

CO₂ sequestration in underground coal mines

Introduction

Concentration of greenhouse gases (GHG) has been on the rise in the past century. CO₂ is the most abundant greenhouse gas and its concentration has risen significantly over last several decades. The increased industrial activity is agreed upon to be the main reason for higher atmospheric CO₂ concentration. The increase of GHGs in the atmosphere has resulted in an average increase of 0.6°C in the world's temperature over the last century (IPCC, 2001).

Energy industry has been the main contributor of the anthropogenic GHG emissions. Fossil fuels are the main stay of global energy mix. Worldwide oil, gas and coal together contribute to ~80% of the energy mix. Coal is the second major source of primary energy production contributing to 27% world's energy needs as compared to oil (32%) which is the largest source of primary energy (IEA, 2013). Coals are widely distributed across the world and constitute major sources of primary energy in many countries including India. Coals contributed to 59% of our primary energy production in the year 2012 accounting for 132,288 MW out of a total of 225,793 MW (MOP, 2013).

Coal will continue to remain the mainstay of India's energy mix for coming several decades. However, more thrust has to be given on adopting clean coal technology so as to minimize the environmental footprints of coal handling and combustion process. Research is being carried out to develop many clean coal technology alternatives that can be practiced industrially. The clean coal technology options can be mainly classified into three categories viz. pre-combustion technology, combustion technologies and post-combustion technology. CO₂ capture and sequestration of (CCS) is one of the post-combustion technologies that have a lot of promise in the near-term and mid-term. CCS technologies involve separation of CO₂ from the flue gas of fossil fuel based power plant and its storage/disposal in various potential geological sinks for geologically significant period of times (thousands of years). CCS technologies have been proposed as an alternative for the continued use of fossil fuel. There are three steps

involved in CCS: (i) capture of CO₂ from power plants and other emission sources, (ii) transport of the gas in pipelines or compressed medium and (iii) underground injection of the gas in geological formations. The potential geological formations that can store CO₂ include abandoned coal mines, saline aquifers, oil and gas reservoirs etc.

Abandoned underground mines in the past have been used for storage of natural gases. Three successful natural gas storage operations in abandoned mines exist worldwide. Two mines namely Peronnes and Anderlusare located in Belgium. The third mine used for storage of natural gas is Leyden Abandoned coal mine in the USA (Jalilli et al, 2011).

Experience of natural gas storage in abandoned mines

Leyden coal mine is situated in the state of Colorado and located near Denver. The mine was in operation during the period 1903 to 1950 during which it produced about 6 Mt of sub-bituminous coals using room and pillar method of mining. Access to the mine was made by four shafts. Coals were extracted from two horizontal seams at depths of 210 m and 225 m from the surface. The cap rock consisted of 20 m claystone. About 35% of coals were extracted and 65% coals were locked as pillars when the mining operations were discontinued. The gas storage operation was commenced in 1961 and is still in operation. During the initial development of the storage system, water was pumped from the mine as gas was being injected. Two active water wells are currently used to continuously remove approximately 50,000 m³ of water from the mine each year. The leak off test shows that the cap rock can only withstand 75% of the hydrostatic pressure which is ~1.8 MPa. It was estimated that the coal in the mine can adsorb 85 to 127 Mm³ at pressure of 1.1 MPa and 100 to 140 Mm³ at pressure of 1.8 MPa (Schultz, 1998).

The Anderlus mine is located in the Hainaut coalfield in Southern Belgium and was operating between 1857 and 1969. A total of 25 Mt of sub bituminous coal were produced during this period. Coals were extracted at depths varying from 600 to 1100 m. The thickness of the overburden in the mine is almost 50 m. Natural gas storage operation began in 1980. After closure, the mine pumping facilities were maintained. The gas pumping was discontinued in 2000 because the cost

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of maintenance of the shafts was becoming prohibitory.

The Peronnes mine is also located in the Hainaut coalfield in Southern Belgium. It was in operation from 1860 until 1969. This mine could store 120 Mm³ of CH₄. The storage operation for both Peronnes and Anderlus mines were stopped in 1996 and 2000, respectively as an unforeseen cost for sealing the shafts was required.

CO₂ storage in coal mines

The successful experience of storage of gas in the mines has encouraged the engineers to think about using the abandoned mines as CO₂ repositories for CCS purpose. Although the storage of natural gas in mines appears to be similar to the proposed storage of CO₂ in mines, there are a few fundamental differences between the two. The first is that the natural gas is stored in the mine for a very small period of time varying between several weeks to several months after which it will be recovered for commercial use. In contrast the CO₂ will be stored in the mine for very long period of time of the order of thousand years. Secondly, the natural gas is an inflammable gas while the CO₂ is an asphyxiant. So the safety standards and procedures to be followed will be different. Thirdly, the natural gas in the mine is mostly stored as a free and compressed gas while for storage of CO₂ a number of mechanisms play important roles. These mechanisms are discussed in the next section. The basic requirement for any gas storage in a mine or any geological reservoir is that it must have an impermeable cap rock so that the gas is not allowed to leak into the surface. Further the rock strata above and below the reservoir should have less permeability. The schematic of CO₂ storage in any geological reservoir is shown in Fig.1. The CO₂ at high pressure mostly in the supercritical state is injected in the targeted formation. The injected CO₂ migrates into the rock away from the well by diffusion, or by pressure gradient.

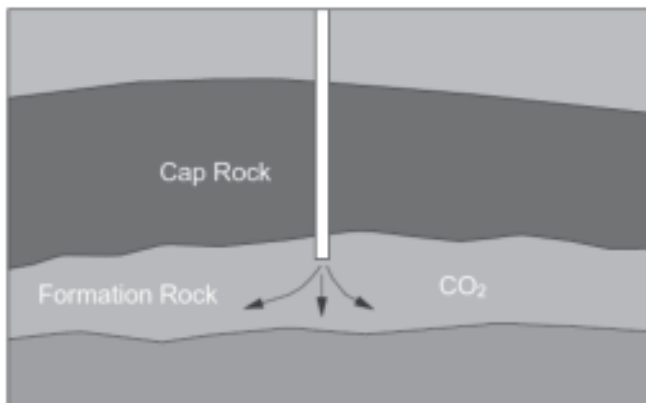


Fig.1 CO₂ sequestration in geological formations

CO₂ storage mechanisms

There are three principal mechanisms of CO₂ storage in an abandoned coal mine. They are: (i) structural trapping in

which free CO₂ gas is stored in the residual space in the mines, (ii) solubility trapping in which CO₂ is dissolved in the formation water, and (iii) adsorption trapping in which CO₂ is stored as adsorbed gas in the left out coals and carbonaceous shale formations in mine as well as in the roof and floor strata. The three mechanisms of sequestration in the mines are shown in a schematic diagram as shown in Fig.2.

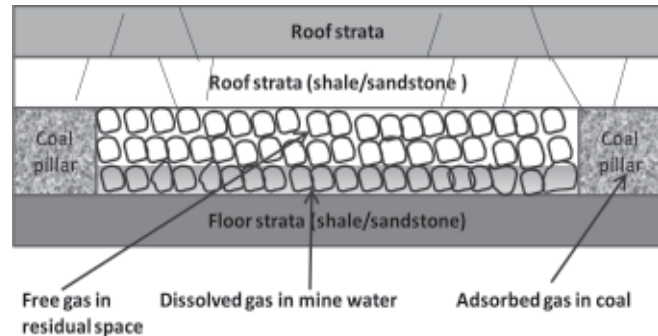


Fig.2 Mechanims of CO₂ storage in abandoned coal mine

STRUCTURAL TRAPPING

After the extraction of coals by mining, the mined out area is converted to goaf. The roof strata break and fall into the goaf and fill up the void space. However a lot of void spaces are created in between the rubbles making the mined out area a porous formations. The total void spaces present in the abandoned mine is known as the residual space. The residual space would be either filled up with gases comprising methane-air mixtures (if the mine is not flooded) or water. In case of a gas filled residual spaces, the injected CO₂ will displace the methane-air mixture and occupy the places taken by the later. In case of a flooded residual space, the injected CO₂ plume travels through the porous rocks, and gets trapped in the small pore spaces by surface tension and then move upwards until it reaches a trap rock and accumulated below it. In the coal mines, usually the cap rock is shale formation which is very less permeable for flow of gas and provides a good capping. Further the likelihood and intensity of leakage is reduced because the CO₂ is adsorbed on the shale matrix and cannot migrate from the (shale) formation to the surface as long as pressure and temperature remain constant in the reservoir (Busch et al, 2009). The trapping of CO₂ by such a cap rock layer is called structural trapping. The residual space in the mine is filled up gradually and the pressure of CO₂ in the residual space gradually goes up. The pressure in the residual space can go up to the pre-mining reservoir pressure of the mine. However, it is desirable that CO₂ is maintained at a pressure less than the hydrostatic pressure so as to prevent uncontrolled leakage of gases from the reservoir to the adjoining strata. Reservoir pressure higher than the hydrostatic pressure will not allow the formation water to be seeping into the residual space. A lower reservoir pressure below than hydrostatic pressure will lead to water infusion to the residual space. The infused water

would compress CO₂ highly and a high pressure gradient is created that result in gas leak to the surface. Even though low levels of leakage are inevitable, it should be within the stipulated limit of less than 0.1% per year assuming a sequestration time of 1000 years (Keith and Wilson, 2002; Bachu and Adams, 2003). The presence of any fractures, or faults in the cap rock would also lead to the leakage of the CO₂ to formations above the reservoir. Residual gas trapping is one of the most important storage mechanisms in storing CO₂ in the mines.

SOLUBILITY TRAPPING

Water in a mine is present in two forms. Some water is accumulated on the dip side of the mine. The residual spaces as well as coal pillars also act as an aquifer and huge quantity of water is available in their pore space. Both forms of water have capacity to dissolve CO₂. A lot of CO₂ can be stored in this water by the process of dissolution which is referred to as solubility trapping. CO₂ comes in contact with the mine water as well as in the mine aquifer through diffusion, convection and dispersion. The rate of solubility trapping depends mainly on the amount of carbon dioxide coming in contact with the water that is unsaturated with carbon dioxide. The dissolution process occurs much slower and the storage capacity is lower compared to structural traps. The solubility of carbon dioxide in the formation water depends on temperature and the salinity of water. It decreases with increasing salinity. The contribution to the total sequestration capacity is only up to 10% of that of the storage capacity of pure CO₂ in natural traps (Keith, and Wilson, 2002).

ADSORPTION TRAPPING

Coals are porous organic materials and have capacity to adsorb CO₂ in their micro-pores. Coals remaining in the mine after abandonment can adsorb significant quantity of CO₂ in their internal pores. Coal seams are characterized by a dual-porosity structure. They consist of both micro-porosity, pores less than < 2 nm and meso- (2 to 50 nm) and macro-pores (> 50 nm). Micro-pores constitute more than 90% of the coal porosity and because of their micro-porosity they provide a very large surface area for adsorption of CO₂. Adsorption is the phenomenon of accumulation of adsorbate molecules (methane/CO₂) on the internal surface of coal micro-pores due to van der Waal's force. CO₂ adsorption capacity of coal depends on physico-chemical property of coals such as porosity, pore size, surface area, and reservoir properties like pore pressure, reservoir temperature and pore water saturation. Adsorption of CO₂ on coal can be modelled by the widely used Langmuir Equation which is as given below.

$$V = \frac{V_L * P}{P + P_L} \quad \dots \quad (1)$$

where V is the volume of CO₂ adsorbed (m³/t of coal), P is the reservoir pressure (bar), V_L is the adsorption capacity at that temperature (m³/t of coal), P_L is a constant called

Langmuir pressure (bar).

MINERAL TRAPPING

The injected CO₂ gets dissolved in water and forms a weak carbonic acid. Over along period of time, this weak carbonic acid can react with minerals in the rock strata to form solid carbonate minerals and this is known as mineral trapping. The rate of mineralization will depend on: contact time and contact surface between mineral grains and the carbonic acid. The size of the contact surface area will depend on the grain/pore size, and porosity of the rock strata. It will also depend on the formation temperature, pore pressure, and the salinity of the water. The mineralization is a very slow process and usually not significant for CO₂ storage in a mine.

Conditions for CO₂ storage in mines

It is important to note that all coal mines are not suitable for CO₂ storage. Potential abandoned coal mines should meet some basic conditions (Piessens and Dusar, 2003; Jalili et al, 2011).

- ♦ It should have an impermeable cap rock layer of sufficient thickness. A layer of shale formation which usually has very less permeability should be a good candidate.
- ♦ The mine should be isolated from other mines. There should be no lateral communication with other mines.
- ♦ There should not be any extensive fracture joining the mine with the surface. Any fracture present would allow the CO₂ to escape to the atmosphere. Existing fractures if any should be sealed by cementation or other means.
- ♦ The amount of the influx of water into the mine should preferably be low. The mine has to be kept dry by pumping for storage of CO₂ to prevent gas pressures which may cause leakage of the gas.
- ♦ The mine should be at a sufficient depth from the surface. A potential mine should have working at a minimum depth of 500 m.
- ♦ The reservoir pressure should be kept below the hydrostatic pressure in order to prevent uncontrolled leakage of the gas. An under-pressured reservoir will result in water seepage into the reservoir; however the water seepage is expected to be very less since the selected reservoir will be tight.
- ♦ A mine where the water seepage is less is a good candidate for sequestration. Since the sequestration is planned for at least 1000 years, even with very less water seepage, significant amount of water will accumulate in the mine. With time the water level in the mine will rise and it will compress the free CO₂ present in the residual space and exert pressure on the cap rock. The pressure to be maintained in the residual space must be designed sufficiently below the hydrostatic stress so that even after compression by the rising water level the pressure of the free CO₂ shall be equal or slightly exceed the hydrostatic

stress but shall in no case exceed the leak-off pressure of the top cap layer.

- ♦ The leak-off pressure of the cap rock is critical for a sequestration project. The leak-off pressure of the cap should be well above the hydrostatic pressure.

Estimation of CO₂ sequestration potential in a mine

The amount of CO₂ that can be stored in an underground coal mine can be estimated by calculating the volume of CO₂ that can be trapped each by structural trapping, adsorption trapping and solubility trapping, the three major mechanisms discussed before.

FREE CO₂ CAPACITY

To estimate the amount of free CO₂ that will be stored by structural trapping, the residual space volume of the mine has to be estimated first. First, the mined out volume is calculated by subtracting the volume of coal recovered from the original panel volume. The residual space volume can be estimated by deducting the subsidence volume from the mined out volume. The volume lost due to the collapse of strata and the subsidence is more difficult to establish. The volume of the subsidence basins for a mine can be ascertained by using empirical equations and also verified by a conducting a surface survey. The subsidence volume will depend on among other parameters: seam thickness, depth of the seam/working, inclination of the seam, swelling factor, panel size, nature of strata (the angle of draw, geo-mechanical strength properties of the strata). The residual space volume is usually calculated empirically from the mined out volume by knowing the porosity of the residual space. The porosity in the mined out area after the formation of the goaf varies between 5-20%. The volume of the residual space is usually calculated to be of the order of 5-20% of the mined out volume (Piessens and Disar, 2003). The volume of trapped carbon dioxide will depend on: rock volume saturated by carbon dioxide, rock porosity, and the CO₂ saturation in the pores.

$$M_f = V_{res} * (1 - S_w) * \rho * P \quad \dots \quad (2)$$

where M_f is the mass of CO₂ stored in free state (kg), V_{res} is the residual space volume (m³), S_w is the water saturation (in percentage), ρ is the density of CO₂ at the reservoir temperature and pressure conditions.

ADSORBED CO₂ CAPACITY

The adsorbed CO₂ capacity in the mine depends on: the amount of coal and carbonaceous shale left in the mine, the Langmuir volume and Langmuir pressure of coal and shales, CO₂ saturation, and the reservoir pressure.

$$M_a = M * V_L * \rho * P / (P + P_L) \quad \dots \quad (3)$$

where M_a is mass of adsorbed CO₂ (t), M is the coal tonnage (t), V_L and P_L are Langmuir volume (m³/t), and Langmuir pressure (bar), P is the reservoir pressure (bar), and ρ is the density of CO₂ (t/m³).

The tonnage of coal left in the mine can be calculated from the following equation.

$$M = V_p * (100 - RF) * D \quad \dots \quad (4)$$

where V_p is the volume of the coal panel in m³, RF is the recovery factor in per cent, and D is the bulk density of coal in m³/t. The adsorption volume has to be calculated separately for the left out coals and for shale layers and then added up to determine the total adsorbed CO₂ in the mine.

SOLUBLE CO₂ CAPACITY

CO₂ is soluble in water, with solubility varying between 3% and 10% depending on pressure and temperature, salinity of the water (Bachu and Adams, 2003). Duan et al. (2006) has proposed a model to calculate the CO₂ solubility in saline water. For calculating the total soluble CO₂ in the mine, the soluble CO₂ in the accumulated mine water and that in the mine aquifer are calculated separately and then both are added. Knowing the rate of seepage of water, the total amount of accumulated water can be calculated. Amount of soluble CO₂ in the mine water can be calculated using the relationship given by Dilmore et al. (2008).

$$M_{s1} = V_w * \rho * C * M \quad \dots \quad (5)$$

where M_{s1} is the mass of CO₂ stored in the accumulated mine water (kg), ρ is the density of the CO₂ saturated brine in kg/m³, C is the concentration of CO₂ dissolved in the mine water in mol/kg, V_w is the accumulated water volume (m³), and M is the molar mass of CO₂ in kg/mol.

The amount of CO₂ that can be sequestered in the mine aquifer is given as:

$$M_{s2} = V_a * E * \phi * \rho * c * M \quad \dots \quad (6)$$

where M_{s2} is the mass of CO₂ stored in the mine aquifer (kg), V_a is the aquifer volume (m³), E is the storage efficiency factor (%), ϕ is the porosity, c is the concentration of CO₂ dissolved in the mine aquifer in mol/kg and M is the molar mass of CO₂ in kg/mol. The total mass of CO₂ trapped by solubility is given by the expression below:

$$\dots \quad (7)$$

Safety risks associated with CO₂ sequestration in mine

Storage of CO₂ in a mine although appears to be technically feasible, there are a few safety risks associated with such operations. Slow and continuous leakage of CO₂ may not lead to any environmental or safety hazards. However it is not desirable since this will lead to the failure of a sequestration project which limits the leakage to be less than 0.1% per year. In the event of any tectonic activity such as earthquake or failure of the cap rock, it may lead to massive leakage of CO₂ within a short period of time to the atmosphere. Since CO₂ is heavier than air, it will accumulate near the ground surface and further CO₂ being an asphyxiant, this may pose safety risk to the people living in the vicinity.

Summary and conclusion

Disposal of CO₂ in an abandoned coal mine appears to be promising after the successful experience of storage of natural gas in the mines. Preventing leakage of CO₂ by maintaining the cap rock stability and integrity over a large period sometimes for several decades is a big challenge. However since in the nature many CO₂ repositories exist, it is believed that mines can be good sinks. Using selection criteria potential mines can be identified and the feasibility of CO₂ storage in such mines shall be studied before any sequestration operation is carried out in practice.

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COMPUTER AIDED COAL MINE VENTILATION PLANNING

(Continued from page 57)

According to input data, the user is able to construct models and optimize them by considering duct type and diameter, shock losses, and the number, type and spacing of fans. The programme may be used for initial design, or to help trouble shoot and improve existing duct installations. It is useful in showing personnel the reasons behind a poor installation and ways to improve the system, resulting in safer working conditions.

Few computer software programmes have been designed to aid mine ventilation and environmental engineers in the prediction of the thermodynamic and psychrometric properties of air as it flows through underground airways. The

programme takes into account geothermal gradient, rock thermal conductivity and diffusivity, airflow, air quality, age of the excavation, wetness of the rock surfaces and the siting and capacity of machinery, heat exchangers or other local or disseminated sources of heat and humidity.

10. Conclusion

Softwares have proved to be essential tool in underground coal mine ventilation planning and monitoring. The views expressed in this paper are of the author and not necessarily of the organization, he belongs to.