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SYSTEM OF ADSORPTION OF CO_2 IN COALBED

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ABSTRACT

Carbon capture and storage (CCS) has been observed as an alternative measure to mitigate emissions from greenhouse gases. CCS systems separate CO_2 during the process of converting fuel and transport to the facilities where they are stored, e.g., under geological formation conditions. Capture and sequestration technologies are now widely used in different industries. CO_2 capture is currently a costly and energy-consuming technology. The costs obviously depend on the size of the plant and the type of fuel used. Generally, capture systems are categorized into three categories: pre-combustion, post-combustion, and oxy-fuel combustion. The saline aquifer, depleted oil, and gas fields are large-capacity storage sinks. The coalbeds also provide as a substitute to geological storage. One of the main advantages of coal storage is renewable methane fuel and coal desulfurization. The studied coal reserves are located in Candiota, State of Rio Grande do Sul, Brazil. These are the largest coal deposits in the country, with a reserve of 1 billion tons. This work will represent a study on CO_2 storage in Candiota coalbed system. The use of a synthetic CO_2 cylinder with a flow of 0.2 L /min varies the time of contact with the coalbed. The results from the volatile matter increase by 11%. The results of the ultimate analysis exhibited an 8% increase for carbon and oxygen after 60 minutes of CO_2 flow in the coalbed. On the other hand, there was a reduction of 32% for sulfur. The resultant phenomena occur due to the adsorption capacity of CO_2, where the compounds are released from the pores of coal.

Keywords: *Greenhouse Gases, Climate Change, Candiota Coalfield, Carbon Capture, and Storage.*

1. INTRODUCTION

The burning of fossil fuels emits a substantial amount of greenhouse gases (GHG), where carbon dioxide (CO_2) is most abundant (Chen *et al.,* 2017). The emission of this gas into the atmosphere causes the natural phenomenon of heat retention on Earth's surface to be aggravated, causing climatic changes (Fernández-Sánchez *et al.,* 2015). These changes in climate, in turn, activate a set of changes related to the operation of ecosystems (Kim and Kim, 2016; IPCC, 2014). As a procedure of mitigating environmental impacts, the geological, oceanic and biological storage of CO_2 are technological alternatives that have been discovered. Geological storage is the process which follows the capture and transport of CO_2 (Man *et al.,* 2018).

Capture and transport take place from the source to reservoirs which are capable of retaining CO_2 for hundreds or thousands of years. This technology system is called Carbon Capture and Storage (CCS). Carbon capture and storage systems occur with separation of CO_2 during the process of energetic conversion of fossil fuel to different geological formations (IPCC, 2014). The reservoirs currently used are depleted oil and gas fields, saline aquifers and Carboniferous formations (Agartan *et al.,* 2018; Hosa *et al.,* 2011; Qi *et al.,* 2009; Hassanzadeh *et al.,* 2008). The use of carboniferous formations as a reservoir occurs when the coal is no longer operable and the extraction of methane (CH_4) adsorbed in the pores of the geological formation and desulfurization of the coal with high SO_2 contents due to the presence of pyrite is desired (Shogenov *et al.,* 2017; Xia and Xie, 2017; Borah and Baruah, 2001).

In order to achieve the storage of large volumes of CO_2, the geological formation requires to have adequate permeability (Jiskani *et al.,* 2018; Marcisz *et al.,* 2018). The logistics of CO_2 capture, transport, and storage are not trivial, and risks of leakage are involved at all stages (Deng *et al.,* 2014; Keller *et al.,* 2008). The characteristics of the reservoir and possible escape mechanisms of CO_2 must be carefully monitored to avoid catastrophe in the environment (Deng *et al.,* 2014).

This particular work aims to study a CO_2 adsorption system in coalbed in order to properly capture and store CO_2 in order to sustain for a long inactive period of time. The coals of Candiota, State of Rio Grande do Sul, Brazil, were utilized for this purpose. In a system simulating coalbeds, CO_2 could be stored. This is evident in the results of proximate and ultimate analyses where there was an increase of 11% in the volatile matter and a decrease of 32% in sulfur contents.

2. CO_2 CAPTURE, TRANSPORTATION AND STORAGE TECHNOLOGIES

Carbon is an essential chemical element of life. It occurs in nature as graphite and/or diamond, it is also associated with other atoms such as carbon dioxide, coal, methane, etc. (Reiter and Lindorfer, 2015). The development of modern society is based on activities which provoke an increase in carbon. In this context, we can mention: soil use (agriculture, forest burning); ii) industrial activities (petrochemical); iii) the burning of fossil fuels (coal, oil and natural gas) for generating power (Koytsoumpa *et al.,* 2018).

As there are no natural mechanisms to convert carbon at the same rate as it is transferred, CO_2 ends up concentrating on the atmosphere. This increase in CO_2 concentration causes the greenhouse effect in the atmosphere (Huaman and Lourenco, 2015). Other gases are also responsible for this disastrous effect on the atmosphere, which includes nitrous oxide (N_2O), methane (CH_4) and chlorofluorocarbons (CFC's). Among GHGs, CO_2 possesses the maximum concentration (approximately 75%) in the atmosphere (Deng *et al.,* 2014).

2.1. CO_2 capture

Generally, capture systems are classified into three categories: pre-combustion, post-combustion, and oxyfuel combustion.

2.1.1. Pre-combustion capture

The technology for pre-combustion is extensively applied for chemical, gaseous fuel (H_2 and CH_4), fertilizer, and also for power production (Mantripragada and Rubin, 2017). In these particular cases, the fossil fuel is partially oxidized in a gasifier. CO from the derived syngas (CO and H_2) reacts with additional steam (H_2O) and is thereby converted into CO_2 and H_2 (Zheng *et al.,* 2017). The resultant CO_2 can be captured from a comparatively pure exhaust stream. H_2 can further be used as fuel, whereas, removal of CO_2 takes place before combustion. There are various advantages and disadvantages when this method is compared to conventional post-combustion carbon dioxide capture (Kanniche *et al.,* 2010). In this method, CO_2 is removed after combustion of fossil fuels, but before the expansion of flue gas in the atmospheric pressure. This method is applied to contemporary fossil fuel burning power plants, or to existing plants where re-powering is an alternative (Park *et al.,* 2015). The capture which occurs before expansion, i.e., from pressurized gas, is the standard in almost all the industrial CO_2 capture processes at the same scale as required for utility power plants.

Pre-combustion capture also has the advantage of excessive control of CO_2 emissions (90 to 95% of CO_2 is captured). It involves low technological risk and also has the possibility of becoming the most efficient method to mitigate CO_2 emissions (Ryu *et al.,* 2017).

2.1.2. Post-combustion capture

Post-combustion systems retain CO_2 capture after the burning of fossil fuel (Wu *et al.,* 2014). In this case, CO_2 is captured from flue gases at several power stations or other massive point sources. The technology is well comprehended and is presently used in distinct industrial applications, although at a different scale than required in a commercial scale power station (Moser *et al.,* 2013). Post-combustion capture is most accepted for research purpose since the existing fossil fuel power plants can be retrofitted for including CCS technology in this specific configuration (Singh, 2013). There are several methods which can be applied to capture CO_2 in post-combustion processes (Jin *et al.,* 2018). One significant method is the separation of the gases produced and emitted into the atmosphere through the absorption process (Plaza *et al.,* 2017). This includes a mass transfer between the gas (combustion gas) and liquid (solvent) phases, which exploits the differences in the gas-liquid solubility of distinct components of the treated mixture. Another post-combustion capture process is adsorption, which involves utilization of a solid surface, such as activated carbon or zeolites to adsorb CO_2 present in a particular gas mixture (Moser *et al.,* 2013. Separation of CO_2 by membranes is another significant process associated with capture. The membranes cannot separate the CO_2 with the required efficiency in only one step. It is, therefore, necessary to repeat the process until a suitable degree of purity has been achieved. This is responsible for increasing the complexity associated with increasing energy consumption and costs in the system (Jin *et al.,* 2018).

2.1.3. Oxy-fuel combustion

In oxy-fuel combustion method, the fuel is burned in oxygen instead of air (Dwivedi *et al.,* 2018). The cooled flue gas is re-circulated and then injected into the combustion chamber to limit the resultant flame temperature up to the levels of conventional combustion (Hu *et al.,* 2018). The flue gas consists of mainly carbon dioxide and water vapor, where the latter is condensed through cooling (Leffler *et al.,* 2017). The outcome of the process is an almost pure carbon dioxide stream, which can be conveyed to the sequestration site and stored. Power plant processes associated with oxy-fuel combustion are frequently referred to as "zero emission" cycles. In these processes, the preserved CO_2 does not occur as a fraction removed from the flue gas stream (like in pre- and post-combustion capture methods), but the overall flue gas stream itself (Carrasco-Maldonado *et al.,* 2017). A certain fraction of the generated CO_2 during combustion will inevitably end up as a constituent of the condensed water. The water has to be treated or disposed of appropriately for authorization of the “zero emission” label. The oxy-fuel combustion technique is promising, but the initial step of air separation requires a lot of energy (Chen, 2018).

2.2. CO_2 transport

CO_2 transport is the least complicated process in the CCS chain, as it is an existing technology for which relevant costs can be estimated (Goldthorpe and Ahmad, 2017). The principal complication with CO_2 transport is that this compound behaves in different ways when subjected to variations in pressure and temperature (Jakobsen *et al.,* 2017). Thus, the transfer of CO_2 has to be carefully managed to prevent solidification and occurrence of blockages. There are two effective methods for transporting massive volumes of CO_2: pipeline transport and ship transport. Transportation is also possible through trucks with CO_2, usually in liquid form. However, these two solutions are not feasible for substantial quantities (Engel and Kathe, 2017).

2.3. CO_2 storage

In several places of the world, CO_2 is trapped in carboniferous formations (He *et al.,* 2011). The idea of geological storage further seeks to reproduce such natural phenomenon which attests to the great potential that geological formations have for storage of gases (Shogenov *et al.,* 2017).

Several aspects need to be considered when it comes to CO_2 storage: (i) the storage period should be extensive, preferably hundreds or thousands of years; (ii) the cost of storage, including transport from the place of capture, injection, and monitoring, should be minimized; (iii) the applied methods may not violate any national or international law or regulation; (iv) the environmental impacts need to be carefully considered, and their minimization techniques should be assessed; (v) the probability of occurrence of accidents should be practically null (Zhao *et al.,* 2016). Coal is the sedimentary rock which results from the decomposition of organic matter and is generally found in veins or layers (Maphala and Wagner, 2012).

If CO_2 is injected into coal deposits, CO_2 will release the trapped CH_4 therein and fill the voids where it was located. Based on laboratory studies, it is estimated that a volume of CO_2 can be stored twice as large as that of CH_4 initially adsorbed (Chai and Shimada, 2011). Since this technique leads to an improvement in the yield of a deposit in terms of collected natural gas, it became popular as Enhanced Coalbed Methane Recovery (ECBMR) (Fan *et al.,* 2018). ECBMR is a more efficient method for capturing more than 90% gas than the traditional methods for CH_4 collection from coal which captures about 50% of the gas (Vishal *et al.,* 2018; Ma *et al.,* 2017).

3.2. MATERIALS AND METHODS

3.1. Coal geology

Coal is a biogenic sedimentary rock, which is easily combustible and is brown to black in color. It is derived from the decomposition of accumulated debris in a basin from the atmosphere, on which sediments were deposited (Wang *et al.,* 2018). The formation consisting of the coal goes through an initial phase of slow biodegradation of the accumulated material by anaerobic microorganisms (Fuertez *et al.,* 2017). Both biochemical and thermodynamic processes occur with the successive deposition of inert gases on this organic sludge (Davis and Gerlach, 2018). Dehydration is accompanied by impoverishment in the volatile matter, along with loss of oxygen and hydrogen. The resulting mass, enriched in carbon, decreases in volume, and increases in density due to the combined action of pressure and temperature (Wang *et al.,* 2018).

The constituents of coal which can be distinguished microscopically are called macerals. The term “maceral” is analogous to the use of the term mineral while speaking of igneous or metamorphic rocks. However, unlike minerals, macerals have no characteristic crystalline form and are not constant in their chemical composition and physical properties. One of the most common macerals is vitrinite, which is derived from woody materials (Zieger *et al.,* 2018). Vitrinite is considered to be the most important maceral due to its extraordinary adsorption power of CO_2, causing a greater increase of volume of the same and, consequently reducing the permeability of coal (Zieger *et al.,* 2018).

The proportion in which the different macerals present themselves, as well as the level of their maturation, have consequences on the chemical, physical, and mechanical properties of the coal. Therefore, there occurred a requirement to classify the various types of coal. One of the most widely used concepts is the rank of coal. This is an important feature as it is widely used by coal-burning industries to infer the calorific value of the fuel (Fuertez *et al.,* 2017).

The degree of coal is estimated by a specific energy, evaluation of the water content, percentage of volatile matter or reflectance of vitrinite. A single parameter is not sufficient to classify the different grades of coal, and however, sometimes more parameters are used for one rank than others. The low-rank coals are generally characterized by the heat energy value and the water content, whereas the high-rank coals are distinguished using the reflectance values of vitrinite, the presence of fixed carbon and the percentages of volatile matter (Zieger *et al.,* 2018).

3.2. Location of the study area

The study area is located in the city of Candiota, at the southern end of the State of Rio Grande do Sul, Brazil. The Presidente Médici thermoelectric power plant belongs to Eletrobrás - Companhia de Geração Térmica de Energia Elétrica SA (CGTEE) and is provided by Companhia Riograndense de Mineração (CRM), which has reserves of $1,419.41 \times 10^6$ tons (CRM, 2010) and a production of 1,600,000 tons per year of ore to meet the needs of the plant.

3.3. Preparation of coal samples and CO_2 injection system

The coal samples were inserted into a system (Figure 1) and connected to CO_2 gas (from Martins PRAXAIR INC. WHITE^{TM}) with a concentration of 5000 ppm packaged in the cylinder, with a purity of 99.95%. For the experiments, a flux of 0.2 L/min was used with contact intervals of 5, 10, 15, 30, 45, and 60 minutes in the coalbed. Proximate analysis and ultimate analysis were measured before the tests and after the contact intervals between the CO_2-Coal interactions.

4. RESULTS AND DISCUSSION

The results of proximate analysis (Table 1) reveal that CO_2 injected into the coal changes its properties. The moisture and ashes underwent an increase of 5% after 60 minutes of the reaction. Also, an overall increase of 11% was observed for the volatile matter and fixed carbon. The ability of coal to adhere CO_2 releasing hydrocarbons manifests such behavior. The greater amount of CO_2 in contact with the charcoal bed produces greater ash content. The volatile matter has increased in content as a consequence of the increased amount of CO_2.

The ultimate analysis (table 1) results in an 8% increase for carbon and oxygen after 60 minutes of CO_2 flow in the coalbed. For sulfur, a decrease of 32% has been observed. This is due to the displacement capacity of sulfur in porous layers of coal. There was coal desulfurization with the increase of CO_2 flow during 60 minutes of contact.

5. CONCLUSIONS

The present study addressed the characteristics of the CO_2 capture and storage technique. In this context, a CO_2 injection scheme was presented in coalbed, and the physicochemical properties were verified after the experiments. It was established that according to the time of contact with the CO_2 flow, there is an increment of 5% of moisture and ash content and 6% of carbon and oxygen, in the coal. Subsequently, for the volatile matter and fixed carbon, there is a decrease of 7 and 11% respectively. The desulfurization caused by the injection of CO_2 into the coal where there was a 32% decrease is another interesting discovery.

Therefore, the results indicate that the proposed coalbed system was efficient in the adsorption of CO_2 and might act as an alternative for the removal of SO_2 in the coal.

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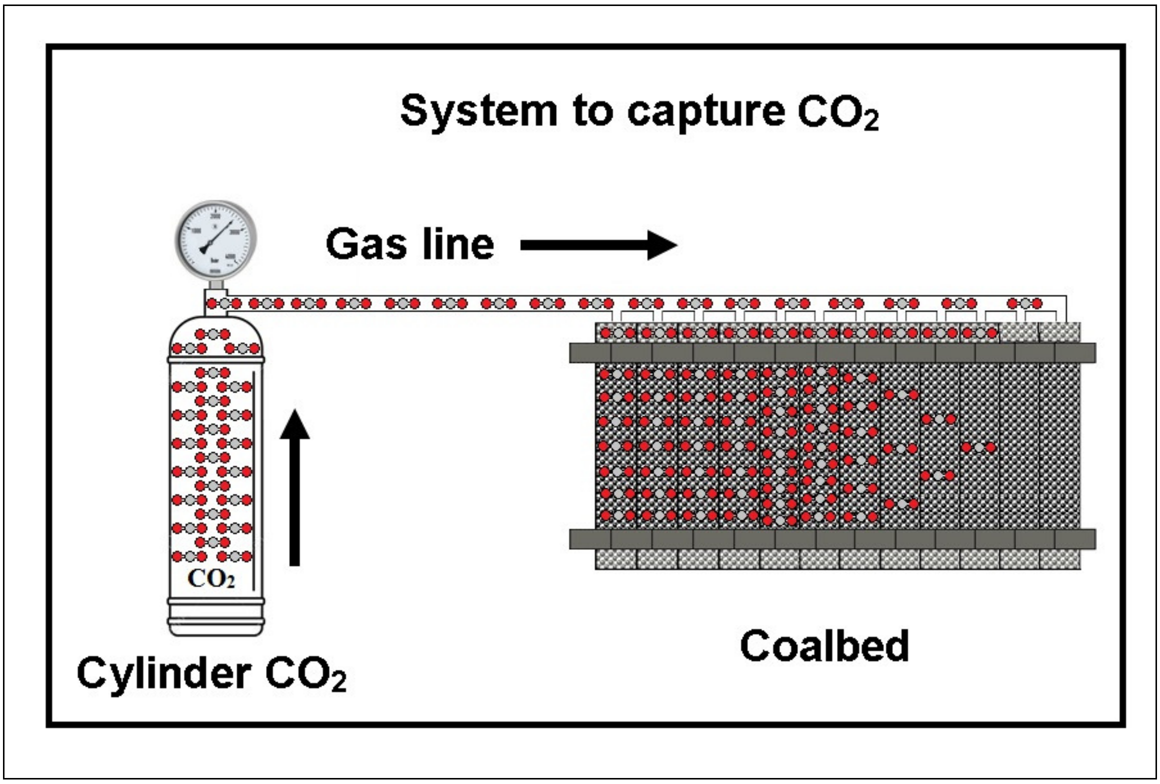

Figure 1. *Schematic system of capture CO_2 in coalbed*

Table 1: *Properties of the coal before and after interaction with carbon dioxide.*

Proximate analysis - Weight (%) on dry basis					
Samples	**Moisture**	**Ash**	**Volatile matter**	**Fixed carbon**	
CF	8.4	52.9	51.2	24.9	
C-5	8.7	53.4	51.7	25.7	
C-10	8.8	53.8	52.4	25.9	
C-15	8.8	54.2	52.6	26.3	
C-30	8.9	54.9	53.1	26.7	
C-45	9.1	55.3	55.9	27.5	
C-60	9.4	55.8	56.8	27.9	
Ultimate analysis - Weight (%) on wet basis					
Samples	**Carbon**	**Hydrogen**	**Nitrogen**	**Oxygen**	**Sulfur**
CF	25.4	4.9	0.9	14.9	2.5
C-5	25.7	5.0	0.9	15.1	2.3
C-10	26.0	5.2	0.8	15.3	2.0
C-15	26.3	5.2	0.8	15.4	1.8
C-30	26.8	5.3	0.8	15.7	1.6
C-45	26.9	5.3	0.8	15.9	1.4
C-60	27.3	5.3	0.8	16.1	1.1

CF (Coal free of CO_2 flux); C-5 (CO_2 flux in coalbed of 5 min); C-10 (CO_2 flux in coalbed of 10 min); C-15 (CO_2 flux in coalbed of 15 min); C-30 (CO_2 flux in coalbed of 30 min); C-45 (CO_2 flux in coalbed of 45 min); C-60 (CO_2 flux in coalbed of 60 min).