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Combined gasification of lignite coal: Thermodynamic and application study

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ABSTRACT

Combined gasification (a combination of steam gasification and CO_2 gasification) without air is an important process for research in coal gasification to reduce the steam generation energy in the gasification system and also to enhance the slow CO_2 gasification reaction. A thermodynamic study involving the effect of temperature, pressure and feed CO_2 and steam ratios in gasification of lignite coal was studied in this paper. The product generation trends of syngas and methane with carbon (in coal) conversion were studied in detail. The carbon (in lignite coal) was converted completely at a lower temperature than pure carbon in the combined gasification. Some applications of the gasifier product gas were also studied. Combined gasification offers great advantages to produce syngas of exact ratio in one step for use in petrochemical manufacture and fuel cell systems. The complete carbon (in coal) conversion occurred beyond the thermoneutral gasification temperature in the study. The combined gasification process was a useful way for CO_2 utilization reducing the net CO_2 emission to the atmosphere.

1. Introduction

Coal is a natural resource available in many countries worldwide. It is a carbon source used mainly for energy generation. Some other coal based value added products include syngas (H_2 + CO). Syngas can be utilized in many ways – for electric power generation in gas engine or gas turbine, for Fischer–Tropsch (FT) synthesis of liquid fuels, for production of gaseous products such as synthetic natural gas and also as fuel in fuel cells. Coal gasification is a popular technology to produce syngas from coal. Some researchers are also working on underground coal gasification (UCG) which is a potential clean coal technology to convert coal gas in situ into syngas using dry air [1].

Traditionally coal gasification is done using steam and air in coal gasifiers. Neogi et al. have experimentally studied coal gasification in a fluidized bed reactor using steam as the fluidizing medium and simulated the dynamic behavior and steady state performance of the gasifier to a mathematical model [2]. Bayarsaikhan et al. have experimentally studied the steam gasification of a Victorian brown coal in an atmospheric bubbling fluidized-bed reactor with continuous feeding of coal [3]. Sekine et al. have studied the reactivity and structural change of coal char during steam gasification [4]. Peng et al. have studied the reactivities of in situ and ex situ coal chars of bituminous coal, subbituminous coal and lignite in steam gasification and found that the reactivities of the resulting in situ chars were up to six times higher than that of the corresponding exsitu chars [5]. Foong et al. have studied the coal gasification in a spouted bed reactor to produce syngas using mixtures of steam and air and compared the results to commercial moving and fluidized bed systems [6]. Chatterjee et al. have studied the gasification of high ash India coal in a laboratory scale, atmospheric fluidized bed gasifier using steam and air as fluidizing media and compared the predicted and experimental data to show similar trends [7].

Coal gasification can also be done using CO_2. Irfan et al. have reported a detailed review of coal gasification in CO_2 atmosphere and its kinetics since 1948 [8]. Ethan et al. have studied the effect of CO_2 gasification reaction on oxy-combustion of pulverized coal char and reported new calculations that give new insight into the complexity of the effects from the CO_2 gasification reaction [9]. Wei-Biao and Qing-Hua have found a general relationship between the kinetic parameters for the gasification of coal chars with CO_2 and coal type [10]. Marcel et al. have experimentally studied the coal gasification at pressure by mixtures of carbon dioxide and oxygen and reported that experimental results could be satisfactorily explained by means of a model based on thermodynamic equilibria [11]. Coal is also gasified with biomass [12] and wood [13].

Use of air in coal gasification results in nitrogen dilution of the gasifier product gas which reduces the value of the syngas produced. Hence gasification without air/oxygen input is beneficial to applications that use syngas. Thermodynamic studies are the starting points to assess the feasibility of new processes. Thermodynamic analysis is the guiding step to determine the

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Nomenclature

UCG	underground coal gasification
FT	Fischer–Tropsch
WGS	water gas shift
GaCR	gasifying agent to carbon (in coal) ratio
TNP	thermoneutral temperature
kJ	kilo Joule
SNG	synthetic natural gas
SOFC	solid oxide fuel cell
PEMFC	polymer electrolyte membrane fuel cell
CCGT	combined cycle gas turbine

optimum conditions for process development. Thermodynamic studies are also popular in coal gasification area. Eftekhari et al. have used a chemical equilibrium model to analyze the effect of process parameters on product composition and use it for an exergy analysis of underground coal gasification [14]. Dufaux et al. have investigated the gasifier involving underground coal gasification using mathematical models based on the simultaneous resolution of the different thermodynamic equilibria of the gasification reactions [15]. Ng and Lipinski have done thermodynamic analyses for solar thermal steam and dry gasification of anthracite, bituminous, lignite and peat coals [16]. Giuffrida et al. have analyzed the thermodynamic performance of IGCC power plants based on an air-blown gasifier [17]. Prins et al. have studied the effect of fuel composition on the thermodynamic efficiency of gasifiers and gasification systems using a chemical equilibrium model to describe the gasifier [18]. Wang et al. have conducted a thermodynamic equilibrium analysis of hydrogen production by coal based on Coal/CaO/H_2O gasification system [19].

Most of the gasification thermodynamic studies have reported the use of Gibbs free energy minimization algorithm [20–22] or Gibbs free energy routine of aspen plus for biomass gasification [23]. HSC Chemistry software has also been used by some researchers in thermodynamic studies related to coal gasification [24], biomass gasification [25,26]. Syed and Isam have performed the thermodynamics analysis of gasification process using the Gibbs energy minimization approach through Lagrange multiplier method to study the three different methods of gasification using air, air-steam and solar-steam to convert bituminous (RTC) coal into syngas [27].

Use of excess steam and excess CO_2 in gasification processes is common to enhance the chemical reactivity in gasification [28]. Steam generation is a very energy consuming task as water has high latent heat of vaporization. CO_2 does not require phase change but the CO_2 molecule is very inert and its chemical reactivity in gasification is low. Combined gasification offers an advantageous middle path to reduce the disadvantages of pure steam or CO_2 gasification. The syngas produced by steam gasification has very high hydrogen content while that produced by CO_2 gasification has significant amount of CO. The high hydrogen containing syngas cannot be generally used in FT synthesis. The syngas required for syngas based applications *e.g.* FT synthesis requires adjustment of syngas ratio (preferably between 1 and 3). This adjustment is usually done by coupling water gas shift (WGS) reactor (sometimes reactors in stages) to the gasifier. The following reaction occurs in the WGS reactor:

$$CO + H_2O = CO_2 + H_2 \tag{A.1}$$

These water gas shift reactors operate at low temperatures than the gasifier *e.g.* high temperature shift reactor operates around 400–500 °C. These are catalytic reactors requiring selective WGS catalyst for operation. The WGS reaction using commercial Fe based catalyst requires high steam input to make the ratio of feed $H_2O/CO = 4.5$. This additional steam requirement is a huge energy consuming process. Kiso and Matsuo have studied the factors to improve the thermal efficiency of fuel production and electricity generation by dry coal feed gasification and have suggested that the primary cause of thermal efficiency loss is the use of steam in the water-gas shift reactor [29]. Moreover the sulfur compounds in coal can poison the WGS catalyst. The WGS reaction is also very slow and requires huge reactors.

Combined gasification of coal using steam and CO_2 in appropriate ratio can produce syngas of the desired syngas ratio in one step. Although several such studies on coal gasification have been published in literature, coal gasification using both steam and CO_2 simultaneously has not yet been studied extensively although such study for biomass is recently reported [30]. This study aims to study the feasibility aspects of coal gasification using steam and CO_2 simultaneously and understand the trends in product generation and their utility to some specific applications.

Different types of coal have been used in various research studies. Lignite coal gasification studies have also been reported in literature [31–34]. Alexander et al. have studied the pyrolysis and gasification behavior at operation conditions relevant to industrial scale entrained flow gasifiers and have reported high level of conversion of Rhenish lignite coal at atmospheric pressure [35]. Shayan et al. have experimentally studied the effect of coal feed rate, coal particle size, and steam/O_2 ratio, and their interactions on the quality of syngas produced from fluidized bed gasification of lignite coal [36]. Lignite coal is reported for its high activity in gasification [37–39] and hence was selected for this study.

Coal gasification is affected by the inorganic matter content of the coal char [40–43]. Coal gasification is also slightly affected the particle size of coal [44,45]. Thus, barring the extreme mass transfer and kinetic limitations, thermodynamic study of gasification can be of universal use to predict the conversions and product distributions.

Coal gasification involves many chemical reactions. The gasification reactions mentioned in earlier literature are given below:

$$C + CO_2 = 2CO \tag{A.2}$$

$$C + H_2O = H_2 + CO \tag{A.3}$$

$$C + 2H_2 = CH_4 \tag{A.4}$$

$$2C + 2H_2O = CH_4 + CO_2 \tag{A.5}$$

$$C + 2H_2O = 2H_2 + CO_2 \tag{A.6}$$

$$CH_4 + H_2O = CO + 3H_2 \tag{A.7}$$

$$2CO = CO_2 + C \tag{A.8}$$

$$CO + H_2O = H_2 + CO_2 \tag{A.1}$$

The coal gasification products reported in literature are H_2, CO, CO_2, CH_4, C_2H_6, C_3H_6, C_3H_8, H_2O, and C. The coal gasification reactions and their products have been discussed in literature earlier [46–51].