KINETIC STUDY AND MODELING OF NIGERIAN SUB BITUMINOUS COAL CHAR USING THE RANDOM PORE TECHNIQUE

ABSTRACT

The kinetics of Carboncarbon-dioxide gasification of two Nigerian sub bituminous coal chars was studied in this work using a thermogravimetric analyser (TGA). This research work presentspaper reportsthe findings on gasification of typical Enugu (Udi) and Kogi (Okaba) coal of Nigeria in carbon dioxide medium at atmospheric pressure. Proximate and ultimate analysis was carriedperformedon the coal samples based on their origin to determine their properties and values. Result shows that Udi coal has a higher heating value ascompared to Okaba coal. The effect of the temperature of gasification rangingfrom 900°C to 1000°C at different concentration of carbon dioxide of 100%, 70% and 40% at atmospheric pressure was studied. The gasification process of coal involves pyrolysis and char gasification. It was found that the reactivity of char gasification increases increasedwith the increase in the of pyrolysis heating rate, which shows demonstratesthe effect of temperature on gasification. A random pore model approach with a novel kinetic scheme was used to describe the behaviour of thecoal char.A comparison betweenTthe simulated predictive model result was compared with and the observed experimental result, was done.The simulated model result fits well with the observed experimental resultandwhich were ina good fit agreementwas observed. Based on the result obtainedresult, it was observed that the rate of carbon conversion increases with increase in concentration of carbon dioxide with overtime, and vice versa.The activation energy for the coal samples varyied depending on their origin and were found to be 97.58KkJ/mol and 103KkJ/mol for Kogi coal and Enugu coal, respectively. Products of coal gasification will serve as a promising fuel alternative.

Key Word: Coal**,** Kinetics, Char Gasification, Random Pore Model, Thermogravimetric Analyser(TGA), and carbon-dioxide.

1. INTRODUCTION

Coal is the largest most abundant fossil fuel resources in the world and has played and will continue to play an essential role in the supplyyingofthe energy needed for economic growth. The current increasingdemand for conventional fuel production, has been on the increase; as a result, a cleaner and more efficient coal technology haves gained more attention (Liu et. al., 2000). Product from coal gasification can be considered as a promising fuel for Nigerian's future growth, since there are large coal reserves in Nigeria the country, which totalof more thanover 2billion tons. Thiese coal reserves are situated in places like Enugu, Okaba, Nnewi, Nasarawa,

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Benue, and Gombe of Nigeria and there are still more reserves that have not been discovered. Different technologieslike IGCC, Integrated Gasification Combined Cycle, PFBC, Pressurized Fluidized Bed Combustion have been and are~~been~~being developed to overcome the many environmental problem associated with the use of coal as a fuel (Nwafor,1985).

The design of a gasifier depends on the kinetic parameter for the gasification of char in addition to the pyrolysis kinetic parameters. Many models have been suggested during the years, moving from the simplest one step to more complicated random pore, grain model. In most model of coal char gasification,particles are normally assumed to be spherical, uniformly porous solids but in realities this is not the case as char from the same coal can exhibit different range of morphologies and porosities (Nwafor,1985).The choice of model solution depends on the conditions of the structure of the solid phase and the transport and reaction rate. The complexity of the model depend mainly on the type of heterogeneous reaction and simplifying assumptions (Essenhigh et. al.,1991). Also the physical properties of the coal char, such as pore structure and surface area varies with conversion, depending upon gasification conditions (Bautista et. al., 1986).

The overall reactivity of coal char depends on the accessibility of the internal surface area to the gaseous reactant, which is determined by the porous structure of the coal particles (Liu et. al., 2004 & Hong et. al., 1999).

The aim of the~~is~~ present work is to study the reactivity and kinetics of char gasification at different concentrations of $CO_2$and at three different temperatures. A predictive random pore model was tested and presented. Char ~~were~~ wasprepared from Okaba and Udi subbituminous coals.

2. MATERIALS AND METHOD

2.1 Coal Sample

Okaba and Udi Subbitiminous coal samples were crushed and sieved to size fraction (75~~μm -~~- 150μm). The coal particles were then dried in oven at 70^{o}C for ~~two~~ 2h~~ours~~.

2.2 Pyrolysis of coal

The subbituminous coal sample was pyrolyzed using a furnace at the temperature of 800^{o}C~~at~~ witha heating rate of 10^{o}C/min and 30^{o}C/min in ~~N~~nitrogen

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atmosphere. The proximate and ultimate analysis for the coal char was carried out.

2.3 Coal Char Gasification in a Thermogravimetric Analyser (TGA)

The Thermo Gravimetric Analysis (TGA) facility at the National Research Institute for Chemical Technology (NARICT) Kaduna, Nigeria was incorporated to obtain the gasification rate. The TGA was used to monitor the weight change by isothermal analysis.Char samples from Okaba coal and Udi coal were gasified in a Thermogravimetric Analyser (TGA) ~~at a~~ in the temperature range ~~of~~from 900^{o}C to 1000^{o}C. The computer controlled TGA constantly measures the weight of materials as a function of time and temperatures. See figure 1. for schematic diagram of TGA.

Gasification of coal char with different carbon dioxide concentration (100%, 70%, and 40% CO_2) was performed at 900^{o}~~C –~~-1000^{o}C at a heating rate of 5^{o}C/min for different carbon dioxide concentrations. Readings~~for the~~of mass change ~~in mass~~ of the char ~~was~~ were taken at every 10mins interval. Nitrogen was used to vary the concentration of carbon dioxide. For each run 20 mg of char was weighed and placed onto the crucible, ~~which is~~placed in the centre of the reaction chamber of the TGA. The volumetric flow rate of the gaseous phase was ranged between 0.2 to 0.6 ~~litre~~L/h~~r~~.

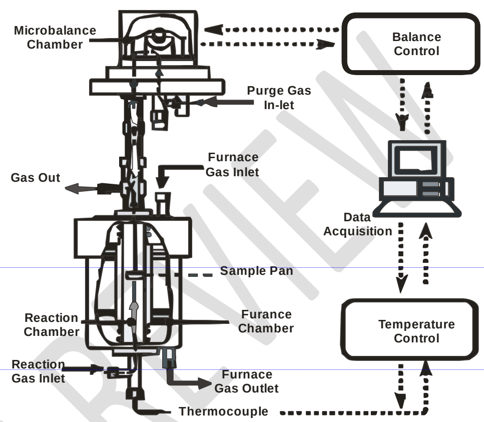

Figure 1: Schematic diagram of the TGA.

The ~~power~~ capability of X-ray diffraction method (XRD) equipped with a diffractometer was used to analyze the crystalline ~~of~~properties of samples. The X-ray patterns were recorded using Cu Kα radiation. The power supply was 55kV and 50mA. The peaks were obtained at 2θ, scan speed at 0.4~~sec~~/step increment of 0.003 degree. Peaks of structures were compared with those of the reference materials. The XRD analysis was carried out at the National Steel and Raw Material Research Institute, Kaduna.

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2.4 Theory

The mathematical model of coal char gasification is presented. ~~For the purpose of~~To perform modeling~~ing~~, it is essential to know the kinetics of the reaction taking place during gasification (Yu et. al., 2002 and Wall et. al., 2002).The ~~well-known~~ generally accepted mechanism of the reaction between carbon dioxide and coal char, involving free carbon active site follows the form below: (Bischoff, 1965 and Colomba, 2002).

$$C_{(s)} + CO_{2(g)} \rightarrow CO_{(g)} + C(O)_{(s)}$$

(ΔG^o= 128KJ/mol)

$$C(O)_{(s)} \rightarrow CO_{(g)} + C_{(s)}$$

The reaction rate is equal to the rate of decomposition of C(O)$_s$ intermediate.

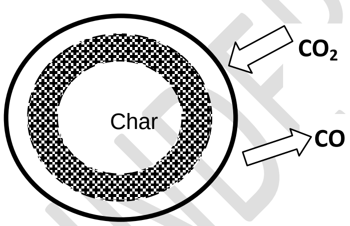

Fig. 2: Schematic diagram of char gasification

The rate r_c of hetrogenous gas/solid reaction for coal char conversion at a given level of carbon conversion and at constant reaction gas pressure can be expressed as Eq. 1~~:~~ (Bautista et. al., 1986 &Liu et. al., 2003).

$$r_c = \frac{\partial c_c}{\partial t} = \frac{1}{1-x_s}\frac{\partial x_s}{\partial t} = KC_c C_{c_{O_2}} \qquad (1)$$

In complementary studies (Hurt & Haynes, 2004)~~stresses~~ found that~~,it have been found that~~ the amount of stable complex remaining on the surface of the char after partial gasification and subsequent removal of higher temperature by temperature programmed desorption on the char also ~~increases~~ increased monotonically with conversion.This is due to the gas diffusion especially ~~in~~at high temperatures.Distinguishing intrinsic and effective reaction rate is indeed particularly difficult, and this led to the usage of apparent reaction rate as the char gasification reactivity index. The apparent reaction rates (dX/dt) are obtained indirectly from the derivative ~~calculation~~ of carbon conversion (X) versus reaction time (t).

The rate of a heterogeneous gas/solid reaction at a given level of conversion X$_s$ and at constant gas pressure can be expressed ~~in the following form~~according to Eq. 2 (Moor, 1998 and Smith, 1982).

$$Rate = \frac{1}{1-X_s}\frac{\partial x_s}{\partial t} = KC_{(s)} \qquad (2)$$

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Char conversion *X*, (dry, ash free.) is expressed as Eq. 3:

$$X = 1 - \frac{W}{W_o} \qquad (3)$$

Where W and W_o are the weights of the remaining solid at time t and initial weight of char at t=0, respectively (Smith, 1981 and Levenspiel,1981).

In thisour research work, the random pore model is applied into determineing the kinetics of the coal charformation, since it considers the physical structural change during the gasification reaction. The model is applied together with a Temperature ProgrammedReaction (TPR) of char gasification reaction carbon dioxide medium. TPR is a method wherein the weight loss is measured during the heating of a solid sample in a reactive gas medium to a desired pre-determined temperature at constant heating rate (Essenhigh, 1991 and Benfell, 2001).The basic idea of random model pore is that gasification follows different steps: adsorption of the reacting gas on the surface, reaction and diffusion of adsorbed species and finally release of carbon containing gases. The following assumptions are present in the model; char is described as a pure carbon matrix, ashes if present are considered as inert, char mass loss during gasification is due to desorption of carbon containing gases. The carbon particle is assumed to be spherical and relatively equal. The reaction gas concentration is uniform throughout the carbon particle.The RPM also assumes that the reaction is initiated on the surfaces of the pores in chars. As further reaction occurs, a layer of gas product is formed around each pore which separates the growing reaction surface of the carbon from the gas reactant within the pores. Reactant diffuses through the product layer to reaction surface where chemical change occurs. Although the RPM considers the physical structural changes during the gasification reaction, it does not consider the random pore overlapping and neglects all diffusion resistances in the char reaction.

The derived random model equation is obtained as Eq. 4:

$$\frac{dx_c}{dt} = k_o e^{-(E_a/RT)} C_{co_2} (1 - x_c) S_{gc} \sqrt{1 - \psi \ln(1 - x_c)} \qquad (4)$$

Where ψ = Structural parameter, as defined by Eq. 5

$$\psi = \frac{4\pi l_o}{\rho_c S_o^2} \qquad (5)$$

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The integral version of the random pore model ~~equation~~ follows the form given in Eq. 6:

$$x_c = 1 - Exp\left(\frac{1}{\psi} - \frac{\psi k_o^2 s_{gc}^2 \left(C_{co_2}\right)^2 e^{-2E_a/RT} t}{4}\right) \quad (6)$$

3. RESULTS AND DISCUSSION.

Normally the char gasification of coal conversion is the rate ~~controlling~~determiningstep ~~for~~ofthe overall process. Experiments were performed to determine the rate of CO_2 gasification of the coal char at temperatures ranging from 1173K to 1273K, with data of conversion rate versus reaction time. A gas-solid reaction kinetic model has been developed and applied to the Char-CO_2 systems. The model prediction compares favourably to the measured data.

Figure 3 below shows raw data of TGA results for coal char gasification. Weight loss measurements were made under isothermal and transient conditions~~, in the TGA~~at atmospheric pressure.

Using the char sample produced from the sub-bituminous coal, test at various temperatures and gas concentrations indicated that 1000K was the highest temperature at which there were negligible mass transport effects during char gasification of 75~~µm –~~ – 150µm diameter particles~~in the TGA~~.

At higher temperatures, the overall mass loss rates were found to depend on the amount of material placed in the balance pan of the TGA, indicative of mass transfer limitation.

Based on the result obtained, which shows that the carbon conversion versus gasification time takes an S-Shapedform. That is to say the reaction rate initially increases during the gasification reaction.

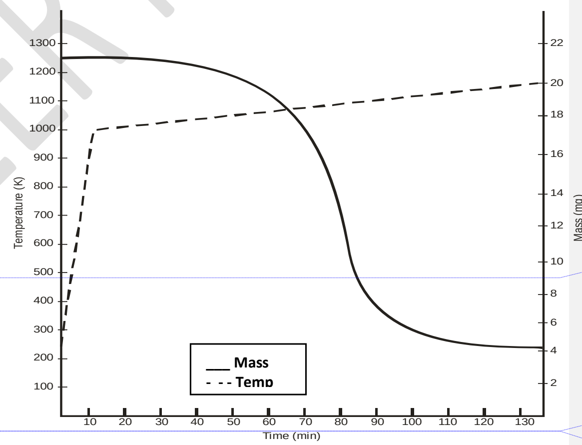

Figure 3: Non isothermal combustion and gasification of coal char in CO_2 at 5^oC/min.

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3.1 The Micro-Structural Parameter

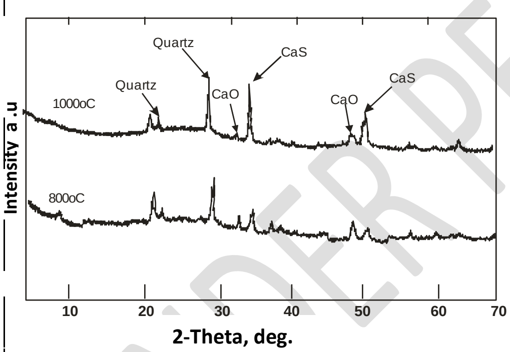

Figure 4: X-ray Diffraction pattern for Udi coal char pyrolized at two different temperatures, ~~(~~1000°C and 800°C, ~~)~~ respectively~~.~~

The~~above~~ XRD ~~result~~in Fig. 4 shows the XRD pattern for Udi coal char sample ~~pyrolized~~pyrolyzed at 800°C and 1000°C.The ~~M~~main peaks of crystal structure ~~which can be detected by the~~detected in XRD consist of quartz (SiO_2), ~~C~~calcium oxide (CaO), and ~~C~~calcium Sulfide (CaS). Peaks of crystal structures were obtained by comparing with structures of reference materials ~~as maintained~~available in the XRD coal library and based on the elements in the sub-bituminous coal. ~~From the~~se ~~result~~s ~~of the XRD, it is~~One can observe~~d~~ that most of crystal structures peaks of char sample from pyrolysis at 1000°C are higher than those of char sample from pyrolysis at 800°C, except for calcium oxide. It is well recognized that ~~calcium oxide~~CaO is a primary specie, which act~~s~~ as acatalyst ~~for~~ inthe gasification process. Thus, low gasification reactivity of char samples is probably due to the formation of inert crystal structures and the consumption of ~~calcium oxide~~CaO at higher pyrolysis temperature. (Campbell et. al., 2002).

Table 1.: Proximate and Ultimate analysis for the coal samples~~.~~

		Enugu (Udi) Coal	Kogi (Okaba) Coal
1	**Average particle Size**	75~~µm~~-150 µm diameter.	75~~µm~~-150 µm diameter.
2.	**PROXIMATE ANALYSIS**		
	Moisture %	4.8	4.6
	Ash%	20.4	22.8
	Volatile matter %	40.8	35.9
	Fixed carbon%	41.3	36.8

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3	**ULTIMATE ANALYSIS**		
	Total ~~Sulphur~~ sulphur%	0.4	0.9
	Carbon%	63.5	61.4
	Hydrogen%	2.59	1.7
	Nitrogen%	1.6	1.3
	Oxygen%	8.3	11.5
	Calorific Value ~~KJ~~kJ/~~K~~kg	24,780	22,350

Proximate analysis is the most often used ~~analysis~~strategyfor characterizing coalsamples in connection with their utilization and usefulness. The proximate analysis on the coal samples in Table1. shows that Udi coal has a higher risk of spontaneous combustion or ignition than Okaba coal, because it contains more volatile matter.The ~~altimate~~ultimateanalysis indicates the various elemental chemical constituents as shown in Table 1.Theanalysis ~~shows~~indicatesthat Udi coal ~~contains~~has aslightly ~~highly~~greatercarbon content and calorific(heating value) than Okaba coal.~~, though,~~Nonetheless,both coal samples are suitable for fuel.

3.2 Comparism of Experimental result with Model result at different temperatures and carbon dioxide concentrations.

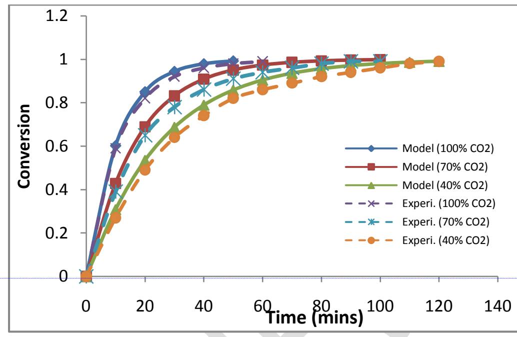

Figure 5: Graphical result of Udi char gasification at 1000°C for different CO_2 ~~Concentration~~ concentrations

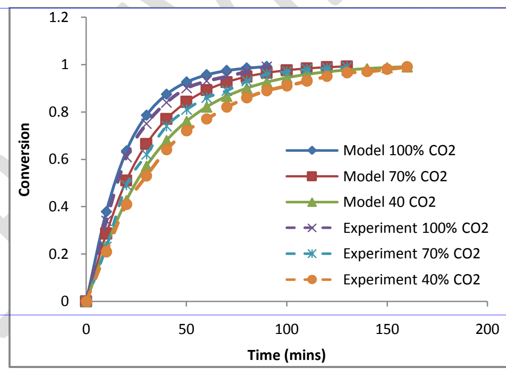

Figure 6: Graphical result of Udi char gasification at 950 °C different CO_2concentrations~~concentration.~~

Figure 6: Graphical result of Udi char gasification at 950°C different CO2 concentration.

From the trends of results, it was observed that thecomplete conversion of char in carbon dioxide is attained with less time at high CO_2 concentration. But as the concentration of CO_2 is reduced, the rate of conversion is reduced, thereby increasing the time required for conversion

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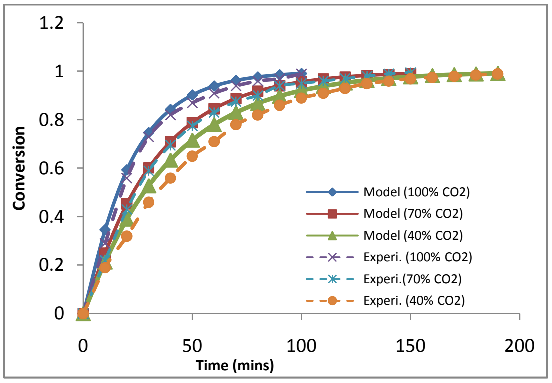

Figure 7: Graphical result of Udi char gasification at 900°C for different CO_2concentrations~~Concentration~~

From the graphical result of thermogram, it can be observed that the model does predict the rate of coal char gasification in a carbon-dioxide atmosphere.

The result for that of Okaba coal char gasification show asimilar trend ~~like that of~~withUdi coal char. The time taken for completion conversion is slightly ~~higher~~longerthan that of Udi Coal char.

Figures 5 to 10 (Udi and Okaba coal) shows the steady rate variation with conversion (time) for sub-bituminous coal char gasified at different CO_2 concentration, at atmospheric pressure at 1000°C, 950°C, and 900°C, respectively A monotonically increasing specific gasification rate is observed in all cases.

The thermogram for simulated model and theexperimental results~~it can be observed~~revealthat the rate of conversion of coal char deceases with decreasingcarbon dioxide~~in~~ concentration~~of~~. As the concentration of carbon dioxide increases, the time for complete conversion decreases.

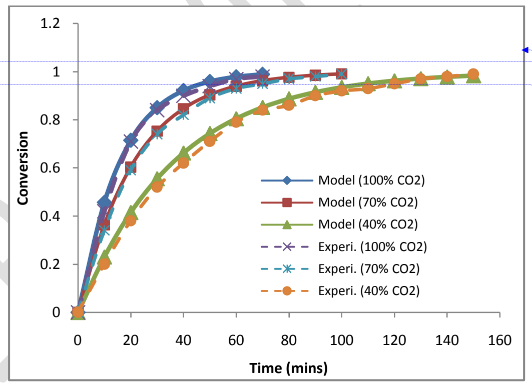

Figure 8: Graphical Result of Okaba char gasification at 1000°C for different CO_2 Concentration.

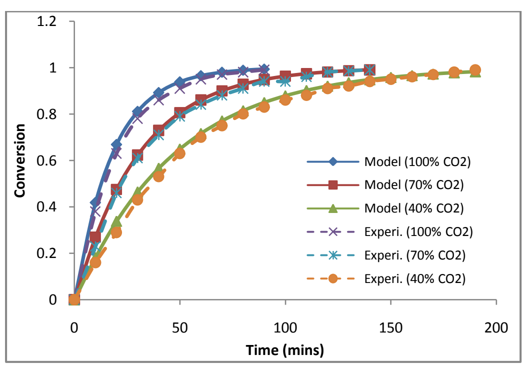

Figure 9: Graphical result of Okaba char gasification at 950°C different CO_2 concentration.

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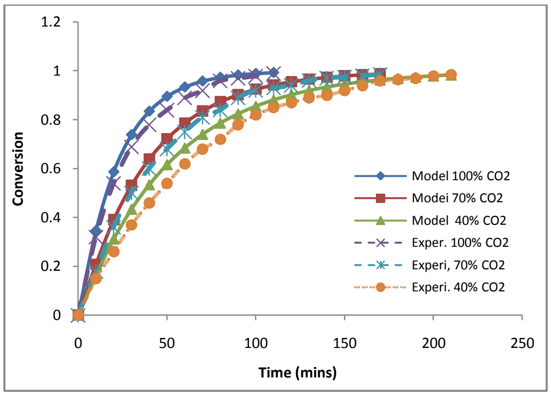

Figure 10: Graphical result of Okabachargasification at 950^{o}C different CO_2 concentration.

From all the thermogram ~~above~~ in Fig. 10it is observed that at higher level of conversion, a steep rise in the rate of conversion ~~against~~overtime is observed. Also, it was observed that increase in concentration of carbon dioxide leads to increase in the conversion.

4. CONCLUSION

~~The~~We carried out the kinetic study and modeling of Nigerian sub-bituminous coal char using the random pore technique~~ was carried out~~. Proximate analysis ~~on~~ofthe coal samples shows that Udi coal has a ~~higher~~greaterheating value ~~as well as~~and poses a higher risk of spontaneous combustion than Okaba coal ~~because~~ as it contains~~higher~~morefixed carbon and volatile matter~~content~~. ~~Though~~Nonetheless, both coal samples are suitable for fuel.

The ~~Char~~char–CO_2 gasification is an endothermic reaction at relatively slow rate and the single product evolved is ~~Carbon~~ carbon monoxide, which ~~makes~~renders theanalysis simple and reliable.

~~It can be noted that t~~Thereactivity of char gasification and increase of pyrolysis heating rate is due to the development of active surface area at higher pyrolysis heating rates ~~and also~~andto theincrease of the surface area ~~during the course of~~during gasification. (Suuberg et.al.,1996).

Based on ~~the~~ results obtained from both coal samples, it was observed that concentration of the gasifying gas (carbon dioxide)affected the conversion rate. A monotonically increasing specific rate of conversion with time was observed in both cases. As the reaction temperature and concentration of the gasifying gas ~~(Carbon dioxide)~~increases, the rate of conversion also increases, but the time required for complete conversion decreases.

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