CARBON DIOXIDE CAPTURE THROUGH BIOMASS GASIFICATION

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ABSTRACT: Gasification, a thermo-chemical process, is explored as a promising technology towards carbon capture. The present work focuses on using CO_2 , from a typical flue of combustion of fuels in an engine or a combustion device, as a co-reactant along with other reacting media. Experiments were conducted with CO_2 volume fraction varied from 0 to 15% in a mixture of O_2 and N_2 . Increase in CO_2 fraction resulted in decrease in bed temperature, primarily due to reduction in O_2 fraction, in the gasifying medium, and the endothermic reaction of char and CO_2 . Low bed temperature was addressed by maintaining the O_2 volume fraction in the input at 21% by introducing additional oxygen. At 15% CO_2 injection, the CO fraction increased from 13.1% to 16.3% and over 55% of the input CO_2 conversion was noted. Recorded an increase in cold gas efficiency by 30% owing to higher conversion rate of char. Working with the engine exhaust also eliminates the cost incurred in separation of CO_2 and makes the system less complicated.

Keywords: CO₂ capture, CO₂ reduction, gasification, CO₂ emissions

1 INTRODUCTION

Ever increasing energy demands have become a primary concern, and meeting this escalating energy need has resulted in renewed interest in alternatives to fossil fuels. The interest amongst all the climate change mitigation researchers is in addressing the reduction of emissions and also attempt sequester the carbon. Amongst the technologies receiving the most such attention to reduce CO₂'s impacts is CO₂ capture and sequestration. CO₂ sequestration involves removing CO₂ from the fuel, either before, during, or after combustion, and then capturing and converting it, to avoid its release to the atmosphere. While other greenhouse gases (e.g., methane) are more potent in terms of global warming effects per unit of mass, the CO₂ emissions of industrialized economies are so great as to dwarf the contributions from other gases in terms of overall impact on global warming. Hence, the focus is on CO₂ sequestration technologies.

The relative contributions of different fossil fuels to total energy-related carbon dioxide emissions have changed over time. In 1990, carbon dioxide emissions associated with liquid fuels made up an estimated 42 percent of the world total; in 2007, their share was 38 percent. The pursuit of greenhouse gas emissions reductions has the potential by reducing global coal use significantly. Limitations on carbon dioxide emissions will raise the cost of coal relative to the costs of other fuels. Under such circumstances, the degree to which energy use shifts away from coal to other fuels will largely depend on the costs of reducing carbon dioxide emissions from coal-fired plants relative to the costs of using other, low-carbon or carbon-free energy source [1].

Amongst the emerging CO_2 capture technologies, absorption of CO_2 has received considerable attention. Absorption processes use a suitable solvent to separate CO_2 from the flue gas stream. Alkanoamines (e.g., MEA -Mono-ethanol amine, and diethanolamine) are typically used in chemical absorption, whereas methanol,

dimethylether, polyethylene glycol, and sulfolane are employed in physical absorption [2]. The major issues with MEA and other solvents include equipment corrosion in the presence of O₂, and the energy-intensive solvent regeneration. The presence of common flue gas contaminants, such as SO_x , and NO_x , also has a negative impact on solvent based process performance. Comparatively, the ammonia scrubbing technique provides advantages of lower material costs, higher absorption efficiency, a greater absorption capacity and less corrosion to absorber, as well as potential to save energy [3][4]. The amount of CO₂ that can be removed from the exhaust depends on the size of the absorption unit and the concentration of CO₂ in the exhaust. For a standard plant the economical recovery limit is approximately 85% for 3% CO₂ in the exhaust, and 90-92% for 8% [5]. Adsorption processes are based on the selective adsorption of CO₂ on a solid adsorbent, such as zeolites, alumina molecular sieves and activated carbon. One very promising high temperature chemical sorbent is that of CaO, which can be used either directly or in modified form, and is freely obtained from limestone, which is abundantly available [6]. The sorption /desorption temperatures of modified CaO are normally in the range of 650-850 °C. Porous membranes, which are capable of separating gas molecules of different molecular sizes, are available in a variety of forms, including polymers, metals and rubber composites. The main disadvantage of membrane separation is its low gas throughput, and the need for multistage operation or stream recycling [7]. The development of a membrane separator for the selective removal of CO_2 in the presence of CO, H_2 , H₂O and H₂S (fuel gas) or N₂, O₂, H₂O, SO₂, NO_x, and HCl (flue gas) would be of tremendous economic value. A membrane separation technique requires less maintenance and energy than a comparable absorption system. Table I gives a glimpse of carbon capture and separation technologies with limitations and advantages [8].

Carbon capture and separation technology	Benefits	Limitations	Energy cost for separation (kW/ ton CO ₂)
Absorption (liquid) – MEA	Solvents can be regenerated easily. Well established and widely used for over past 65 yrs.	Desulfurization and removal of NO_x , SO_x and particulate matters required. O_2 content in flue gas damages the system.	309
Adsorption – CFCMS	Can play significant role in hybrid system (coupled with another separation process).	Not capable enough to handle large concentration of CO_2 (>1.5%) – not suitable for engine exhaust.	617
Adsorption – PSA	Can play significant role in hybrid system (coupled with another separation process).	Not capable enough to handle large concentration of CO_2 (>1.5%) – not suitable for engine exhaust.	154
Cryogenic distillation	Product is liquid CO_2 , ready to transport. CO_2 recovery is very high compared to other methods.	All components except N_2 and CO_2 needs to be filtered from flue gas.	726
Membrane Separation	Design and construction simpler and cheaper, as no requirement for high temperature or pressure. Flue gas can be directly used without SO_x/NO_x removal.	CO ₂ separation efficiency is low (~50%)	610

Table I: Overview of Carbon Capture and Separation (CCS) technologies [8]

This work, involves an alternative strategy for dealing with CO₂. This propitious use of CO₂ involves its recycling into the fuel making process. Biomass gasification is explored as a promising technology to convert CO₂ to a fuel gas, identified as producer gas. CO₂, from the engine exhaust, is used as a co-reactant in the gasification medium, along with air. This work concentrates on capturing CO₂ from a typical combustion system like, engine exhaust where CO₂ fraction varies from 12% (CNG) to 15% (diesel), on dry basis. Scrubbing of SO_x/NO_x or particulate matter is not required. Condensing H₂O and mixing exhaust with proportionate amount of O₂, eliminates the cost of separation and storage of CO₂ as well.

With energy consumption expected to double over the next 40 years and with heightened environmental consciousness as to the need for limiting CO_2 atmospheric emissions, the feasibility of biomass-derived fuels as one of the significant carbon neutral energy solutions has emerged.

The paper addresses use of biomass gasification process to capture CO_2 from a typical combustion system, say an internal combustion engine. Biomass constitutes one of the more promising carbon neutral energy sources that can be part of the solution. Biomass gasifiers converts solid fuel(biomass) to gaseous fuel through thermochemical process. It undergoes the process of pyrolysis, oxidation and reduction under sub-stoichiometric conditions. Oxidation of pyrolysis products (volatile matter) takes place which in turn reacts with char to produce H₂, CO, CO₂, CH₄, H₂O and small fractions of higher hydrocarbon (HHC). It is a heterogeneous reaction between the char and gaseous species of pyrolysis combustion products. These, as sources of combustible gas for energizing internal combustion engines, have been in existence for nearly a century.

Following are the majorly involved reactions in the reduction zone:

Oxidation:

 $C + O_2 \Leftrightarrow CO_2 + 393.8 \text{ kJ/mole}$ (1)

Water gas reaction:

$$C + H_2 O \Leftrightarrow H_2 + CO - 131.4 \text{ kJ/mole}$$
 (2)

Boudouard reaction:

$$C + CO_2 \Leftrightarrow 2CO - 172.6 \text{ kJ/mole}$$
 (3)

Water shift reaction:

$$CO + H_2O \Leftrightarrow CO_2 + H_2 + 41.2 \text{ kJ/mole}$$
 (4)

Methane reaction:

$$C + 2H_2 \Leftrightarrow CH_4 + 75 \text{ kJ/mole}$$
 (5)

Introducing CO_2 as a co-reactant, along with air in the open top downdraft gasifier reduces CO_2 to CO, owing to reactions 3 and 4 above. The CO_2 gasification of char has been extensively studied.

The mechanism of heterogeneous reaction of CO_2 with C, proposed by Blackwood and Ingeme has been based on the adsorption of process where CO_2 breaks down at the C surface to form CO and O in the adsorbed state. The O atom then reacts with C to form molecular CO [9]. The kinetic mechanism considered for the C + CO₂ reaction are as follows:

$$CO_2 \xrightarrow{k_1} CO + (O) \tag{6}$$

$$C + (O) \xrightarrow{\kappa_2} CO \tag{7}$$

The retarding effect of CO is explained by the competition of CO with O for the active sites; i.e.

$$CO \underset{K_4}{\overset{K_3}{\Leftrightarrow}} (CO) \tag{8}$$

$$CO + (CO) \xrightarrow{K_5} CO_2 + C \tag{9}$$

The reaction 9, involves the introduction of reverse of $C-CO_2$ reaction by neglecting the adsorption of CO. In the above set of reactions (O) and (CO) are species in the adsorbed state at the active sites which are present in different fractions at each site. Assuming (O) and (CO) to be in a quasi-steady state, the overall rate expression with the elementary reactions has been proposed as:

$$\omega_{c}^{'''} = \frac{-k_1 p_{CO_2} - K_2 p_{CO}^2}{1 + K_3 p_{CO} + K_4 p_{CO_2}}$$
(10)

$$k_1 = 2.2 \times 10^9 e^{-E/RT} \text{ mol/cm}^3$$

K₂ is obtained from equilibrium

$$K_3 = 15.0 \text{ atm}^{-1}$$

 $K_4 = 0.25 \text{ atm}^{-1}$

where $\dot{\omega}_c^{(i)}$ is the , reaction rate of carbon per unit volume, k_1 is the rate constant, and K_2 , K_3 , K_4 are obtained from detailed kinetic steps, with p_i being the partial pressure of the species. This reaction has been extensively studied by Dasappa et al [15].

Apart from enhancing CO in producer gas, introducing CO_2 as a co-reactant changes the H₂/CO ratio as well. In recent years, the availability of biomass and the successful research on it have provided momentum in the use of Fischer Tropsch (FT) technology. Production of syngas from biomass and its subsequent transformation via FT synthesis has potential to produce a wide spectrum of linear and branched chain synthetic hydrocarbons. This process is called biomass to liquid fuel (BTL) and has become an excellent option to obtain liquid transportation fuels. These fuels are ultraclean; contain no aromatics, no sulfur, and no nitrogen containing compounds. Therefore, they can easily satisfy the upcoming stricter global environmental regulations [10]. Presence of CO2 would help in establishing H₂/CO ratio in a syngas generation unit, using CO₂-O₂-steam gasification. Using CO₂, the opportunity exists to adjust the ratio of H₂ / CO in the syngas produced, by selecting the concentration of CO₂ fed into the reactor system resulting in operational and economic advantages.

Butterman and Castaldi had done studies on gas evolution on several biomass and organic compounds with steam-CO₂ gasification in a thermally controlled environment [11]. Feedstock were heated from 300-1300 K at rates of 1-100 K/min in H_2O , CO_2/N_2 and 100% CO_2

environment. Higher conversion of biomass was reported in CO_2 gasification. Results showed the variation of H_2/CO ratio from 0.25 at 50% CO_2 to 5.5 at 0% CO_2 .

The current work addresses use of biomass gasification process to capture CO_2 from a typical combustion system in self-sustained manner.

2 METHODOLOGY

The development work at the Indian Institute of Science has resulted in a design with an open top downdraft gasifier, with air entering both at the top and at the air nozzles in the lower part. Stratification of the fuel bed keeps larger part of the bed volume at higher temperature and thus helps in maintaining low tar levels. Figure 1 shows the outline of the open top downdraft gasifier, developed at Indian Institute of Science [12]. Typical producer gas composition (dry basis) is as follows; CO (19%), H₂(19%), CH₄(1%), CO₂(11%) and rest N₂.

Experiments were conducted initially with air as the gasification medium, followed by injection of measured quantities of CO_2 along with air, as a co-reactant, to analyze CO_2 conversion levels. During gasification process, once the hot char bed was established; input air mass flux rate was fixed to desired value. Producer gas flow rate, gas composition and bed temperature at different heights were recorded. After 45 minutes of stable operation, CO_2 and O_2 were introduced, along with air, maintaining the mass flux rate. With the introduction of CO_2 , change in gas composition and producer gas flow rate were noted. Net CO_2 conversion was obtained from following equations:

$$m_{PG(1)} = m_{air} + m_{bio(1)} \tag{11}$$

$$m_{PG(2)} = m_{air} + m_{Bio(1)}$$

$$\tag{12}$$

$$m_{CO2(out1)} = m_{PG(1)} \times \chi_{CO2(1)}$$
(13)

$$m_{CO2(out2)} = m_{pg(2)} \times \chi_{CO2(2)}$$
(14)

$$CO_2 seq = m_{CO2(in)} - (m_{CO2(out2)} - m_{CO2(out1)})$$
(15)

$$CO_2$$
 conversion (%) = $\frac{CO_2 \text{ conversion}}{\frac{1}{m_{CO2(in)}}}$ (16)

Efficiency
$$(\eta) = \frac{LHV(PG)}{LHV(Biomass)}$$
 (17)

$$\Delta \eta = \frac{(\eta(2) - \eta(1))}{\eta(1)}$$
(18)

where, m_{air} = mass flow rate of air during operation, in kg/hr



Figure 1 Schematic diagram of open top downdraft gasifier

 $m_{PG(1)}, m_{CO2(out1)}, and, m_{Bio(1)}$, are mass flow rate of producer gas, CO₂ in output producer gas and biomass consumption rate respectively in case 1 (without passing CO₂), in kg/hr. $\chi_{CO2(1)}$ is mass fraction of CO₂ in the producer gas in case 1. $m_{PG(2)}, m_{CO_2(out 2)}$ and $m_{Bio(2)}$, are mass flow rates of producer gas, CO₂ in output producer gas and biomass consumption rate respectively in case 2 (while passing CO₂), in kg/hr. $\chi_{CO_2(2)}$ is mass fraction of CO₂ in the producer gas in case 2.

Hence, an attempt has been made to convert CO_2 to CO, which is an active component of producer gas.

3 EXPERIMENTAL SETUP AND MEASUREMENT SCHEMES

Casuarina wood chips were used as a fuel for gasification. All the wood chips were dried at 373 K prior to gasification process. The result of ultimate analysis of dried casuarina wood samples was obtained from CECRI, India and is listed in Table II. The chemical composition of sample wood chip is $CH_{1.62}O_{0.88}$

Table II Ultimate analysis result of dry casuarina wood

Element	Mass fraction (%)				
Carbon	42.83				
Nitrogen	0.12				
Sulphur	0.42				
Hydrogen	6.24				
Oxygen	50.39				
Molecular Weight	27.89				

Set of experiments were conducted in a 10 kg/hr opentop downdraft gasifier. Ash extraction provision was fixed at the bottom of the reactor. This ensured continuous operation for longer durations. Bed temperatures were measured using K-type thermocouples, placed at 100 mm distance in the reactor. Exit gas composition was measured using *Sick Maihak S 517* gas analyzer. CO, CO₂, CH₄, O₂ and H₂ fractions were measured using the gas-analyzer. The gas analyzer data were taken at an interval of 30 s. Exit

gas flow rate and air flow rate were measured using a precalibrated venturimeters. So, at any point of time any changes of air flow-rate into the gasifier were adjusted using a control valve, thus maintaining desired input mass flux rate of air or mixture of air, CO₂ and O₂. Char was loaded initially up to 300 mm height. Dry biomass (casuarina wood chips with moisture content < 5%) were fed above the char bed. Char bed was ignited from ignition ports and a blower was used to induce air flow inside the reactor. Reactor was kept under negative suction pressure throughout the experiments to maintain air flow at required rate. Once flame reached the wood particles; temperature, flow and gas composition data were recorded. After 45 minutes of air gasification, exact amount of CO2 were injected through regulated flow meters from pressurized CO₂ cylinders, by mixing with air at the top. As, CO₂ participates in endothermic reactions 3 and 4; introduction of CO₂ induces endothermicity in the system, thus reducing bed temperatures with increased tar levels. Dasappa has worked on the modeling of biomass char gasification with O₂/CO₂/H₂O [14]. Hence arrangement was made to introduce O_2 by mixing it with air and CO_2 . The flow rates of O₂ and CO₂ were measured using pre-calibrated flow meters.

4 RESULTS AND DISCUSSIONS

Chemical equilibrium analysis has been done under adiabatic conditions for reaction of biomass employing Gibbs free energy minimization. NASA SP-273 code was used for chemical equilibrium analysis. Results were obtained using air as an oxidizer ($\Phi = 0.25$) and subsequently adding CO₂ as a reactant. Over 45% CO₂ conversions were obtained from equilibrium studies. Reduction in adiabatic temperature was also noted with increase in CO2 fraction. Addition of CO2 along with air reduces the O₂ fraction, leading to reduction in bed temperatures and thus the reaction rates. Boudouard reaction is endothermic in nature and conversion of CO₂ to CO requires high bed temperature to be maintained in reduction zone. Mixing CO₂ with air reduces the volume fraction of O₂ which leads to reduced reaction rates and subsequently lower conversion rates of biomass/char. Rashbash and Langford [13] and Dasappa [14] suggest similar findings on working with varying fractions of O_2 with wood and char respectively, where flame extinction was observed at 14% O_2 fraction. On mixing 10% CO_2 , O_2 fraction reduces to 19%. Hence, O_2 was mixed along with CO_2 to maintain O_2 fraction as that in air (21%). Equilibrium analysis, maintaining O_2 fraction as 21% in inlet gas mixture, results in stable adiabatic flame temperatures at varying CO_2 fractions. Figure 2 shows the equilibrium analysis results for varying CO_2 input fraction. The results suggest reduction in percent of CO_2 conversion with increase in input CO_2 fraction.

In gasification system, which involves, diffusion limited heterogeneous reactions and heat loss, equilibrium condition is never attained. But, equilibrium studies provide us the tool to analyze this system, which in turn suggests the optimum working conditions. Results of equilibrium analysis were verified in experiments where drop in bed temperatures were observed with CO2 as reactant compared with with air alone. A drop in 50 K was recorded for 8.5% dilution of CO₂ with air. This issue of drop in bed temperature was resolved after increasing the inlet O_2 fraction to 21% (as that in air).



Figure 2 Equilibrium analysis - CO₂ conversion, CO₂ and CO output with varying CO₂ input fraction

A gas evolution plot showing the distribution of the three gases (CO, CO_2 and H_2) that were monitored while

conducting experiments with 15% CO₂ input (by volume fraction), is shown in Figure 3. It represents the air gasification composition with and without CO₂ injection. Changes in CO and CO₂ fractions occur when CO₂ and O₂ mixture is injected. After steady operation, CO₂ and O₂ injection is stopped and volume fraction of CO & CO₂ return to its original fractions. Figure 3 clearly shows the enhanced CO fraction which is the outcome of Boudouard and water shift reactions. Figure 4 summarizes the results at different CO₂ input fractions. Data points obtained, were averaged through the several experimental data points, and were checked for consistency through repeated experiments. CO2 conversion of 52%-55% was recorded while varying the CO_2 input fraction from 8.5 to 15%. It was observed that there was no appreciable change in H₂ fraction in the output gas. CO and CO₂ fraction followed the similar trend as shown in figure 3 with varying CO₂ fraction. It enhanced relative cold gas efficiency of system by up to 30%, as evaluated using equations 18 and 19. CO₂ conversion rates were calculated on the basis of measured input air flow rate, output gas flow rate and char left in the gasification process.

Gas composition was measured on dry basis. To account for condensed H₂O during cleaning and cooling process, species balance of C, H and O was performed, based on exit gas composition, which accounted for 10% H₂O in the producer gas. H₂O was accounted for all the calculations of mass balance to calculate total CO₂ fraction converted and captured in process. CO₂ conversion to CO was calculated as described in the methodology section. Substituting the parameters in equations 11 - 16, result in the total CO₂ percentage converted. Table III shows carbon balance for all the sequestration experiments. Case 1 considers Carbon input into the system only in the form of biomass. Case 2, on the other hand, considers Carbon input into the system in the form of biomass as well as additional CO2. The input and output mass flow matches quite reasonably. It indicates that the CO2 measurements are well accounted, and proves validity of the CO₂ conversion levels. The results of varying CO₂ input fraction are tabulated in Table III. The lower heating value of the gas varies from 3.8 to 4.3 MJ/Nm³.

CO ₂ input fraction (%)		Input Carbon in kg/hr			Output Carbon in kg/hr						
		Biomass	CO ₂	Total Input	СО	CO ₂	CH_4	Char	Total Output		
15.0	Case-1 (Air)	4.27	0	4.27	1.37	1.5	0.28	0.99	4.14		
	Case-2(Air+CO ₂)	3.30	0.88	4.18	1.51	1.9	0.25	0.77	4.43		
12.1	Case-1(Air)	3.71	0	3.71	1.43	1.33	0.2	0.86	3.82		
	Case-2(Air+CO ₂)	3.67	0.71	4.38	1.69	1.72	0.2	0.85	4.46		
8.5	Case-1(Air)	3.31	0	3.31	1.4	1.35	0.32	0.77	3.84		
	Case-2(Air+CO ₂)	2.85	0.45	3.3	1.5	1.57	0.28	0.66	4.01		

Table III: Mass balance for CO₂conversion



Figure 5 Experimental results with varying CO₂ input fractions

As seen from equation 10, at high CO_2 concentrations, the last term in the denominator becomes large compared to the other terms and the rate expression is approximated as:

$$\omega_c^{'''} = -k_2 \tag{19}$$

This is the rate expression for reaction 3, implying that this reaction becomes the rate limiting step. Similarly, at low CO_2 concentration the expression can be approximated as

$$\omega_c^{''} = -\frac{k_1 p_{CO_2}}{1 + \frac{k_3}{k_4} p_{CO}}$$
(20)

In this case the rate expression is proportional to the concentration of CO_2 [15]. By mixing CO_2 with air as a gasifying medium, p_{co2} increases and, as the above reaction states, the reaction rate of carbon (char) per unit volume changes. This has also been confirmed with the experiments conducted and are consistent with the literature [11]. Introduction of CO_2 as a gasifying medium contributes to better char conversion in reduction zone.

The increase of CO_2 fraction and increase in CO fraction in producer gas with time can be clearly seen in the Figure 3. Induction of CO_2 in input gasifying medium increases the fraction of CO_2 in the gas passing through the reduction zone, resulting in further drop in bed temperature of the reduction zone owing to endothermic reaction with char (reaction 3). Passing 20% of air through bottom nozzle helped in maintaining the bed temperature and thus enhancing the CO_2 conversion process.

4 CONCLUSIONS

The present study suggests that using low concentration CO_2 as in a typical flue gas of a combustion system, can be used to capture CO_2 and convert to energy. The issues with reduction in O_2 fraction were addressed and resolved by maintaining O_2 level as in ambient (i.e. 21% by volume). CO_2 conversion of as much as 55% was achieved with CO_2 input fraction of 15%. The enhanced cold gas efficiency was observed owing to higher char conversion rate.



Figure 4 Gas composition of producer gas with and without injection of CO₂

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