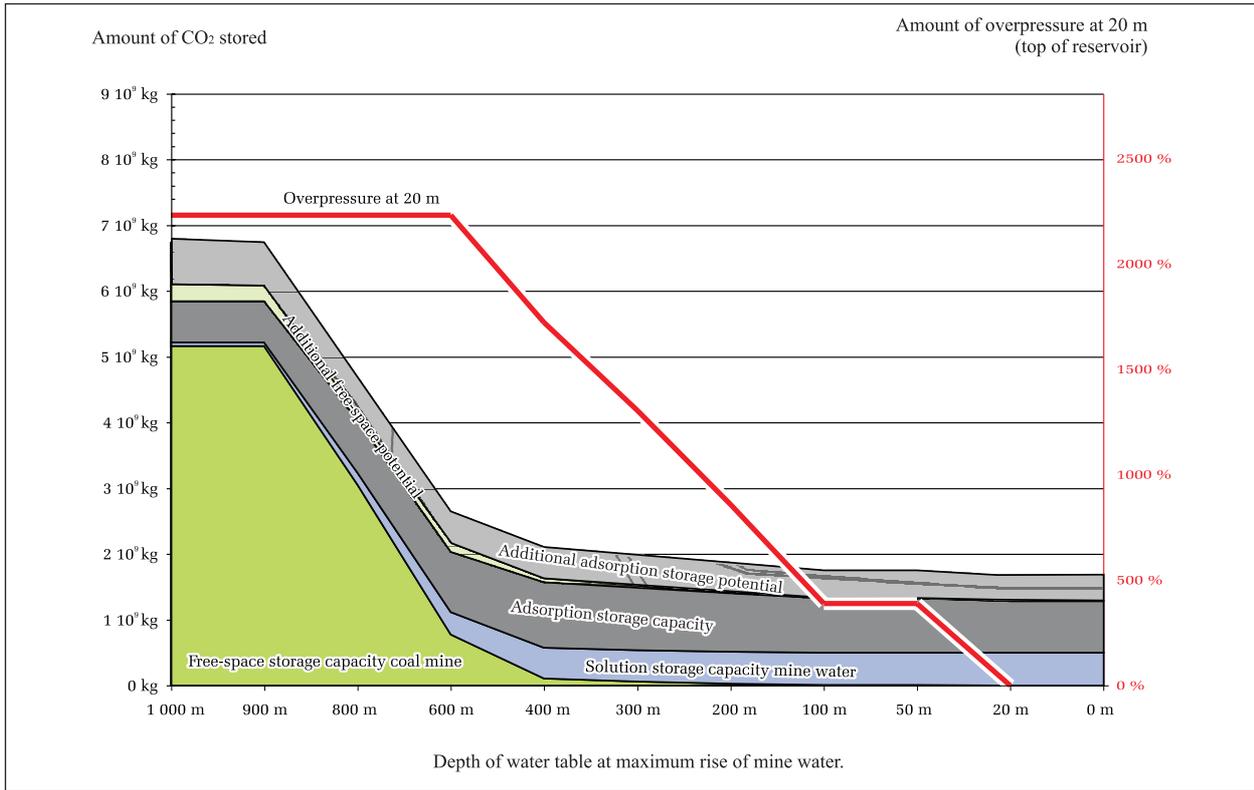


Figure 8. The sequestration capacity of the Anderlues colliery for different amounts of overpressure for the current configuration with a seal at ~20 m depth. Note that unrealistic high amounts of overpressure are needed to create free-space sequestration potential. Calculation with CO₂-VR (Piessens & Dusar, 2003c). X-axis: depth of the water table; leftmost Y-axis: amount of sequestered CO₂; right Y-axis: amount of overpressure at the top of the reservoir (20 m), in percentage relative to the hydrostatic pressure.

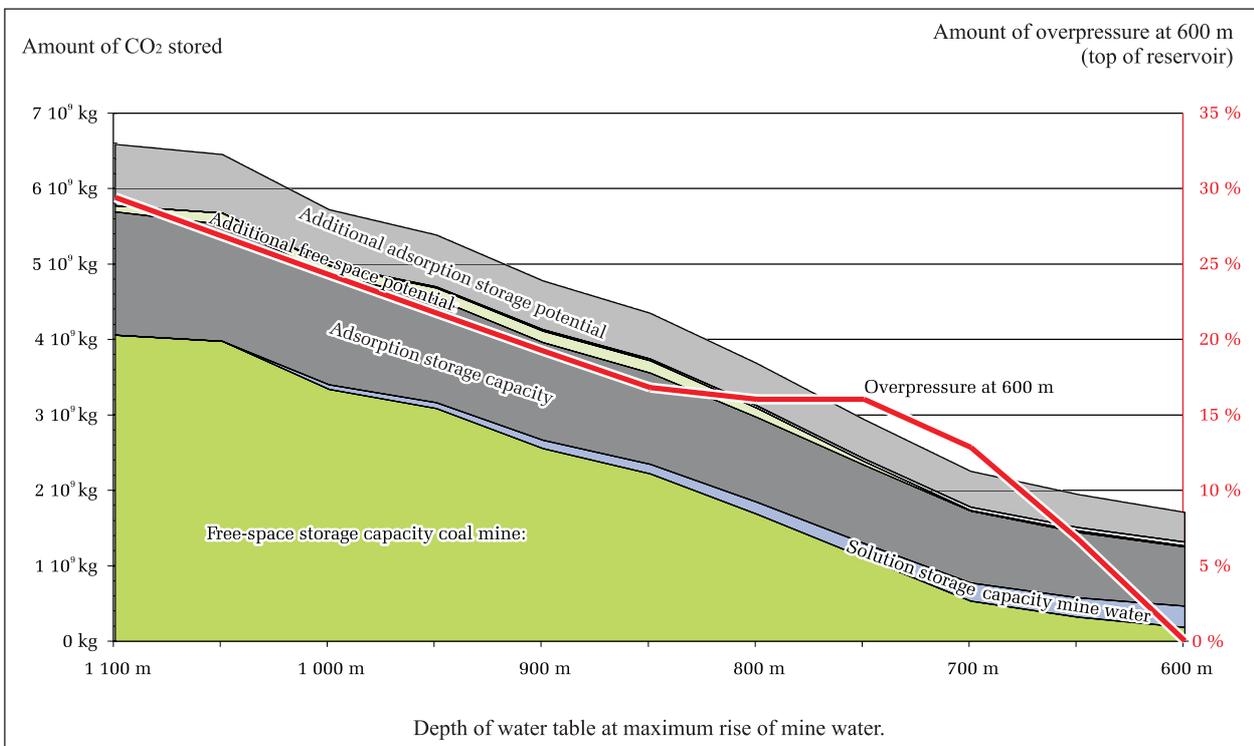


Figure 9. The sequestration capacity of the Anderlues colliery for different amounts of overpressure, assuming that the shafts are sealed at a depth of about 600 m. Calculation with CO₂-VR (Piessens & Dusar, 2003c). X-axis: depth of the water table; leftmost Y-axis: amount of sequestered CO₂; right Y-axis: amount of overpressure at the top of the reservoir (600 m), in percentage relative to the hydrostatic pressure.

Company	City	Country	Distance (km)	CO ₂ emission (ton/y)	CO ₂ concentration (%)
Electrabel SA	Péronnes	Belgium	20	629	15
Electricité de France	Pont-sur-Sambre	Belgium	24	478	15
Electrabel SA	Roux	Belgium	25	1481	15
Cockerill Sambre SA	Charleroi	Belgium	27	3166	15-20
Kemira SA	Tertre	Belgium	35	696	100
Grande Paroisse	Waziers	France	75	194	100

Table 3. Nearest large producers of CO₂ within 30 km from the Anderlues colliery, and pure CO₂ producers within 100 km (source Ecofys).

(Fontaine l'Évêque, Bois-du-Cazier, Monceau-Fontaine, Centre and Anderlues coal mines)-(Frenay, 1981). A main disadvantage is the presence of numerous old shafts that present a risk for leakage when remaining undetected or untreated. Mines that were not used until recently for gas drainage or gas storage are not monitored and may be largely flooded.

The colliery that has been selected for evaluation is the Anderlues coal mine. It was used for storage of natural gas until 2000. Since then, gas is being recovered, and it is foreseen that the reservoir will be depleted around 2005. More details on this mine are given in §2.2.1. The sequestration capacity of this mine is low due to the very shallow depth of the seals. This results in a low maximum reservoir pressure and a large amount of flooding, although flooding proceeds very slowly. A non-flooded zone of 50 m thickness at the top of the reservoir would result on long term in almost 400% overpressure for an overburden of 20 m. This means that it is virtually impossible to maintain free-space sequestration capacity. The total sequestration capacity is therefore the sum of solution and adsorption capacity, or about $1.7 \cdot 10^6$ tons of CO₂ (Fig. 8). Higher sequestration capacity can be reached when only the deeper part of the mine is used, this is the part below the impermeable thrust zone at about 600 m depth (Masse Fault). This does however imply that the shafts would have to be sealed at this depth, which is a technically feasible but expensive operation. In this configuration close to $6 \cdot 10^6$ tons of CO₂ could be sequestered safely if an overpressure of 30% would be acceptable (Fig. 9). This is sufficient to sequester CO₂ during 30 years at a rate of 200 000 tons/y.

The overview of CO₂-producers in Table 3 demonstrates the nearness of several potential providers, including Kemira SA in Tertre that produces pure CO₂. The reduced thickness or absence of a seal, the great number of shafts and the danger of undocumented older shallow workings in many coal mines from the southern Belgian coal fields, makes these mines, in their current state, probably unsuited for CO₂ sequestration. Where the Carboniferous is cross-cut by impermeable thrust

faults at greater depths, as is the case for the Péronnes and Anderlues collieries (Delmer, 1997; Mostade, 1999), the residual mine structure may be adapted to allow the sequestration of CO₂ in the deeper parts. This, however, involves significant initial investment. The Anderlues mine and comparable collieries are therefore considered as second-line projects.

6. Conclusion

It is likely that sequestration of CO₂ in abandoned coal mines such as those of the Campine coal field is possible in the short-term. The main item that needs further confirmation is the sealing capacity of chalk. The problem of injecting in low-pressure reservoirs requires special attention, but is technically feasible. The estimation of the total capacity of a mine reservoir is different from conventional reservoirs, since CO₂ can be stored in free space, in solution, and as an adsorbate. The total capacity is strongly dependent on the final level of flooding, which is directly proportional to the maximal reservoir pressure. The Campine coal mines are therefore better early opportunity candidates than the less-well sealed mines from the Hainaut coal field. First-line sequestration projects will probably use concentrated CO₂ available from several chemical production processes.

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