Technical paper

The Emissions of CO_2 as the Chance for the Sustainable Management With Natural Sources

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Abstract

According to the regional and wider national strategic goals concerning the use of natural resources and electricity production in the future, the sustainable aspect of the exploitation of the nonrenewable lignite stock is emphasized. We also discussed (quantitativly evaluated) the potentially produced electricity and the potentially emitted CO_2 as one of the main environmental issues regarding the Kyoto document when the researched lignite deposit would be used in the nearest Thermal power plant (TPP) Šoštanj.

In 2004 an experiment was performed on sequestration of carbon dioxide (CO_2) in the coal seam. The aim of the experiment was to estimate the suitability of the coal bed for storage of CO_2 released during combustion of coal and the usage of subsequently released CH_4. The wells were 3000 m far from the mine entrance. The first three wells were 6 m apart from each other, the other three 3 m. In the wells the amounts of CH_4, CO_2, dimethylsulfide, oxygen, carbon monoxide and hydrogen, as well as flow, humidity and temperature of desorbed gases were measured. CO_2 was sequestered in the second and fifth well for four hours. In the remaining wells desorbed gases were sampled in 2 l tedlar bags. During sequestration of CO_2 gases were sampled from the wells every half hour. Eight weeks after the experiment was performed, the composition of gases was remeasured.

Keywords:

1. Introduction

The sequestration of CO_2 in coal seams is seen as a possible way to mitigate the rising atmospheric concentrations of CO_2 (Reichle et al. 1999). One of the earliest studies of the adsorption of CO_2 on coal used BET equation to calculate the CO_2 surface areas of anthracites (Walker and Geller 1956). That the diffusion of CO_2 through coals of various ranks is an activated process was established not long afterward (Nandi and Walker 1956). The adsorption of CO_2 and other gases, especially methane, has also been studied in efforts to increase the safety of coal mining (Kim and Kissell 1986). Recently, CO_2 adsorption on coal has been studied as a means of enhancing the gas production in coal bed methane projects. Burlington Resources, the largest producer of coal-bed methane, has been injecting CO_2 in the USA' San Juan basin, New Mexico to enhance methane production since 1996 (Stevens et al. 1998).

The Velenje coalmine implemented a project of Clean Coal Technology (CCT). The possibility of using coal bed methane (CBM) is one of these CCT projects. Research has been carried out starting from the year 2001. In 2001, the content of methane was measured on existent vertical wells from the surface at the Velenje coalmine. In 2002, an estimation of quality and quantity of emitted gas from horizontal and cross-measure wells in the coalmine was performed. Gas samples were taken from three different locations: coalseam at the G plate, during mining at area –20a and at collapsed area –90c.[4] In the year 2003 the measurements of gas quality from horizontal wells at the G plate and at the collapsed area were continued. The gas was of medium quality and methane content was between 33 and 70 %.[5] In 2004 an experiment was performed on sequestration of carbon dioxide in coal seam. For the experiment wells 3242-II/04 and 3243-II/04 on the G plate were used. The composition of desorbed gases was measured from wells 3242-I/04, 3242-III/04, 3243-I/04 and 3243-III/04. The aim of the experiment was to check the possibilities of the sustainable usage of the emitted CO_2: to estimate the suitability of the coal bed as the nonrenewable source for storage of CO_2 released during combustion of coal and the usage of subsequently released CH_4.[6]

Regarding safety hazards of the 2004 experiment the experts were present due to take some measures – temporary or permanently stop the experiment if it would be necessary. There were expected the following equipment and experiment irregularities: damaged well/valve, disproportionate pressure increase in the well, raised content of CO_2 at the well entrance.

2. Experimental

Whereas in the y. 2003 the natural content of methane from the horizontal wells at the G plate, mining area and collapsed area were measured (the samples showed 61 % of CH_4 in the area of G plate, 68 % of CH_4 in the mining area and 51 % of CH_4 in the collapsed area), in the 2004 experiment the sequestration of carbon dioxide (CO_2) in coal seam was performed. The main goals of the research were to find out (1) the difference between the adsorption affinity of CH_4 and CO_2 and how CO_2 helps to desorb CH_4 from the coal, (2) to measure CH_4 concentration during experiment comparing to previous research, that would help us (3) to estimate the gas quality.

The wells of 30 m length were 3000 m far from the mine entrance. The first three wells of diameter were 6 m apart from each other, the other three 3 m. In the wells, which incline was 40, the amounts of CH_4, CO_2, dimethylsulfide, oxygen, carbon monoxide and hydrogen, as well as flow, humidity and temperature of desorbed gases were measured. Working pressure of CO_2 was 10 bar and the amount of sequestrated CO_2 was 75 kg to each well.CO_2 was sequestered in the second and fifth well for four hours. In the remaining wells desorbed gases were sampled in 2 l tedlar bags. During sequestration of CO_2, gases were sampled from the wells every half hour. Eight weeks after the experiment was performed, the composition of gases was remeasured. In Figure 1, the situation in the mine where the experiment of CO_2 sequestration was performed is presented (G plate).

The collected gases during the experiment and the calibration gases were analysed under the same conditions in the ERICo laboratory. The calibration curves were linear ($R^2 >$ 0,99); the limits of quantification were 0.01% for CH_4 and CO_2, 1.0 ppm for dimethylsulfide (DMS) and hydrogen (H_2), 1,0 ppm for carbon monoxide (CO), and 1,0 % for oxygen (O_2). The measured components were determined by methods validated under ISO 17025 requirements.[7]

2. 1. GC Analysis

The gas samples were analysed using a Perkin Elmer AutoSystem XL chromatograph. A 100-L loop connected to a 6-port gas sampling valve for injection of gas samples was used. Behind the injector, the sample was delivered into two capillary columns of the same dimensions. CO_2 and CH_4 were separated on a Carboxen 1006 PLOT capillary column, 30 m in length (530 ìm id.), connected to a Flame Ionisation Detector (FID) equipped with a Zr/Ni catalytic reactor. The catalytic reactor reduced CO and CO_2 to CH_4, which could be detected at considerable lower concentrations by FID. A Supel Q PLOT capillary column was used for separation of DMS. This column was connected to a Flame Photometric Detector (FPD), which is a highly sensitive and selective detector for sulphur compounds and is appropriate for detecting low concentrations of DMS. The other hydrocarbons and sulphur species were not determined at the operating conditions, which were as follows:

- Carrier gas: helium flow: 8 ml min^{-1}
- Gas sampling valve on: 0.01 min to 0.05 min
- Oven temperature (start): 50C, 1 min
- Heating rate: 18C min^{-1} to 170C
- Detectors: FID with catalytic reactor ZR/Ni, FPD
- FID: Air flow: 400 ml min^{-1}, Hydrogen flow: 40 ml min^{-1}
- FPD: Air flow: 100 ml min^{-1}, Hydrogen flow: 75 ml min^{-1}

Messer's secondary gas standards of various concentrations (as expected in our samples) for each gas component were used for qualification and quantification.

2. 2. Electrochemical Analysis

Concentrations of CO, H_2 and O_2 were measured by electrochemical sensors, built into a special device, made by the ECHO Company, Slovenia. With this device it is possible to measure concentrations of CO and H_2 from 1 to 500 ppm and O_2 from 1,0 to 30 %.

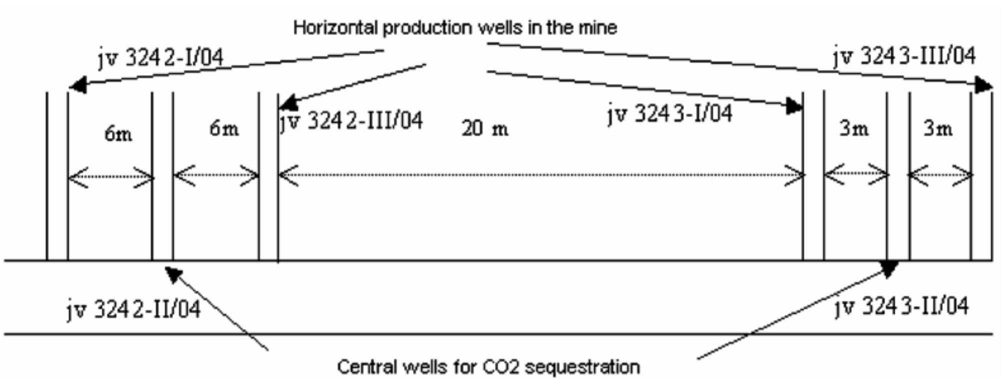

Figure 1: Situation in the mine at the G plate, where CO_2 sequestration was performed.

3. Results and Discussion

In gas desorbed from the wells after sequestering CO_2, the amount of methane reached the value of 78,2 %, which is essentially higher than in previous research, when the amount of methane was on average between 55 and 65%. It was shown in the experiment that the methane concentration increased during the experiment while the CO_2 concentration remained at the same level (18 %).

During the CO_2 sequestering the gas flow from the production wells was between 24 and 33 l/h. In the weeks after sequestering the flow from the production wells was in general lower (22 l/h) than it was on the day of CO_2 sequestering.

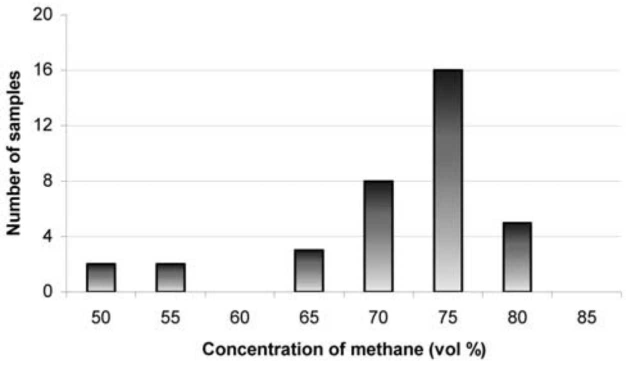

Figure 2: The number of samples, containing methane in the interval from 50 to 80 % during CO_2 sequestering in the wells jv 3242-II/04 and jv 3243-II/04.

In Figure 2 the numbers of samples with a certain concentration of methane are presented. The most frequent methane concentration was from 70 to 75 %. In the previous investigations (in the period 2001–2003) only three samples reached this concentration level on the G plate.

The average concentration of methane during CO_2 sequestering was 71% and 69% eight weeks after sequestering. The average content of methane was higher in this experiment than it was in previous investigations, where the average content of methane was 61%.

In the wells which were at a distance 3 m from the central well where CO_2 was sequestered, higher individual (78 %) and average concentrations (74 %) of methane were determined in comparison to those wells, which were at a distance of 6 m from the central well (75 % in the individual samples and 70 % on average). The measurements confirmed the data from literature, where the optimal distance between wells from the central well is 3 m.[8]

The higher concentrations of methane in released gases were the consequence of CO_2 sequestering in the coal seam. Regarding the fact that a released molecule of methane needs two to three molecules of carbon dioxide[9] and 150 kg of the sequestered amount of carbon dioxide, between 18 to 27 kg (25–40 m^3) of methane were released. From the results determined in the experiment, 10–15 kg of methane were daily released from wells jv 3242-I/04, jv 3242-III/04, jv 3243-I/04 and jv 3243-III/04. If the released amount of methane would remain at the same level for the whole year it would mean 3,5–5,5 t of released methane of medium quality.

The average concentration of carbon dioxide was 17 % during sequestration, which increased to 18 % in the two-month interval after sequestration. The determined concentrations of CO_2 and O_2 from the four wells in the experiment are comparable to the CO_2 and O_2 concentrations from the investigations from the G plate in previous years[5].

4. Conclusion

The sequestration of CO_2 in the coalmine is one of the possibilities for CO_2 usage. In the year 2004 an experiment was performed on sequestration of carbon dioxide in coal seam. While CO_2 has a higher adsorption affinity than CH_4, it helps to desorb CH_4 from the coal. For the experiment, wells 3242-II/04 and 3243-II/04 on the G plate were used. The composition of desorbed gases was meas-

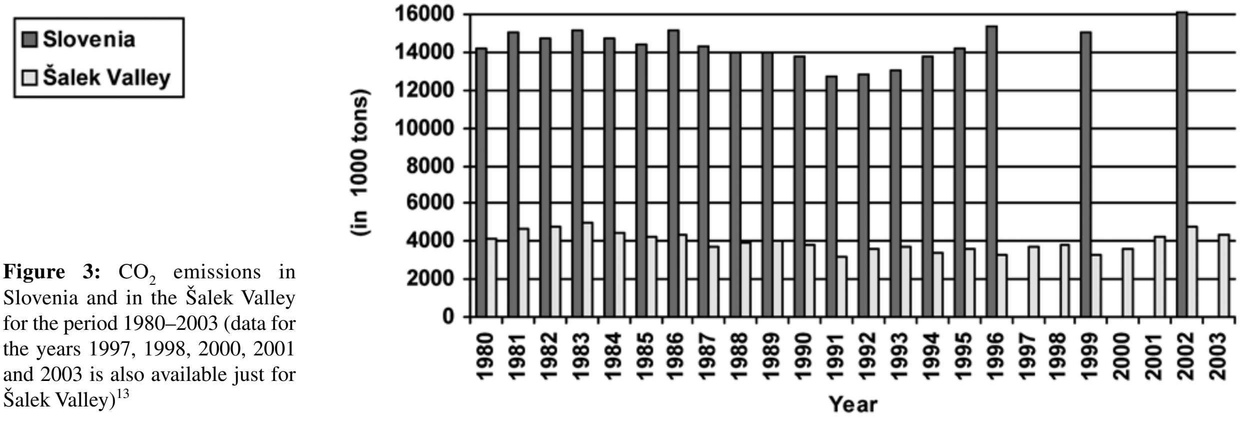

Figure 3: CO_2 emissions in Slovenia and in the Šalek Valley for the period 1980–2003 (data for the years 1997, 1998, 2000, 2001 and 2003 is also available just for Šalek Valley)[13]

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ured from wells 3242-I/04, 3242-III/04, 3243-I/04 and 3243-III/04. The amount of methane was between 60 and 78%, which is essentially higher than in previous research. Gas desorbed from these four wells was still of medium quality and can be used for electricity production or for space and water heating on-site. When Thermal power plant Šoštanj will install a gas turbine, the degasification system in Velenje coalmine will be an interesting option due to coal bed methane use in Thermal power plant Šoštanj.

The other possibilities for CO_2 usage are also in the food and chemical industries, in agriculture, for extinguishing fire, for production of dry ice or it can be used as a solvent at supercritical conditions.

CO_2 is the most important anthropogenic greenhouse gas. Over the past 2000 years its concentration in the atmosphere has increased from 0,0275% to 0,0370%, mostly as a result of the combustion of fossil fuels.[10] Šoštanj power plant (PP) as the largest Slovenian thermal power plant in the vicinity of Velenje coalmine is known to be the biggest source of CO_2 emissions in Slovenia (Figure 3). Since approximately 4 Mt of the lignite coal per year is used in Šoštanj PP it emits almost the same amount of CO_2 every year. Although they have already installed the desulphurisation devices, which decreased the sulphur pollution by 90% the problem of NOx and especially the CO_2 emissions remains.

Slovenia has ratified the Kyoto protocol regarding the United Nations Framework Convention on Climate Changes in y. 2002. With the ratification we agreed on decreasing GHG emissions in the period 2008–2012 by 8% compared to the 1986 emissions. In 1999 CO_2 presented 80% of GHG emissions, mostly because of fossil fuels usage. Although the Slovenian government has passed a law on Order about taxes for air pollution with CO_2,[14] the data for 2002 show an increase in CO_2 emissions (Figure 3). The CO_2 emissions licence trading in Slovenia has already started in 2005 in the appropriate manner. So the polluters in Slovenia have to pay already the real price for emitted CO_2 and Slovenia is short of coupons at the moment; since the price for emitted CO_2 at European emissions' licence exchange varies a lot[15], the above presented data and results are of great national importance. Slovenia has already accepted the external costs' concept in energy management, since the sustainable development paradigm is being followed. The ecosystem's goods and services should not be free of charge.

5. Acknowledgement

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Povzetek

Glede na regionalne in širše, nacionalne strateške cilje (NEP) o trajnostni rabi naravnih virov, povezani s proizvodnjo električne energije v prihodnosti, je v prispevku poudarjen sonaravni vidik uporabe lignitnih zalog v Premogovniku Velenje (PV). Prav tako smo upoštevaje načela sonaravnega ravoja (in sporazuma iz Kyota) ovrednotili okoljske pritiske, ki bi nastali z emisijami CO_2 ob potencialni izrabi preučevanih zalog lignita za proizvodnjo električne energije v bližnji TE Šoštanj.

S poskusom vtiskanja CO_2 v premogov sloj l. 2004 smo ugotovili ustreznost sloja za shranjevanje CO_2, ki se sprošča pri izgorevanju premoga, in posledično uporabnost sproščenega metana (CH_4) kot energetskega vira. Vrtine so bile 3 km oddaljene od vhoda v glavni jašek; prve tri vrtine so bile 6 m vsaksebi, ostale tri pa 3 m. V njih smo merili koncentracije CH_4, CO_2, dimetilsulfida, kisika, ogljikovega monoksida in vodika, prav tako pa tudi pretok, vlago in temperaturo desorbiranih plinov. CO_2 smo štiri ure vtiskali v drugo in peto vrtino, v preostalih vrtinah pa vzorčili desorbirane pline.