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Ideas for storing CO_2 from the Turceni Power Plant, in closed mining areas from the Jiu Valley, Romania

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Abstract. Considering the Getica project, and the feasibility study prepared in 2011 in order to capture and storage CO_2 from the Turceni Power Plant and in view of the temporary cessation of this project, we propose a study on the storage of CO_2 in disused and closed mining areas, from the Jiu Valley, with impact on the environment and on exploitation and monitoring for long-term more than 1000 years and also alignment with similar projects in other countries, Europeans or not. Mainly, the majority of long-term capture and storage projects are carried out in deep-water aquifers, such as aquifers under the North Sea and the Barents Sea, or specially storage projects created in dissolutted salt mines, such as those in the Santos Basin in the Atlantic Ocean in Brazil, as well as others, like the pilot projects in India, with storage in volcanic rocks, etc. Storage projects in large-capacity coal mines such as those in Romania, Serbia or Bulgaria, to discuss common issues with neighboring countries, can create an exchange of knowledge with those countries on long and very long-term storage of CO_2 in coal mines, with an obvious gain in greening the atmosphere and in the health of the environment.

1 Introduction

The history of the Jiu Valley (Table 1) is related to the discovery and exploitation of coal deposits. The process of industrialization from the communist period until 1992, when were the first signs of mine closure, produced a significant migration to the Jiu Valley, with an impact on population growth. The massive restructuring followed by the closure of the mines and their preservation, negatively affected both the number of inhabitants and the number of employees in the extractive industry, the number of employees being reduced with about 75% up to 2017, compared to the existing one in 1989 [1].

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Table 1. Table of history for Jiu Valley extractive industry [1].

<table>
<tr><th>Year</th><th colspan="2">Description</th></tr>
<tr><td>1855</td><td colspan="2">The first geological explorations in Jiu Valley</td></tr>
<tr><td rowspan="2">1859</td><td colspan="2">Petrila is offcially the first mining perimeter in the Jiu Valley and the deepest coal mine in Europe</td></tr>
<tr><td colspan="2">The extensive industrialization process of the second half of the communist period has led to significant flows of migration to the mining cities of the Jiu Valley, with an obvious impact on the population growth</td></tr>
<tr><td rowspan="2">1989</td><td colspan="2">60,679 employees in the extractive industry</td></tr>
<tr><td colspan="2">The massive restructuring followed by the mine closure; negatively affected both the number of inhabitants and the number of employees in the extractive industry</td></tr>
<tr><td>2017</td><td colspan="2">3767 employees in the extractive industry</td></tr>
<tr><td rowspan="3">2030</td><td rowspan="3">Scenarios</td><td>I. inaction scenario</td></tr>
<tr><td>II. modernization of one of the mines and the gradual closure of four others by 2030</td></tr>
<tr><td>III. development of the primary, secondary and tertiary sectors in the period 2020–2030</td></tr>
</table>

The first scenario presented in the table 1 is the worst. The second scenario, including the modernization of one of the mines with the gradual closure is an interesting view for arrangement of the mines for CO_2 storage, and after that gradually closing with careful monitoring. The third scenario also is an available if the CO_2 capture is closely related to CH_4 extraction [2-4].

Before CO2 can be stored, it must be captured as a relatively pure gas. The flue gases of coal-fired power plants contain about 10-12% of the volume of CO_2 and of natural gas plants about 3-6%. In the United States, CO_2 is commonly separated and captured as a by-product from industrial processes [2]. One of the CO_2 sepparation method is that the sorbent molecule is an amine, a derivative of ammonia. The exhaust is bubbled through an amine-containing solution, and the amine chemically binds the CO_2, removing it from the exhaust gases. The CO_2 is then separated from the amine and converted back to a gas for disposal with some energy consume, up to 25 % of a plant's power-generating capacity [5]. Another technology is the membrane technology for CO_2 capture that is not as mature as conventional amine processes, but has many benefits including, simplicity of operation, modular construction, small footprint, no hazardous by-product emissions, no changes to the power plant steam cycle, and potentially lower capital and operating costs. A number of groups are working to improve different membrane processes and materials tailored for CO_2 capture [6].

2 The Getica project for Turceni Power Plant

The CCS demo project developed in Romania as Getica CCS, aims to be an integrated CCS project, covering the entire CCS chain: carbon dioxide (CO_2) capture, transport and storage. The Getica project was planned to start in December 2015 at one of the six existing units in PP Turceni, namely at Unit no. 6 of 330 MW. The optimal choice of technology in terms of post-combustion CO_2 capture technologies has been focused on the Chilled Ammonia Process (CAP) and the Advanced Amine Process (APA), as these are the furthest post-combustion CO_2 capture technologies in development, and closest to commercialization. The selected Alstom technology [6], like in the most recent results of operating 20 MWe CAP in AEP Mountaineer see the Figure 1, confirm the assumptions taken into consideration in the Feasibility Study of the Getica project. There are already implemented a number of successful pilot and demonstration units across the globe [7].

Fig. 1. The Mountaineer project aimed to achieve a minimum 90% carbon capture rate [8].

The project location was thought out to be implemented in Gorj county, in the South West Development Region, at Turceni Energy Complex, Romania. The South West Development Region comprises five counties: Dolj, Olt, Valcea, Mehedinti and Gorj. For transportation, the technology is by pipeline, which is suitable for not very long distances (maximum 100 ÷ 200 km). In order to reduce the CO_2 volume and therefore the pipeline diameter the CO_2 is transported in supercritical phase, at a pressure higher than 90 bar (abs), and at the exhaust of the carbon dioxide capture plant's compression line the pressure being commonly between 110 ÷ 120 bar (abs). The CO_2 product will have a purity of 99.7%, at 30÷40°C and 120 bar (abs) [7, 8].

3 Petrosani Basin located in the Jiu Valley

Between the Retezat and Sebeş Mountains to the N., the Vâlcan and Parâng Mountains to the S. is located the Petrosani Basin, with a maximum length of 45 km and a width of 9 km. It is crossed by the Jiu river, and it was probably formed in the Eocene by tectonic sinking. The filling deposits belong to the Paleogene, Neogene and Quaternary. Within them, several lithological complexes were separated:

a. the complex of lower red conglomerates, 200 to 600 m thick, are attributed to the Eocene and Oligocene.

b. the productive clavey-marly complex (300 - 600 m thick), includes up to 25 layers of coal, are attributed to the Oligocene - Lower Aquitanian.

c. the pebbles and torrential gravels (400 - 800 m thick) are attributed to Pliocene.

d. the conglomerate complex, (1,200 – 1,500 m thick), are attributed to Burdigalian.

Structurally, the deposits of the Petroşani Basin form a wrinkled and faulted syncline. The basin is developed on a crystalline foundation, with a discontinuous frame of Jurassic limestone and Cretaceous deposits. It has a foundation composed of crystalline schists and

Mesozoic limestones, over which appear Aquitanian, Burdigalian, Tortonian and Sarmatian-Pliocene Oligocene formations (Lupei, 1968) (Figure 2) [9].

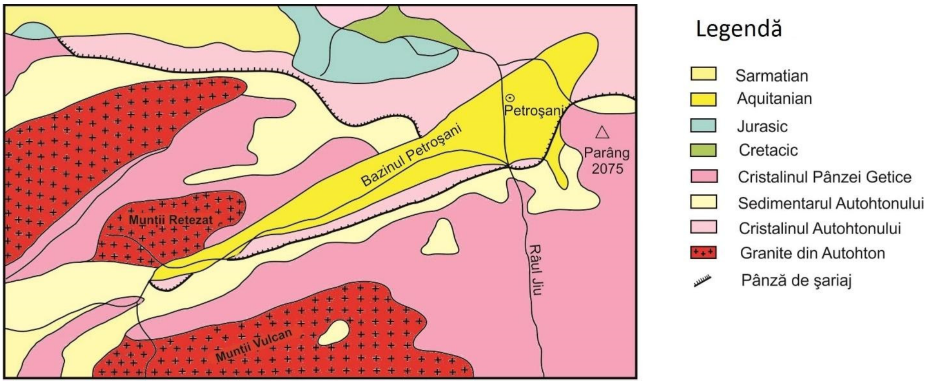


Fig. 2. Geological sketch of the Petroşani Basin (after N. Gherasi and Gh. Manolescu) [9].

The sedimentary formations on the southern flank of the Petroşani Basin rest on rocks belonging to the metamorphic series of Sebeş - Lotru (of Austrian age) within the Getic Canvas. From a tectonic point of view, the Petroşani Basin takes the form of a strongly fractured syncline, especially on the flanks. A system of major faults oriented along the basin (west-east direction) delimits the syncline so that the depression appears as a graben. A second fault system divides the sedimentary filling of the basin into numerous blocks detached from each other, both vertically and horizontally. The northern flank of the basin is represented by an inverse fault within the Cerna - Jiu system [9].

An example is the history of coal mining at Petrila opened in 1860 and officially ceased the activity in 2015, with an exploitation maximum depth of 940 m see the Figure 3 [1].

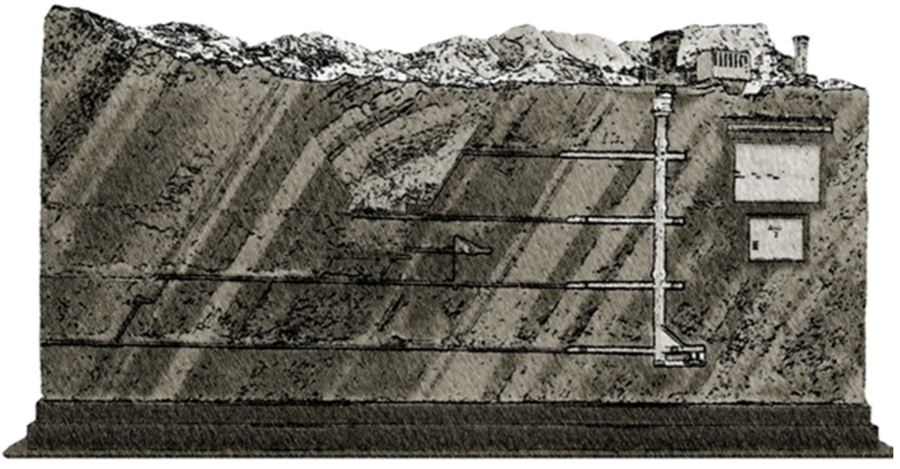


Fig. 3. Outline of the opening, preparation and exploitation works of the coal deposit from Petrila.

The study of stratigraphic columns based on results of research drilling, mapping of horizontal mining works (transversal, directional) and geo-mechanical studies related of mining fields from Jiu Valley Basin, have shown the existence of a wide variety of sedimentary rocks that have been classified into five main and distinct categories: sandstones category, clays category, marls category, marl-limestones category and microconglomerates category and the varieties of these types. The closure of the underground space consisted mainly of embankment works, filling works and watertight insulation works of concrete or masonry, resistant in time, respecting the shape and dimensions of the mining works profiles [1]. A very short, concise and easy to understand description of the phenomenon is this that in underground mines, after the extraction of useful minerals from a seam, the stresses inside the massif change which leads to the destruction of the stability of the surrounding rocks. After the stresses inside the surrounding rocks are redistributed, the rocks are set in motion and occupy the space created after the mining. In some cases the shifting of the rocks conglomerate takes place within certain limits, without affecting the integrity of the surface. Most of the times, though, the movement is transmitted to the surface, affecting it and, consequently, degrading civilian and industrial facilities situated within the mining area [2].

A technical obstacle for injection of CO_2 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 CO_2 is liquid or liquid-like. In coal mines, the initial pressure is often close to atmospheric (Figure 4). If liquid CO_2 would be injected into such a reservoir, then liquid CO_2 would evaporate and cool parts of the reservoir significantly below 0°C. The water present at the injection point freezez and blocks the subsequent injection. The freezing also may damage and collapse parts of the reservoir. Partialy the problem can be solved by customising the injection pressure. If CO_2 is transported by pipeline in supercritical phase, then it will come as "liquid" CO_2 at high pressure (110 ÷ 120 bara). A schematic example of customised injection equipment to increase the injection rate of CO_2 at a low density and so prevent freezing of the reservoir when injecting it, can be seen in the figure 4, a, b, c [10].

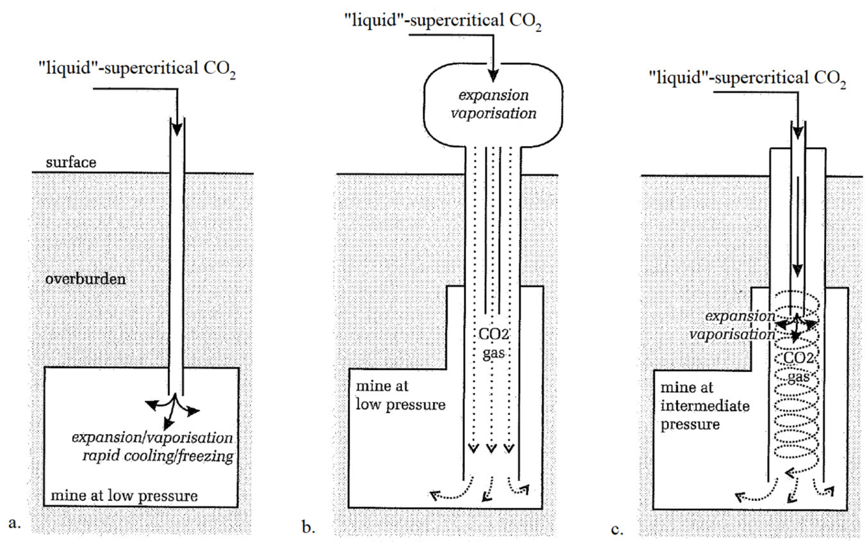


Fig. 4. Schematic example of customised injection equipment [10].

As with depleted gas reservoirs and salt caverns, CO_2 store in coal mines is inspired by storage projects for natural gas in abandoned coal mines, the oldest of which dates back to 1961. The Leyton coal mines, located near Denver Colorado, were in operation from 1903 until 1950, producing 5.4 million tonnes sub-bituminous coal from two horizontal seams at 210 m and 225 m depth in the upper Cretaceous Laramin formation. There are two other abandoned mine converted natural gas storage reservoirs, both located in the gassy Hainaut coalfield in southern Belgium. Piessens and Dusar (2003) have recently carried out a detailed feasibility study on using abandoned coal mines for long-term CO_2 storage, with special reference to a Belgian colliery, see the Figure 5, for the sites of Peronnes and Anderlues (highlighted with a circle in magenta) [10].

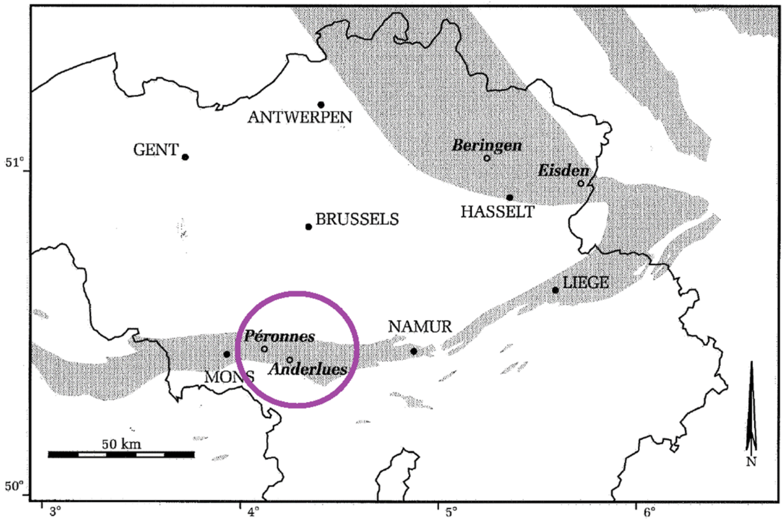


Fig. 5. Map of the outcropping or shallow surface coal basin (gray area) in Belgium [10].

4 Capacity of CO_2 storage

Coal layers considered unusable for technical or economic reasons are proving to be of major importance for the storage of carbon dioxide captured in CCS processes. Coal is characterized by the existence of a large volume of micropores to which different gases have an absorption affinity, a ton of coal being able to absorb more than 25 m^3 of methane. At the same time, coal is characterized by a higher affinity for gaseous CO_2 than for methane see the Figure 6, so that the ratio between the volumes of CO_2 and methane (CO_2/CH_4), absorbed in the coal bad can take values from 1 (in the case of anthracite), to 10 or even more in the case of lower quality coals, like lignite.

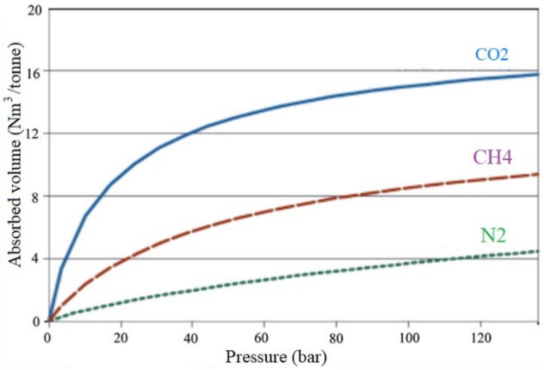


Fig. 6. Absorption curves of different gases in coal [4].

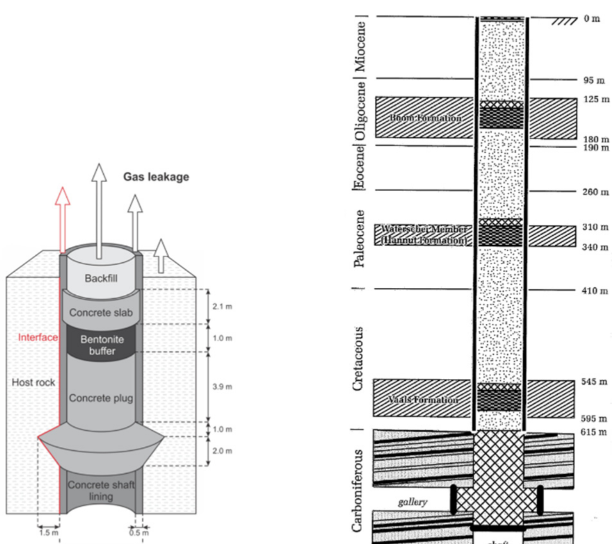
The CO_2 injected, moves through the existing crack slopes on the stratification planes and by diffusion it spreads in the coal bad, being absorbed by the micropores, releasing the existing gases with lower affinity. CO_2 also acts as a plasticizer for coal and under specific conditions of temperature and pressure, the coal changes its state, turning into a plastic environment. Both plasticization and absorption will reduce the permeability of coal, a phenomenon that can be counteracted by increasing the injection pressure [4].

There are some equations for estimation of CO_2 storage capacity in coal deposits based on the amount of CH_4 that can be extracted from coal seams. The estimates are based on the assumption that coal usually has a higher affinity for CO_2 than for methane, so when injected with CO_2, it will completely replace methane. A formula was used in the EU GeoCapacity pan-European research project, assuming that one molecule of CO_2 can replace 1.5 up to 6 molecules of CH_4 depending on pressure and depth, as well as the maturity of the coal deposit (CGS Europe Key Report 2) [9]:

$$CS_{CO2} = PGIP \times \rho_{CO2} \times ER = \left(V_{CP} \times \rho_C \times V_{CH4} \times C \times R_f\right) \times \rho_{CO2} \times ER \quad (1)$$

where: CS_{CO2} [million tonnes] - is the storage capacity of CO_2; PGIP (productible gas in place)- [Bcms] is the production capacity of coal methane; ρ_{CO2} - [g/cm^3] is the density of CO_2 under specific reservoir conditions, 0.6 - 0.8 g/cm^3; ER - [-] CO_2-CH_4 exchange rate, 1.5 up to 6 for brown/lignite coal; V_{CP} - [m^3] pure volume of coal; ρ_C - [g/cm^3] density of coal; V_{CH4} - [m^3/tonne] methane content of pure coal, 2,5 to 50 m^3/tonne, typically 5 to 10 m^3/tonne; C [%] - completion / productivity factor, up to 60%; R_f [%] - recovery factor, 20 - 80% [9].

In 2008, the Policy Support System for Carbon Capture and Storage (PSS-CCS) project analysed the opportunities of implementing carbon capture and storage in the Belgian context [9]. The supercritical storage of CO_2 would be more ideal from an economical point of view, coal mines have ideal location close to important point sources and the cost for CO_2 transport would therefore be reduced. A schematic cross section of the shaft meticulously sealed is in the Figure 7.a. Concrete filling extended over a vertical elevation of 100-200 m, close to the top of the coal measures Figure 7.b [10].



a. b.

Fig. 7. a. Layout of the sealing system used for the shaft n°6 of Anderlues coal mine. b. A schematic cross section of one ofthe shafts sealed as is typical for Campine coal mines [10].

Leakage may occur along infrastructure, such as injection wells or along geological structures, such as faults. The effect of faults on the sealing properties of the overburden (also in mining called waste or spoil) is site-specific. For example, in the Campine mine district, faults of post-Carboniferous origin are known to cross the overburden and the roof of the mines. 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 is shown in the figure 8. Calculation is done with CO_2-VR (vertical reservoir) software made by Piessens & Dusar [10]. X-axis: depth of the water table; leftmost Y-axis: amount of sequestered CO_2, right Y-axis: amount of overpressure at the top of the reservoir (600 m), in percentage relative to the hydrostatic pressure [10-20].

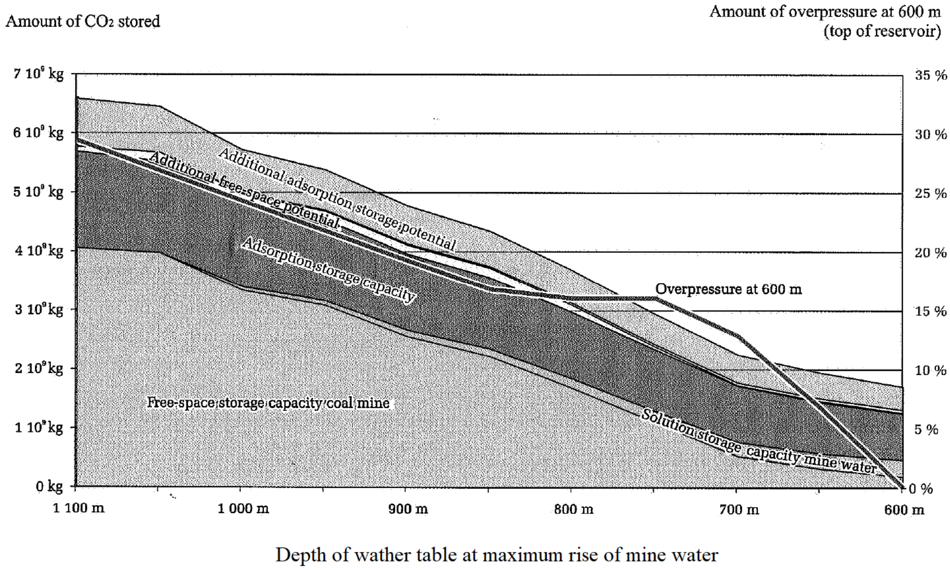


Fig. 8. The sequestration capacity of the Anderlues colliery for different amounts of overpressure, assuming that the shafts are sealed at a depth of about 600 m [10].

Piessens and Dusar (2003) have suggested some special requirements which need to be met in order to obtain a safe and stable reservoir with sufficient capacity. Firstly, the highest level of the mine should be at least 500 m deep, with well-sealed shafts and a tight, dry cap rock. Secondly, in order to prevent mine flooding, the storage pressure should be higher than the hydrostatic pressure of the surrounding strata. This overpressure, typically: hydrostatic pressure + 30% (for high depht) see the figure 8 above, require a stringent leak-proof on the top seal of the reservoir and the existing shafts [10].

5 Production of H_2 from coal mine methane

Over 95% of the world's hydrogen is produced using the steam methane reforming process (SMR). In this reaction, natural gas is reacted with steam at an elevated temperature to produce carbon monoxide and hydrogen. A subsequent reaction - the water gas shift reaction - then reacts additional steam with the carbon monoxide to produce additional hydrogen and carbon dioxide [21].

The reforming reaction with methane as:

$$CH_4 + H_2O \rightleftharpoons CO + 3H_2 \tag{2}$$

The water-gas shift (WGS) reaction is:

$$CO + H_2O \rightleftharpoons CO_2 + H_2 \tag{3}$$

From [2] Table 1, the medium of the reserves of coal in the not yet exploated mines: Lonea Pilier, Petrila, Dâlja, Aninoasa, Bărbăteni, Câmpul lui Neag and Valea cu Brazi (Figure 9), is about 110 million tones, at a medium depth of 200 meters. From the same table 1, we have a medium deposit of methane of 543 million m^3, and the medium potential of CO_2 that will replace the methane, about 16 million tones.

Fig. 9. The Jiu Valley Coal Basin [22].

If we take from [2] that the price for 1MCF methane is about \$2.00 at the well head, that is about \$56.64/1000 m^3, results \$30,755,500/coal mine and \$215,300,000 per all 7 coal mines that have to be explored. If we consider that the mine methane used for a cogeneration plant see the figure 1, and with a cogeneration efficiency at minimum of 75%, the price of the CH_4 recovered will be about \$164,475,000. The CO_2 production and injection underground, from [2] is also about 60÷70% the costs from CH_4 production, that results in a total of \$96,885,000 the value of the CH_4 extracted. If a part of CH_4 is used for hydrogen generation by reforming with CO_2 injection in the coal and if we consider the equations 2 and 3, from 1 mole CH_4 and 1 mole of H_2O we obtain 1 mole CO and 3 mole H_2, and from 1 mole CO and 1 mole H_2O we obtain 1 mole H_2 and 1 mole CO_2 results that from 16 tonnes CH_4 we obtain 8 tonnes H_2. For 0.55 tonnes/1,000m^3 CH_4, results 68,750 tonnes H_2 per 1 million m^3 CH_4. If we consider from [22] that the price for H_2 reformation is about \$1.48/kg, we can obtain a production of 1000 tonnes H_2, with a price of \$1,480/tonne, means \$101,750/1 mil m^3 CH_4 (pure)/68,750 tonnes H_2. For mine gases with 25% CH_4, the price will be greater. From [23] 9.3 kg CO_2/kg H_2 are prodused. The CO_2 should also be injected underground and the cicle is repeated.

For CH_4 recovery, a small compressor natural gas station can assure about 5,000 m^3/h natural gas flow at 45 bar (abs) [24]. In 1 year (8,000 hours) can be extracted and pumped in the National Pipe System 10 million pure CH_4 from about 40 million m^3 mine gas, at 25% vol. purity. The amount of 543 million m^3 of CH_4/coal mine, can be extracted in about 14 years that is approaching the 20 years lifetime of a high scale implemented project.

For H_2 recovery, if we consider the \$1.48/kg H_2 produced, the ROI (return of investment) is about 21.63% in 20 years [25]. In 14 years results about 15%, that is a good value for such development scale.

6 Conclusions

The concept of geological storage of CO_2 as a measure to combat climate change dates back to the 1970's, but was not much researched until the early 1990s. First initiatives in the field of geological storage of CO_2 were the storage in deep geological formations of the acid gas mixture (H_2S and CO_2 with small traces of hydrocarbons) from and separated from some oil fields in the Alberta Basin (Canada) since 1994 (Bachu and Gunter, 2005) and with the start in 1996 of separate CO_2 injection from the gas mixture extracted from the gas field Sleipner (Norway, North Sea) in the sandy Utsira Formation in the first large-scale CCS project [3].

Potential CO_2 reservoirs, implicitly in the case of the Jiu Valley coal basin, must satisfies several criteria, the essential ones being the following:

- sufficient low porosity, low permeability and high storage capacity;
- the presence of an impermeable covering rock (marl, clay, rock salt), which prevents upward migration of CO_2;
- the presence of a trap structure, in other words a structure like the dome, to control CO_2 migration within the reservoir formation;
- located at a depth of over 600 meters, where the pressure and temperature are sufficient raised to allow storage of CO_2 in the supercritical fluid phase to maximize the amount of storage;
- absence of drinking water: CO_2 will not be injected into waters that will be used for consumption and human activities [4].

The hydrocarbon processing industries (HPI) are expanding the production of hydrogen whose demand is approximately 50 million tonnes/year. The hydrogen demand is directed to many different process industries like fats and oil processing, chemicals, pharmaceutical, metallurgy, semiconductor production and aerospace industries. The investments of the capital, the operating costs and primary energy requirements should be evaluated by decision makers [26]. Because the hydrogen can be extracted also by coal gasification, results a great opportunity in reshaping the coal mining industry and transforming the Jiu Valley into green area.

Due to collaboration with University of Petroşani, National Research and Development Institute for Gas Turbines COMOTI start the developing of the storage projects in depleted gas deposits or in depleted or conserved coal or salt mines. The activities of the Research Laboratories of the Petroşani University, are aimed at developing topics with fundamental and applicative impact, in integrative, multidisciplinary and interdisciplinary approaches, in accordance with the priority issues at regional, national and international level, which will ensure the increase of visibility and competitiveness.

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