Carbon Dioxide Capture from Power Plant Flue Gas using Regenerable Activated Carbon Powder Impregnated with Potassium Carbonate

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Abstract

Adsorption is considered one of the more promising technologies for capturing CO₂ from flue gases. This research shows an efficient chemical adsorption method capable of capturing carbon dioxide under moist conditions from flue gases of coalfired power plants. Carbon dioxide was chemically adsorbed by the reaction $K_2CO_3*1.5H_2O + CO_2 \leftrightarrow 2KHCO_3 + 0.5H_2O + heat.$ Moisture however, plays a significant role in the chemical adsorption process, which readily facilitates the adsorption process. Moisture usually contained as high as 8-17% in flue gases, badly affects the capacity of conventional adsorbents such as zeolites, but the present technology has no concern with moisture; water is rather necessary in principle as shown in the equation above. Carbon dioxide uptake occurred at a temperature of 60°C and the entrapped carbon dioxide was released by the decomposition of potassium bicarbonate to shift the reaction in the reverse direction. The decomposition occurred at high enough temperatures of 150°C to ensure complete regeneration of the sorbent. For the purpose of this research, emphasis was placed more on the adsorption process. When compared to other processes such as the conventional amine process, it provided an efficient, low utility cost and energyconservative effect. The activated carbon was prepared by 20% by weight of K_2CO_3 and samples used during the experimental runs were dried at 60°C for the 26-hour runs and at 25°C and 125°C for the air-dried and oven-dried samples respectively for the 48-hour runs. The samples all got to the saturation point after 6 hours of exposure to carbon dioxide and gave adsorption capacities in the range of 2.5 to 3.5mol CO_2 /mol K₂CO₃ for all experimental runs performed in this research.

Chapter 1 - Introduction

1.1. Global Warming and Green House Gases

Global warming, caused by rising greenhouse gases (GHG) in the troposphere (or enhanced greenhouse effect), has received increasing attention in recent years. The greenhouse effect is a natural process in which solar energy is transmitted through the atmosphere warming the earth. The infra-red radiation reflected from the earth surface is trapped by greenhouse gases in our atmosphere thereby causing a warming effect on the earth and making it able to maintain life. Global warming occurs when these GHG increase beyond natural levels resulting in increasing temperatures.

The Intergovernmental Panel on Climate Change (IPCC) Fourth Assessment Report on climate change 2007 stated that world temperatures could rise by between 1.1 and 6.4° C (2.0 and 11.5° F) during the 21^{st} century and that sea levels will probably rise by 18 to 59 cm. Other probable consequences of global warming include droughts, expanding deserts, heat waves, ecosystem disruption, increasingly severe weather, and loss of agricultural productivity. Greenhouse gases include carbon dioxide (CO₂), water vapor (H₂O) [6, 7], Ozone (O₃) [6], methane (CH₄), nitrous oxide (N₂O), and chlorofluorocarbons (CFCs) [6]. Since the beginning of the industrial period, the concentrations of the anthropogenic greenhouse gases have increased [6]. Of those, CO₂ is the principal greenhouse gas of interest because of "its large current greenhouse forcing, its substantial projected future forcing, and its long persistence in the atmosphere". The concentration of CO₂ in the atmosphere has increased from 280ppm in the preindustrial era to about 378ppm. With the projected increase in consumption and demand for fossil fuels, CO₂ emissions will correspondingly increase in the absence of any capture/sequestration strategy. In view that CO_2 is a greenhouse gas with the potential to contribute to global warming, existing and improved technologies to mitigate the release of CO_2 to the environment are being considered as a prudent precaution against global warming.

Prior to the industrial revolution, the relatively constant concentration of CO_2 in the atmosphere implied that the amounts of CO_2 generated by natural processes are almost equal to the amount absorbed by natural processes. However, human activity, mainly burning fossil fuels, produces about 24 billion tons of CO_2 per year and only half of that is being absorbed by natural processes [8]. The projected growth of total global annual CO_2 emissions by 2030 is forecasted to be 16 billion metric tons, resulting in total global CO_2 emissions of 43 billion metric tons. The United States' share of this growth is expected to be 12.7 percent, with the portion allocated to U.S. coal-fired power generation being 6.4 percent [10].

1.1.1. Effects to Reduce GHG Emissions

Research on reducing GHG emissions, including developing non-carbon energy sources, improving energy efficiency, and CO_2 capture and sequestration, has increased in recent years. In 1991, the International Energy Agency (IEA), funded by a consortium of governmental and Industrial organizations, established a Greenhouse Gas Research and Development Program [7]. In 2001, the Department of Energy (DOE) committed almost \$25 million to co-fund eight new exploratory projects to study methods to capture and store CO_2 . The US Electric Power Research Institute (EPRI), funded by power and fossil fuel companies from all corners of the world, was founded in 1973 and they now support research on CO_2 capture and disposal [7].

Improving energy efficiency and using non-carbon energy sources are the most effective in the short term (next 20 years) to reduce GHG emissions. Four important areas are involved: improvement of thermo-electric energy conversion efficiency of power generation plants, using technology such as natural gas combined cycle systems (NGCC); better fuel efficiency in transportation, particularly automobiles, such as the introduction of hybrid cars, fuel cell vehicles (FCV), and electric vehicles; more efficient heating and hot water supplies in buildings and houses; and development of small scale power sources like fuel cells [9].

1.1.2. CO₂ Capture and Sequestration

Since fossil fuels account for 90 percent of global energy consumption currently and cannot be phased out rapidly, we must rely on fossil fuels as the main energy source for the next several decades [3]. Thus, carbon capture and sequestration to reduce the release of CO_2 to atmosphere from stationary sources like fossil fuel fired power plants is of critical importance. The main anthropogenic source of CO_2 emissions is the burning of fossil fuels. There are a number of different anthropogenic sources of CO_2 emissions, predominantly from the combustion of fossil fuels in power generation, industrial facilities, buildings and transportation.

The idea behind carbon sequestration is to find large reservoirs for storing CO_2 rather than allowing it to discharge to the atmosphere. After separating and compressing CO_2 from combustion stack gases, liquid CO_2 can be transported and discharged into the bottom of the ocean, stored in geological formations, stored in the form of dry ice, or fixed by *in situ* lakes of algae [7], or converted to benign solid materials or fuels through biological or chemical processes [1].

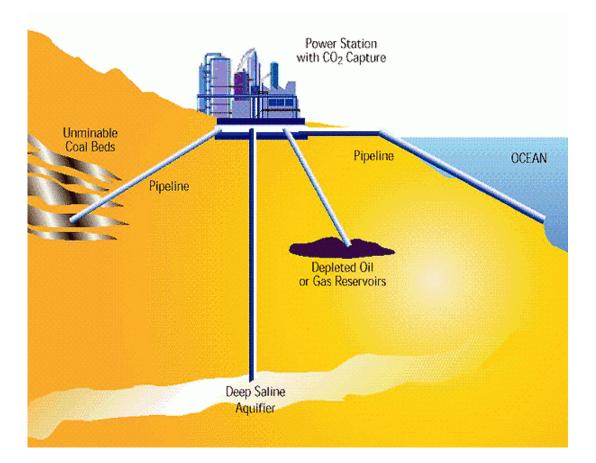
Carbon sequestration in geologic formations, one of the options for carbon management, entails adapting natural processes that have been storing CO_2 and Methane (CH₄) (another greenhouse gas) for geologic times. There are numerous natural sinks for CO₂-depleted oil and gas fields, deep unminable coal seams, rich gas bearing shales, and deep saline formations [1]. In addition, the captured CO₂ can be an effective solvent to improve oil recovery in Enhanced Oil Recovery (EOR) and used to increase the production of Enhanced Coal Bed Methane (ECBM) [5].

The various CO_2 capture options include adsorption of the gas using molecular sieves, physical and chemical absorption by solvents, low temperature (cryogenic) separation processes, and use of membranes [2]. The first step, separation and compression (i.e. capture), is currently considerably more costly than transportation and sequestration. Thus, developing new technology to reduce capture costs is the principal research topic at present.

1.2. Subsurface Storage of CO₂

The most suitable geologic settings for CO_2 storage are depleted oil and gas fields and deep saline formations [4]. These are layers of porous rock (such as sandstone) over 1km underground (either on land or far below the sea floor), located underneath a layer of impermeable rock (known as cap-rock), which acts as a seal. In the case of oil and gas fields, it was this cap-rock that trapped the oil and gas underground for millions of years.

The safety of geological storage of CO_2 will rely on the application of appropriate operational practices, regulations, monitoring and materials. The economics of geological storage depend largely on the type of reservoir being used. When storage is combined



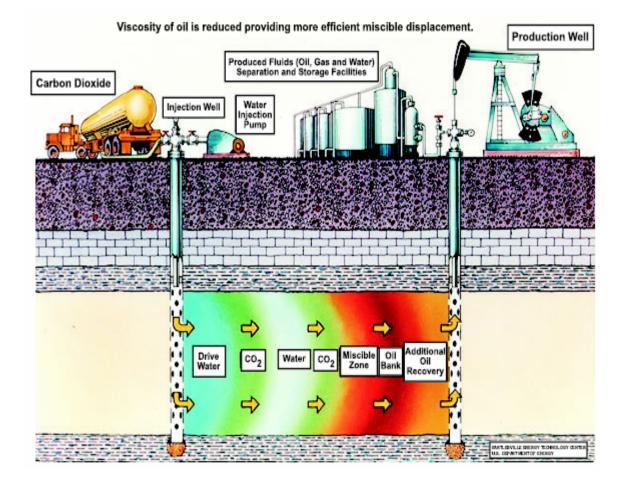
with enhanced oil recovery, as described in the next section, value is added to the process.

Figure 1. Options for the Geological Storage of CO₂[3]

1.2.1. History

The practice of pumping CO_2 into geological formations has been practiced for more than three decades now, mostly in oil and gas reservoirs, albeit not with the primary goal of storing CO_2 but rather using it as an injectant to pump oil and gas out of reservoirs. This process is known as Enhanced Oil Recovery (EOR). To this end, the focus of the research was on the technical aspects of injecting CO_2 and recovering the resource rather than on the capture of the CO_2 and the effect it would have in reducing GHG emissions. Moreover, the stability of the injected CO_2 was not at stake once the resource had been extracted and hence monitoring was not a primary concern. However, these activities established a starting point for geologic sequestration activities, especially in oil and gas reservoirs. Brief descriptions of a few selected projects in three broad categories of geologic sequestration follow to provide a quick overview of past and current research activities in the area.

1.2.2. Storage in Oil and Gas Reservoirs



1.2.2.1. Enhanced Oil Recovery

Figure 2. Schematic of CO₂ Enhanced Oil Recovery (EOR) [11]

EOR refers to techniques that allow increased recovery of oil in depleted or high viscosity oil fields. This has the potential to not only increase the yield of depleted or high viscosity oil fields, but also to sequester CO_2 that would normally be released to the atmosphere. In general terms, carbon dioxide is flooded into an oil field through a number of injection wells drilled amidst producing wells within individual petroleum reservoirs (Figure 2). Injected at a pressure equal to or above the minimum miscibility pressure (MMP), the CO_2 and oil mix and form a liquid that easily flows to the production well. Pumping can also be enhanced by flooding CO_2 at a pressure below the MMP, swelling the oil and reducing its viscosity.

Current (2005) oil production from CO_2 EOR is approximately 237,000 Bbls/day [11]. Use of CO_2 EOR in additional basins and reservoirs could increase domestic oil supply and provide effective storage of CO_2 produced from unconventional fuels production.

1.2.2.2. Weyburn CO₂ EOR Project

In late 2000, EnCana injected CO_2 into the Weyburn Field of the Williston Basin in order to boost oil production. The Weyburn oilfield covers over 70 square miles in southeastern Saskatchewan and is one of the largest medium-sour crude oil reservoirs in Canada, containing approximately 1.4 billion barrels of original oil in place [28]. Overall, it is anticipated that some 20 Mt of CO_2 will be permanently sequestered over the lifespan of the project and contribute to the production of at least 122 million barrels of 21 incremental oil from a field that has already produced 335 million barrels since its discovery in 1955.The oil field began operation in 1954 and currently there are about 650 production and water injection wells in operation. The Weyburn field produces about 10% of EnCana's total oil production. Average daily crude oil production is 2900m³/d (18,200barrels/day). Over its lifetime the field has produced some 55 million m³ of oil from primary and water flood production. The field is in production decline, having produced more than 25% of the estimated. The IEA Weyburn CO₂ Monitoring and Storage Project is coordinated by 20 research organizations in the U.S., UK, France, Italy and Denmark including the US Department of Energy (DOE)/National Energy Technology Laboratory (NETL) Carbon Sequestration Program, and co-administered by the Petroleum Technology Research Centre, Natural Resources Canada, Saskatchewan Industry and Resources, the Saskatchewan Research Council, the University of Regina and IEA GHG Research and Development Program.

1.2.3. Sequestration in Deep Saline Formations

Deep Saline formations consist of porous rock saturated with brine. They exist in most regions of the world and have the potential to store CO_2 by three main mechanisms:

- Hydrodynamic trapping of a CO₂ plume (primary mechanism)
- Solubility trapping through dissolution in the formation water.
- Mineral trapping through geochemical reactions with the formation fluids and rocks.

Currently the geology of saline formations is less well understood than for oil and gas fields. Several large saline formations underlie the United States, but there is no injection of CO_2 into them yet.

1.2.4. Sequestration in Deep, Unmineable Coal Seams

One of the few value-adding approaches to sequestering carbon dioxide is to inject it into deep, unmineable coal seams. The advantages of coal seam sequestration are that coal seams can store several times more CO_2 than the equivalent volume of a conventional gas reservoir because coal has a surface area or large volume of voids in the

form of fractures. In addition, methane is displaced and can be recovered and sold to help offset costs. This process is known as Enhanced Coalbed Methane Recovery, or ECBMR. Two existing ECBMR pilots are located in the San Juan Basin in northwest New Mexico and southwestern Colorado. The knowledge gained from studying these projects is being used to verify and validate gas storage mechanisms in coal reservoirs, and to develop a screening model to assess CO₂ sequestration potential in other promising coal basins of the U.S.

Chapter 2 - Literature Review

2.1 CO₂ Capture Technologies

A wide range of technologies currently exist for separation and capture of CO_2 from gas streams (see Figure 3). Current commercial processes employ a variety of physical and chemical mechanisms including absorption, adsorption, membranes and cryogenics [12-16]. The choice of a suitable technology depends upon the characteristics of the CO₂-laden gas stream, which in turn depends mainly on the type of power plant technology. Figure 4 shows the different types of fossil fuel power plants and technologies that affect the choice of a CO₂ capture system. Future coal-based plants may be designed to separate and capture CO₂ prior to combustion (using coal gasification systems), or they might employ pure oxygen combustion instead of air so as to obtain a concentrated CO₂ stream for treatment. Plants fueled by natural gas similarly have options to capture CO₂ either before (via gas reforming) or after combustion [17].

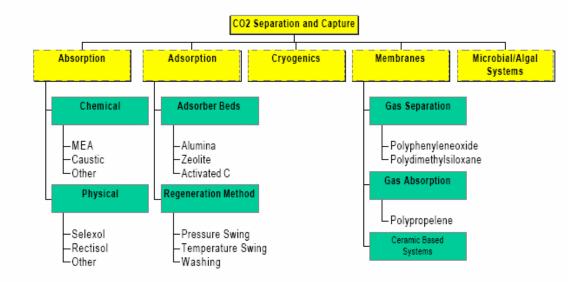


Figure 3. Technology Options for CO₂ Separation and Capture (Courtesy of [17])

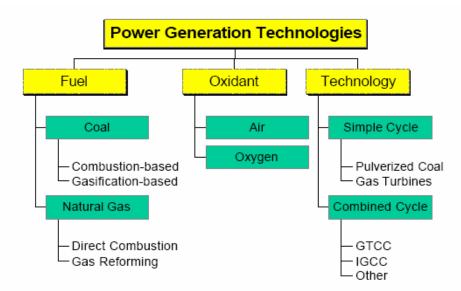


Figure 4. Technology Options for Fossil-Fuel Based Power Plants (Courtesy of [17]).

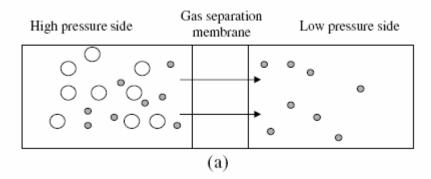
2.1.1. CO₂ Capture with Membranes

Membrane gas absorption makes use of porous, water- repelling membranes for the transfer of components between a gas and a liquid. The membrane forms a gas-permeable barrier between a liquid and a gas. Components diffuse through the pores and are absorbed by a suitable liquid. In the membrane absorber CO_2 is chemically bound in an aqueous solution. It is removed from the rich solution using thermal regeneration, using strip gas or vacuum. The lean solution is then fed back to the membrane absorber where it is reused [18]. Some of the benefits of the Membrane Gas Absorption are

- Separation process not influenced by gas-liquid ratios.
- No entrainment, flooding, foaming or channeling.
- High specific surface area through use of small size hollow fibre membranes, hence compact equipment.
- Use of modular equipment.

- Operation independent of gravity.
- Small visual impact of membrane absorbers.

The fundamental difference between membrane gas absorption and conventional membranes for gas separation is illustrated in Figure 5. In membrane gas absorption the advantages of absorption technology and membrane technology are combined. The membrane gas absorber acts as a different way of contacting the gas and the liquid phase and gives a number of advantages compared to conventional absorption towers, which may be considered dispersed phase contactors. Membrane contactors have a number of possible applications in both gas absorption and liquid/liquid extraction [18]. In some situations where gas side resistance is dominating, it may be desirable to operate the membrane in wetted mode i.e. with liquid filled pores. When operated as a liquid-liquid contactor the pores should be wetted by phase with the lowest resistance to mass transfer. A dense polymer or gel layer may be added on either side of the porous membrane in order to invoke selectivity in the membrane.



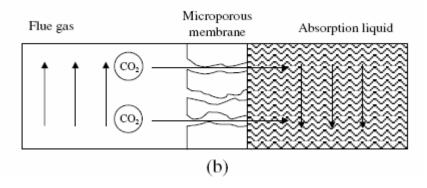


Figure 5. Principle of gas separation membrane (a) and membrane gas absorption (b) [18].

2.1.2 CO₂ capture with Cryogenic processes

Cryogenic processes are low temperature processes, which separate CO_2 directly or through a solvent. CO_2 can be physically separated from other gases by condensing it at low or cryogenic temperatures. Cryogenic processes can produce liquid CO_2 ready for transportation prior to use or sequestration, but is only worth considering when the CO_2 concentration in flue gas is high [19] (>90% CO_2) [20].

2.1.3 Chemical absorption

Currently, all commercial power plants that capture CO_2 use the processes based on the chemical absorption with a solvent. In these processes, a solvent, such as monoethanolamine (MEA), is used in a scrubbing system to remove CO_2 from the flue gas stream [21].

2.1.4 Adsorption

Adsorption methods involve a physical attraction between the gas and the active sites on a solid. This process contrasts with absorption, which causes a chemical reaction to capture CO_2 . These processes are used commercially in process industries and may be applicable to power plants in the future [21].

2.2 Amine solvents

2.2.1 Existing MEA Process

Alkanolamines, simple combinations of alcohols and ammonia, are the most commonly used category of amine chemical solvents for CO_2 capture. Reaction rates with specific gases differ among the various amines. In addition, amines vary in their equilibrium absorption characteristics and have different sensitivities with respect to solvent stability and corrosion. Alkanolamines can be divided into three groups [23]:

- Primary amines, including monoethanol amine (MEA) and diglycolamine (DGA).
- Secondary amines, including diethanol amine (DEA) and diisopropyl amine (DIPA).
- Tertiary amines, including triethanol amine (TEA) and methyldiethanol amine (MDEA).

MEA, relatively inexpensive and the lowest molecular weight, is the amine that has been used extensively for the purpose of removing CO_2 from natural gas streams. MEA has a high enthalpy of solution with CO_2 , which tends to drive the dissolution process at high rates. However, this also means that a significant amount of energy must be used for regeneration. In addition, a high vapor pressure and irreversible reactions with minor impurities such as COS and CS_2 result in solvent loss [24].

The currently preferred chemical solvent technology for carbon capture is aminebased chemical absorbent. CO_2 in the gas phase dissolves into a solution of water and amine compounds. The amines react with CO_2 in solution to form protonated amine (AH⁺), bicarbonate (HCO₃⁻), and carbamate (ACO₂⁻) [22]. As these reactions occur, more CO_2 is driven from the gas phase into the solution due to the lower chemical potential of the liquid phase compounds at this temperature. When the solution has reached the intended CO_2 loading, it is removed from contact with the gas stream and heated to reverse the chemical reaction and release high-purity CO_2 . The CO_2 -lean amine solvent is then recycled to contact additional gas. The flue gas must first be cooled and treated to remove reactive impurities such as sulfur, nitrogen oxides, and particulate matter. Otherwise, these impurities may react preferentially with the amines, reducing the capacity for CO_2 , or irreversibly poisoning the solvent. The resulting pure CO_2 stream is recovered at pressures near atmospheric pressure. Compression and the associated energy costs would be required for geologic storage.

Chapter 3 – Materials and Methods

3.1 Purpose of this Research

The high cost of separating CO₂ from flue gases is a major barrier to wider use of CO₂ removal technology. Therefore, a relatively inexpensive process for CO₂ capture from flue gas is needed to make reduction of CO₂ emissions an economically viable global goal. The objective of this research is to study a simple, inexpensive CO₂ separation process using a dry, regenerable potassium-based sorbent that may be applicable to existing fossil fuel combustion sources. The role of moisture in the adsorption process was also investigated considering the fact that moisture usually contained as high as 8-17% in flue gas, badly affects the capacity of conventional adsorbents such as zeolites, but the present technology has no concern with moisture; water is rather necessary in principle as shown in the equation 1 below. The energy loss was also calculated from the Integrated Environmental Control Model (IECM) based on a 500MW plant.

$$K_2CO_3*1.5H_2O + CO_2 \leftrightarrow 2KHCO_3 + .5H_2O + heat$$
 (Eq. 1)

3.2 Initial Screening of Sorbents

Several sorbents were reviewed and evaluated in the course of the research. The measure of evaluating these sorbents was based on certain criteria:

- Regeneration energy
- Temperature extremes involved in the adsorption and desorption processes.
- The CO₂ removal efficiency based on the kg of CO₂/kg of adsorbent.
- Parasitic energy compared to the liquid amine process.

- The relative ease, cost and availability of the sorbent.
- The binding energy.

The Integrated Environmental Control Model (IECM) (prepared for the U.S. DOE/NETL by the Carnegie Mellon University, PA, was used to calculate the performance of each of the adsorbent listed below in the table. Some data from the literature reviewed was used in order to determine the calculated values in Table 1. The model consists of a base plant and various control technology modules; these modules may be implemented together in any desired combination. The IECM model was used to calculate the energy loss with respect to a 500MW power plant. The regeneration energy required to desorb the CO_2 and make available the sorbent for re-use is calculated and used in the IECM model to obtain the energy loss in using the sorbent based on a 500MW plant.

The regeneration energy was calculated using the following formula:

$$E = mc_p \Delta T \tag{Eq. 2}$$

Where *E* = Regeneration Energy

m= Mass c_p = Heat capacity ΔT = Temperature change

Table 1.

Evaluation of Carbon Capture Technologies for the Sequestration of Carbon Dioxide from Coal-Fueled Power Plant Flue Gas

Type of Adsorbent	CO ₂ Removal	Temperatu	ire Extremes	Regeneration Energy	Energy Comparison	Energy Loss in 500 MW Plant
	kg CO ₂ /kg adsorbent	Min, ^o C	Max, ^o C	kJ/kg CO ₂	% of Liquid Amine process	MW
Diamine-Grafted						
SBA-15	0.044	25	120	1636	16.36	26
Lithium Silicate						
Pellets	0.180	500	800	1263	12.63	20
Lithium Zirconate	0.160	450	700	1184	11.84	19
Immobilized and						
aminated SBA-15	0.180	25	60	147	1.47	2
Dry sodium-based						
Pellets-AC	0.300	60	120	152	1.52	2
K ₂ CO ₃ -on-Carbon	0.075	100	150	507	5.07	8
Calcium Oxide						
based Sorbents	0.130	550	800	1457	14.57	23
Amine-Grafted						
SBA-15	0.032	25	150	2960	29.60	46
Fly Ash						
Impregnated with						
Organic Bases	0.040	25	100	1421	14.21	22

3.3 Experimental System, Procedure and Materials.

A gas cylinder containing air and 15%CO₂ was connected to a vacuum oven model 282A (46 x 31 x 31cm) with the aid of rubber tubing. The CO₂ flowed from the gas cylinder through the rubber tubing into the vacuum oven. A vacuum pump was used to evacuate the oven properly before each experimental run was performed. This was done effectively by closing the oven door, and then closing the valve leading to the CO₂/Air tank and then connecting the vacuum pump until the pressure dropped to 12.5inHg. Immediately after, the CO₂/Air valve connecting the gas cylinder to the oven is opened slowly until the pressure gets back to 28.5 inHg. The above procedure is repeated before the oven is switched on and heated to a temperature of 60°C. The gas flow rate used in the research was set at 1L/m and the pressure of the gas flowing from the cylinder was 15psig.

The potassium-based sorbents used in this study were prepared by the impregnation of K_2CO_3 on powder porous activated carbon (AC) support. Eight grams (8g) AC was prepared to form the powder with the aid of a mortar and pestle and added to two grams (2g) of anhydrous K_2CO_3 in 7ml of de-ionized water. The water was sufficient enough to form the hydrated complex of potassium carbonate. In the experimental set up, there was 20% (.2 impregnation) loading of potassium carbonate unto the activated carbon powder. It was then mixed thoroughly with the aid of a stirrer. After stirring, some samples of the potassium-based sorbents were air-dried (25°C) (and this was used in the 48-hour run), dried in an oven at 60°C (26-hour run samples), some were dried in an oven at 150°C (oven-dried samples for 8 and 48-hour runs) for different experimental runs. The CO₂ adsorption process was done in a CO₂ oven model 282A

under a pressure of 28.5inHg and the temperature in the oven was set at 60°C. The samples were removed every hour for the first 8 hours from the oven and the weight change recorded. After each weight measurement, the oven is again evacuated with the aid of the vacuum pump to ensure that the concentration of the gas in the oven is same as that flowing from the gas cylinder. The weight gained as a result of CO₂ uptake was attributed to the stable formation of the potassium bicarbonate. Weight decrease was noticed for the control samples containing just AC and moisture. This could be attributed to the loss of moisture as a result of the dry CO₂, drying it off. The weight measurements were measured to the fourth decimal figure for all samples. Drying the samples at 60°C, made sure any moisture not in the hydrated form was lost and only the hydrated complex was involved in the actual adsorption process. Samples prepared at 150°C (oven-dried samples at 150°C) lost most of its moisture but picked up significant amounts of moisture prior to the adsorption process with dry CO₂. Samples prepared at 25°C (air-dried samples), did not lose much of its moisture content since it was placed at room temperature so could not pick up significant amounts of moisture since they were almost saturated with moisture. Upon switching to CO_2 , the samples prepared at this temperature showed weight gain as a result of the stable formation of the potassium bicarbonate.

Chapter 4 - Results and Discussion

4.1 Results and Discussion

Table 2 below shows the weight change as a result moisture and CO_2 uptake by the samples. This initial run for 8 hours was simply to show the role of impregnation and moisture in the adsorption process. Water was placed in a dish alongside the samples during the first 5 hours of the run and was immediately removed, the oven evacuated with the aid of a vacuum pump before the adsorption with dry CO₂. The first 5 hours of this run was with just in the presence of moisture until the samples reached a saturation point and the next 3 hours (i.e. from the 6th hour) were in the presence of dry CO₂. The role of moisture was to enable the activated carbon and potassium carbonate mixture to be hydrated enough to pick up carbon dioxide. Samples containing pure K₂CO₃ and activated carbon showed a slight increase in weight as a result of moisture uptake. The oven and air-dried samples were prepared by mixing approximately 8g of AC and 2g of K₂CO₃. However, the oven –dried sample had slightly more K₂CO₃ than the air-dried sample because the measurements were done manually and lacked the accuracy of measuring equal amounts of the AC and K_2CO_3 on all samples. The oven-dried sample picked up more moisture than the air-dried sample since it had lost most of its moisture when dried in an oven at 150°C compared to the air-dried sample which was dried at 25°C. In the presence of dry CO₂, the oven and air-dried samples showed weight gains whereas the pure K₂CO₃ and activated carbon samples did not show any significant weight gain.

Time(Hr)	Anhydrous K ₂ CO ₃ (g)	Act. Carbon (g)	Oven-dried(g) (150°C)	Air-dried(g) (25°C)
0	5.0116	6.4823	12.0051	12.6124
1	5.0243	6.4889	12.1617	12.6243
2	5.0254	6.4966	12.5861	12.7505
3	5.0339	6.4969	12.7643	12.8565
4	5.0402	6.4983	12.8433	12.8910
5	5.0499	6.4521	12.8630	12.949
6	5.0606	6.4581	13.0046	13.0821
7	5.0617	6.4619	13.1750	13.1665
8	5.0656	6.4619	13.4370	13.3674

Table 2. Weight Changes for Activated Carbon (AC) Impregnated with Potassium Carbonate (K_2CO_3) after 8-Hour Run with Moisture, CO₂ at 60°C.

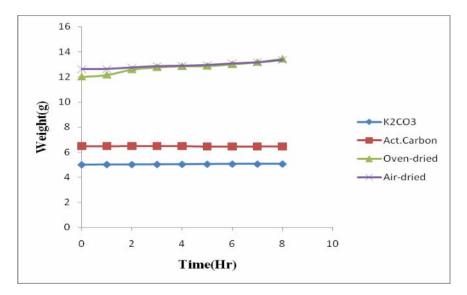


Figure 6. Weight Changes for Activated Carbon (AC) Impregnated with Potassium Carbonate (K₂CO₃) after 8-Hour run with moisture, CO₂ at 60°C.

4.1.1. 26- Hour Run 1 with CO₂

Table 2 shows the weight change for potassium powder-based sorbent and the pure Activated carbon powder after a 26- hour run. The values show that there was a gradual increase in weight until it started leveling off from the 24th to the 26th hour though the saturation level was reached at the 6th hour. Samples 1 and 2 made up of mixtures of activated carbon, K₂CO₃ and moisture were thoroughly mixed and dried in an oven at 60°C for 12 hours. The moisture was just sufficient enough to mix the activated carbon and K_2CO_3 . Samples 1 and 2 both made up of the potassium -powder based sorbent showed similar trends in weight gain while the control made up of just pure Activated carbon powder and moisture showed a decrease in weight as a result of moisture loss. Three experimental runs were performed with the weight change trend consistent throughout the separate 26 hour experimental runs performed. The run was performed at 60°C for 26 hours in the presence of dry CO₂ flowing from the gas cylinder at 1L/m, weight measurements were taken every hour for the first 8 hours of the run and allowed to run till the 24th hour where the weights were taken after each hour till the 26th hour. It should be noted samples 1 and 2 are replicates.

Tables 3 and 4 weight change values also show a similar trend as seen in the previous 26-hour run discussed above. The level of CO_2 saturation was attained the 6th as seen in the figure 7. This shows that after this time the reverse reaction could be possible by increasing the desorption temperature to 120°C to release CO_2 . The same trend was seen in Figures 8 and 9 below. Sample 3 and 4 are replicate samples.

Time	AC+K ₂ CO ₃ ,S1	AC+K ₂ CO ₃ ,S2	AC Powder
0	11.005	10.749	9.0102
1	11.221	10.934	8.8998
2	11.458	11.173	8.8506
3	11.771	11.446	8.7008
4	12.169	11.706	8.6041
5	12.535	11.973	8.5916
6	12.848	12.281	8.5044
7	12.860	12.389	8.4533
8	12.868	12.416	8.4347
24	12.894	12.474	8.4176
25	12.908	12.537	8.4170
26	13.016	12.567	8.4098

Table 3. Weight Changes for Activated Carbon (AC) Impregnated with Potassium Carbonate (K₂CO₃) after 26-Hour Run 1 with CO₂ at 60°C.

Where (AC+K₂CO₃), S1= Activated carbon powder impregnated with potassium carbonate, (sample 1) for 26-hour run prepared at 60°C for 12 hours.

Where (AC+K₂CO₃), S2= Activated carbon powder impregnated with potassium carbonate, (sample 2) for 26-hour run prepared at 60° C for 12 hours.

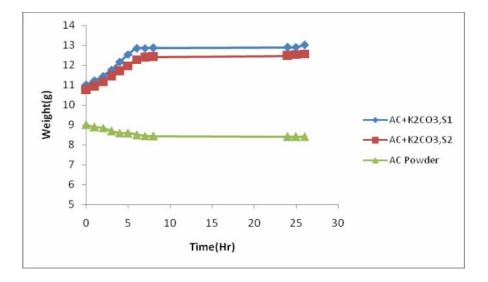


Figure 7. Weight Changes for Activated Carbon (AC) Impregnated with Potassium Carbonate (K₂CO₃) after 26-Hour Run for Run 1 at 60°C.

Time (Hr)	AC+K ₂ CO ₃ ,S3	AC+K ₂ CO ₃ ,S4	AC Powder
0	11.0971	11.366	8.2536
1	11.2470	11.4989	8.0559
2	11.5308	11.6287	7.9409
3	11.7735	11.8875	7.8573
4	12.0330	12.1874	7.6439
5	12.2935	12.5281	7.5396
6	12.4648	12.6005	7.5121
7	12.5466	12.7140	7.4814
8	12.6955	12.8127	7.4398
24	13.0058	13.1845	7.4109
25	13.0536	13.2674	7.3996
26	13.0836	13.2784	7.3948

Table 4. Weight Changes for Activated Carbon (AC) Impregnated with Potassium Carbonate (K₂CO₃) after 26-Hour Run 2 with CO₂ at 60°C.

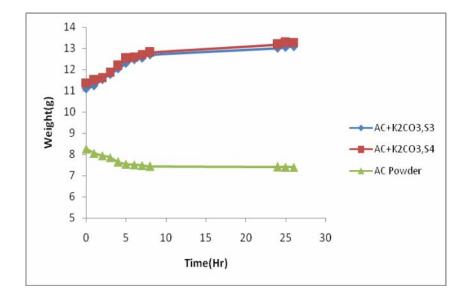


Figure 8. Weight Changes for Activated Carbon (AC) Impregnated with Potassium Carbonate (K_2CO_3) after 26-Hour Run for Run 2 at 60°C. (Samples 3 and 4 are replicates).

Time	AC+K ₂ CO ₃ ,S5	AC+K ₂ CO ₃ ,S6	AC Powder
0	13.9485	14.3043	8.241
1	14.1244	14.4218	8.1147
2	14.2406	14.5547	7.9112
3	14.3971	14.6625	7.9622
4	14.5201	14.8147	7.7112
5	14.8994	14.9954	7.5412
6	15.0582	15.1147	7.4487
7	15.1247	15.2156	7.4011
8	15.2571	15.3451	7.3945
24	15.7844	15.9145	7.3666
25	15.8275	15.9512	7.3510
26	15.8357	16.1136	7.3560

Table 5. Weight Changes for Activated Carbon (AC) Impregnated with Potassium Carbonate (K₂CO₃) after 26-Hour Run 3 with CO₂ at 60°C.

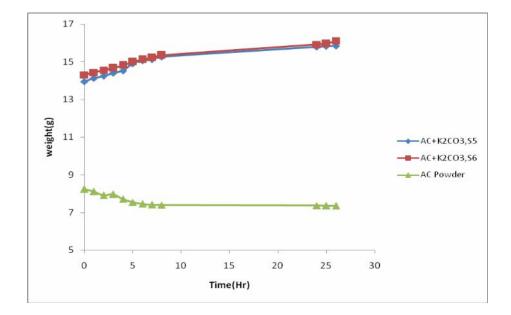


Figure 9. Weight Changes for Activated Carbon (AC) Impregnated with Potassium Carbonate (K_2CO_3) after 26-Hour Run for Run 3 at 60°C. (Samples 5 and 6 are replicates).

The table 6 below shows the calculated amount of CO_2 adsorbed in mmol, the ratio of mol CO_2 to mol K_2CO_3 (mol CO_2 /mol K_2CO_3), in both potassium powder based sorbent samples during the 26-hour run1. The figure 10 below shows the adsorption capacity for samples 1 and 2. The adsorption capacities of approximately 3.3 and 3.0 mol

 $CO_2/molK_2CO_3$ where obtained for samples 1 and 2 respectively. It should be noted noted that samples 1 and 2 are both activated carbon impreganted with potassium carbonate but have slight differences in the impregnation amounts of the K₂CO₃.

Sample 1				Sample	2
Weight gain(g)	CO ₂ ads(mmol)	molCO ₂ /K ₂ CO ₃	Weight gain(g)	CO ₂ ads(mmol)	molCO ₂ /K ₂ CO ₃
0.2155	8.2885	0.5251	0.1846	7.1000	0.4819
0.4531	16.755	1.0615	0.4234	15.651	1.0622
0.7662	23.871	1.5123	0.6964	21.855	1.4832
1.1640	32.912	2.0850	0.9559	27.753	1.8835
1.5298	41.226	2.6117	1.2230	33.823	2.2955
1.8429	48.342	3.0625	1.5315	40.835	2.7713
1.8550	48.617	3.0800	1.6394	43.287	2.9377
1.8628	48.794	3.0912	1.6659	43.889	2.9786
1.8890	49.389	3.1289	1.7245	45.221	3.0690
1.9025	49.696	3.1484	1.7869	46.639	3.1652
2.0113	52.169	3.3050	1.8175	47.335	3.2124

Table 6. Stoichiometric Calculation of CO₂ Adsorbed in mmol for Activated Carbon Impregnated with Potassium Carbonate for 26-hour run 1.

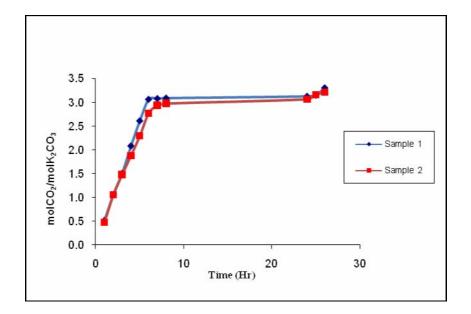


Figure 10. Adsorption Capacity for Activated Carbon Impregnated with Potassium Carbonate (for samples 1 and 2) during 26 –Hour Run for Run 1at 60°C.

Figure 11 summarizes the correlation between the amounts of entrapped carbon dioxide and potassium carbonate in the oven for various preparations of K_2CO_3 -on-activated carbon powder. It shows that the more impregnation with K_2CO_3 , the more CO_2 uptake.Table 7 gives the mmol of CO_2 uptake per mmol of K_2CO_3 used. The efficiency is the mole ratio of the amount of CO_2 uptake to the amount of K_2CO_3 impregnated onto the activated carbon powder.

Run 1	mmol K ₂ CO ₃	mmol CO ₂ ads	Efficiency(ratio of mmol CO ₂ /k ₂ CO ₃)
sample 1	15.784	52.168	3.3050
sample 2	14.734	47.334	3.2124
run 2			
sample 3	15.220	63.500	4.3096
sample 4	14.724	53.467	3.5129
run 3			
sample 5	14.594	48.853	3.3519
sample 6	14.580	47.148	3.2336

Table 7. Efficiency for Entrapping CO_2 on the Activated Carbon Powder Impregnated with K_2CO_3 during 26 –Hour Run at 60°C.

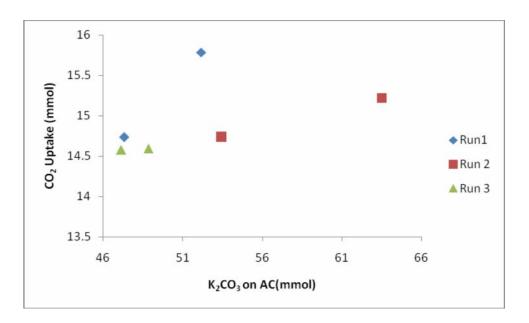


Figure 11. Efficiency for Entrapping CO₂ on the Activated Carbon Powder Impregnated with K₂CO₃ during 26-Hour Runs at 60°C.

4.1.2 48-Hour Run with Moisture, CO₂

Figure 12 shows the weight gain during a 48-hour run. The two samples used in this run were the oven and air-dried samples which were both prepared by impregnating the activated carbon powder with an aqueous solution of potassium carbonate. The oven dried sample was dried in the oven at 150° C for an hour while the air-dried sample was dried at 25° C for an hour. The first 24 hours of the 48-hour run showed an increase in weight as result of moisture uptake until it reached a saturation level at the 7th hour where there was not any moisture uptake. The samples were placed in the oven at 60°C for the first 24 hours in the presence of humid air (moisture). After 24 hours of saturation with moisture, the moisture was removed and the samples were then CO₂ was allowed to flow in the oven for another 24hours.

After hydrating the sample sufficiently enough with moisture, there was a significant CO_2 uptake until it reached saturation at the 30th hour of the run. This shows that the samples had reached saturation point after 6 hours of exposure to CO_2 . This shows that the sorbent reached its saturation level with CO_2 uptake during the 48-hour run at the 6th hour of exposure to CO_2 . Figure 13 also shows the adsorption capacity during the 48-hour run for both the oven-dried and air-dried samples which are approximately 2.7 and 2.5 mol CO_2 /molK₂CO₃ respectively. Table 8 shows weight change as result of the experimental run.

Time	Oven-dried(g) (150°C)	Air-dried(g)(25°C)
0	11.4203	11.6260
1	11.5980	11.8660
2	11.9966	12.1048
3	12.0139	12.2335
4	12.1291	12.2762
5	12.2381	12.3499
6	12.3670	12.3700
7	12.3534	12.3478
8	12.3713	12.3521
24	12.3738	12.3566
25	12.9103	12.6839
26	13.1173	12.8921
27	13.3086	13.0869
28	13.5792	13.4320
29	13.7981	13.6848
30	13.8453	13.7076
31	13.8906	13.7100
32	13.9386	13.7191
48	13.9362	13.6941

Table 8. Weight Changes for Activated Carbon (AC) Impregnated with Potassium Carbonate (K_2CO_3) after 48-Hour Run with Moisture, CO_2 at 60°C.

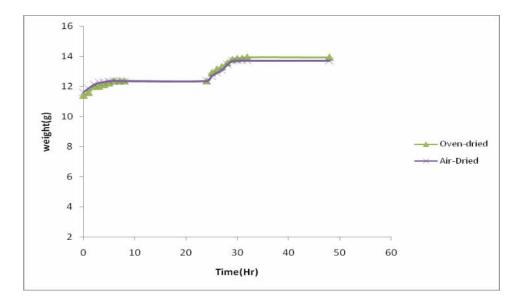


Figure 12. Weight Changes for Activated Carbon (AC) Impregnated with Potassium Carbonate (K_2CO_3) during 48-hour run at 60°C.

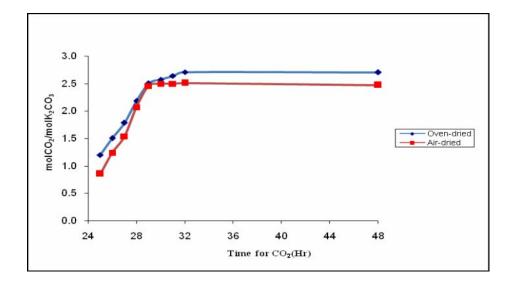


Figure 13. Adsorption Capacity for Activated Carbon Impregnated with Potassium Carbonate during 48- hour run at 60°C.

Figure 13 also summarizes the correlation between the amounts of entrapped carbon dioxide and potassium carbonate in the oven for various preparations of K_2CO_3 -on-activated carbon powder during the 48-hour run.

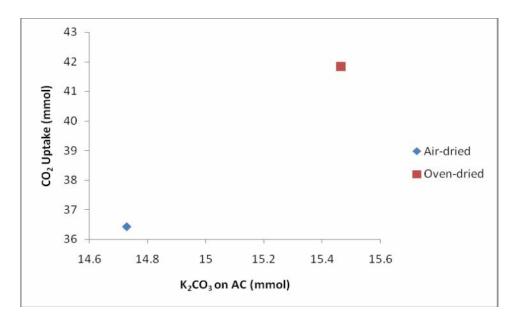


Figure 14. Efficiency for Entrapping CO_2 on the Activated Carbon Powder Impregnated with K_2CO_3 during 48-hour run at 60°C.

4.2 Practical Energy Usage

26-Hour run 1				
Sample 1			Sample 2	
$Mass(g)K_2CO_3$	2.1783		Mass(g)K ₂ CO ₃	2.0334
Mass(g) CO ₂	0.6945		Mass(g) CO ₂	0.6483
Mass of H ₂ O	0.1421		Mass of H ₂ O	0.1326
Net. Weight gain	0.5525		Net. Weight gain	0.5157
$XS CO_2(g)$	1.4588		$XS CO_2(g)$	1.3018
Mass(g) AC	8.0298		Mass(g) AC	8.0143
gH ₂ O	0.7969		gH ₂ O	0.7019
cp AC (kJ/molK)	0.0085		cp AC (kJ/molK)	0.0085
cpH ₂ O (kJ/molK)	0.0754		cpH ₂ O (kJ/molK)	0.0754
cpK ₂ CO ₃ kJ/molK)	0.1251		cpK ₂ CO ₃ kJ/molK)	0.1251
Hrxn (kJ/mol)	37.9000		Hrxn (kJ/mol)	37.9000
HvapCO ₂ (kJ/mol)	25.2295		HvapCO ₂ (kJ/mol)	25.2295
HvapH ₂ O (kJ/mol)	40.7061		HvapH ₂ O (kJ/mol)	40.7061
dT	60.0000		dT	60.0000
dHrxn kJ	0.5982	E1	dHrxn kJ	0.5584
dH xs CO ₂ kJ	0.8365	E2	dH xs CO ₂ kJ	0.7464
dH free H ₂ O kJ	1.4809	<i>E4</i>	dH free H ₂ O kJ	1.2874
dH AC kJ	0.3419	E3	dH AC kJ 0.3413	
dH K ₂ CO ₃	0.1185	E3	dH K ₂ CO ₃	0.1106
Total Energy	3.3760		Total Energy	3.0442
Total CO ₂ g	2.1534		Total CO ₂ g	1.9501
Energy per gCO ₂ kJ/gCO ₂	1.5678		Energy per gCO ₂ kJ/gCO ₂	1.5610
Energy per kgCO ₂ kJ/kgCO ₂	1567.8028		Energy per kgCO ₂ kJ/kgCO ₂	1561.0340
Power per MMlbCO ₂ (MW)	177.8361		Power per MMlbCO ₂ (MW)	177.0683
Compression Power (MW)	60.0000		Compression Power (MW)	60.0000
Total Power Req.	237.8361		Total Power Req.	237.0683
% facility output	47.57%		% facility output	47.41%

Table 9. Practical Energy	Usage in a 500MW	⁷ Plant from tl	his Research for 26-Hour
run 1 with Dry CO ₂ .			

Where cp= Heat Capacity

AC= Activated Carbon

Hrxn= Enthalpy of Reaction

mCO₂=Mass of CO₂

Hvap= Heat of Vaporization.

XS= Excess

Considering the fact that the research is based on a 500MW power plant, 47.57% and 47.41% from the facility output for samples 1 and 2 for 26-hour run 1 respectively, can be directed towards carbon dioxide compression. The percentage facility output was calculated by dividing the total power requirement by 500 and expressed as a percentage.

The total energy per gram of CO₂ calculated

The formulas used in the calculations above include:

The mass of CO₂ adsorbed will be:

$$M_{CO_2} = G \left(\frac{MW_{CO_2}}{MW_{K_2 CO_3}} \right)$$
(Eq.3)

Where m_{CO2} =mass of CO₂ adsorbed

G = mass of K₂CO₃ used

 MW_{CO2} = Molecular weight of CO₂

 MW_{CO2} = Molecular weight of K₂CO₃

The amount of water released will be:

$$M_{H_2O} = 0.5 \left(\frac{MW_{H_2O}}{MW_{K_2CO_3}} \right)$$
(Eq.4)

 M_{H2O} = amount of water released.

 MW_{H20} = Molecular weight of water

 MW_{K2CO3} = Molecular weight of K₂CO₃

The net weight gain will be:

Net Weight gain = M_{CO2} - M_{H2O}

(Eq.5)

Any weight gain above the net weight gain will be adsorption of carbon dioxide to the surface without chemical reaction, which we can call excess CO₂.

*Excess CO*₂= *Actual weight gain - Net weight gain* (Eq.6)

To calculate the energy required to desorb the carbon dioxide, several steps are required. The first is to desorb the carbon dioxide that has reacted with the K₂CO₃. The heat of reaction (ΔH_{rxn}) is calculated based on the chemical reaction above using the heats of formation of each component on a mole basis (kJ/mole). The amount of energy for the reaction is calculated by:

$$E_1 = \left(\frac{M_{CO_2}}{MW_{CO_2}} \Delta H_{rxn}\right)$$
(Eq.7)

The energy required to desorb the excess CO2 is calculated by:

$$E_2 = \left(\frac{ExcessCO_2}{MW_{CO_2}} \Delta H_{vap,CO2}\right)$$
(Eq.8)

Where $\Delta H_{vap,CO_2}$ is the heat of vaporization of liquid carbon dioxide. The amount of energy required to heat up the solid is given by:

$$E_{3} = (\mathbf{m}_{carbon} \mathbf{C}_{\mathbf{p}, carbon} + \mathbf{m}_{carbon} \mathbf{C}_{\mathbf{p}, carbon} + \mathbf{m}_{water} \mathbf{C}_{\mathbf{p}, water}) \Delta T$$
(Eq.9)

Where m_{water} is the amount of water remaining after the powder was dried before the carbon dioxide adsorption experiment and ΔT is the temperature change. The energy required to vaporize the remaining water is given by:

$$E_{4} = (M_{water} - M_{H_{2}O})\Delta H_{vap, H_{2}O}$$
(Eq.10)

The total energy per gram of carbon dioxide is given by:

$$E_{Total} = \frac{E_1 + E_2 + E_3 + E_4}{M_{CO2} + ExcessCO_2}$$
(Eq.11)

CO ₂) 1567 1561
1561
1677
2812
1971
2285
1785
2250
4545
2033

Table. 10 Comparisons of Heat Requirements for CO₂ Recovery in Practical Plants.

* denotes present work

<u>Chapter 5 - Summary, Conclusions and Recommendations</u> 5.1 Summary

The relatively high adsorption capacity results in significant savings in construction, maintenance and equipment costs. The time taken to reach the saturation point for all runs with similar impregnation (in weights) was fairly constant; this was evident in the three different experimental runs performed for 26 hours where the peaks for the saturation level occurred during the 6th hour of the runs.

The role of moisture was significant in the adsorption process as shown in the equation; moisture usually contained as high as 8-17% in flue gas, badly affects the capacity of conventional adsorbents such as zeolites, but the present technology has no concern with moisture; water is rather necessary in principle as shown in the equation 1. The experimental runs had 20% impregnation unto the activated carbon powder.

For the experimental runs performed, it took 6 hours to reach saturation level with the amount of CO₂ adsorbed after which there was not any significant CO₂ pick up. On average the adsorption capacity was in the range of 2.5 to 3.3 mol of CO₂ for all runs for both the 26-hour runs and the 48-hour runs per kg of adsorbent used. The energy required per kilogram of CO₂ recovered in this present research is 1567 KJ/Kg CO₂ and 1561KJ/Kg CO₂ for samples 1 and 2 respectively during 26-hour run 1,167 KJ/Kg CO₂ for and 2812 KJ/Kg CO₂ for 26-hour run 2, 1971 KJ/Kg CO₂ and 2285 KJ/Kg CO₂ for samples 1 and 2 respectively for 26-hour run 3, 1785 KJ/Kg CO₂ and 2250 KJ/Kg CO₂ for oven-dried and air-dried samples respectively for 48-hour run, compared to 4545 KJ/Kg CO₂ for the conventional MEA process [26]. The low energy consumption translates to a low utility cost.

5.2 Conclusion

The weight gain in the experimental run predicts the formation of the stable carbonation product of KHCO₃ at 60°C. The saturation level with CO₂ reached after 6 hours of experimental run indicating that there was not any significant increase after that time. Regeneration of the sorbent and the eventual release of CO₂ could be achieved at this point. K_2CO_3 on its own could not serve as a substrate to adsorb CO₂ but rather could increase the adsorptive capacity of other materials such as the activated carbon pellets used in this experiment.

5.3 Recommendations

A lifetime test should be carried out using the best performing sorbent and reaction conditions in which the highest CO₂ removal efficiency and consistent sorbent durability are achieved. Further tests should be performed on how to achieve optimum CO_2 uptake /adsorbent used. A test to compare the adsorption capacity between a run containing CO_2 and moisture at the same time with a run of just CO_2 to determine which method gives the optimum adsorption capacity. The economic evaluation of the process needs to be researched for commercial purposes such as the capital cost for the construction.

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