COAL GASIFICATION

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Summary

Coal gasification is one of the clean coal technologies. The purposes to convert coal into coal gas are stated in this article. The emphasis is put on integrated coal gasification combined cycle as one of the applications of coal gasification because of its higher efficiency and the greatest potential for meeting stringent emission control requirements. Coal gasification reactions, thermodynamics, and kinetics of gasification reactions are presented in brief. After coal gasification processes are divided into several categories, 4 types of coal gasification processes are demonstrated respectively, these are moving bed, fluidized bed, entrained bed, and molten bed. Some typical or advanced gasifiers introduced are restricted to these in commercial operation and those in large size pilot plant. They are Lurgi gasifier and BGC-L gasifier for the moving bed process, Winkler gasifier, HTW gasifier and U-Gas or KRW gasifier for the fluidized bed process, and the K-T gasifier, Prenflo, SCGP, TCGP and Dow gasifier for the entrained bed process. These gasifiers are discussed according to their methods for feeding coal and ash removal, principal operating conditions, gasification efficiency, and other process characteristics.

1. Introduction

Coal gasification, just as the term suggests, means that coal can be gasified into gas, which is then largely used as fuel gas or chemical feedstock, and sometimes used as industrial reducing gas, in other words coal can be converted into a more easily utilized gaseous form. The technology to convert coal into gas is hardly new and coal gasification has been developed and abandoned periodically during the past two centuries. Fuel gas was first produced commercially from coal in the early nineteenth century. In 1839 the first fixed-grate producer was introduced. In 1852 cyclic operation to produce water gas was invented. During 1900–1920 “the mechanical producer” was developed which in a sense had already reached its present form. The synthesis of ammonia and methanol from water gas grew strongly during 1920s. After the Second World War cheaper natural gas and oil displaced coal gas from many of its former user in the world except in South Africa and some developing countries. In response to the first oil crisis in 1973 the altered price and availability relationship between oil and coal increased a strong incentive in coal gasification technology, therefore, a lot of coal gasification processes had been developed. Some processes developed are quite successful; these processes are called the second-generation technology of coal gasification.

Why should coal be gasified? One reason for coal gasification is to make up the shortage in liquid and gaseous fuels. Now the world economy depends on energy provided principally by oil and natural gas, but in the twenty-first century world petroleum and natural gas production will begin to decline because of increasing world population and aspirations for an improvement in the quality of life. Therefore the need for a transition to other energy sources is necessary. Coal is an abundantly available energy source and coal gasification is a primary way to produce liquid fuels for transportation and gaseous fuels for heating and chemical production. Some technologies for producing liquid fuels and gaseous fuels have been commercialized for quite some years and some are still under development. In many cases these technologies are available, obstacles for commercial application have been the high cost. Another reason for coal gasification is necessity to develop advanced power generation system. The world electric utility industry has greatly depended on the relative abundance of coal. Clearly, the expanded use of coal is still vital to future electrical power generation and the expanded use of coal must be carried out in an environmentally acceptable and economically competitive manner. For conventional pulverized coal fired power plants, it is difficult to meet these requirements. As an advanced power generation system, integrated coal gasification combined cycle (IGCC) offers an attractive approach to produce electrical power at high efficiency and has shown the greatest potential for meeting stringent emission control requirements. Therefore coal gasification is a very important clean coal technology from the viewpoint of sustainable development. We should pay attention to coal gasification technology and its applicable technologies.

Coal gas is a mixture of gases. This mixture is usually made up of carbon monoxide (CO), hydrogen (H_2), carbon dioxide (CO_2), nitrogen (N_2), and varying amounts of methane (CH_4). Coal gas is classified by heating value. Low-heat-content (also termed low-Btu) gas has a heating value below 7 MJ m^{-3} and is mainly a mixture of nitrogen and carbon dioxide, with the combustible components, i.e. carbon monoxide, hydrogen, and methane. High-heat-content (or high Btu) gas has a heating value about 37MJ m^{-3} and consists mainly of methane. The gas is also called substitute or synthetic natural gas or SNG. Medium-heat-content (or medium Btu) gas has a heating value between 7 and 15 MJ m^{-3}. Gas at the lower end of this range consists of carbon monoxide and hydrogen, while gas at the higher end contains more methane.

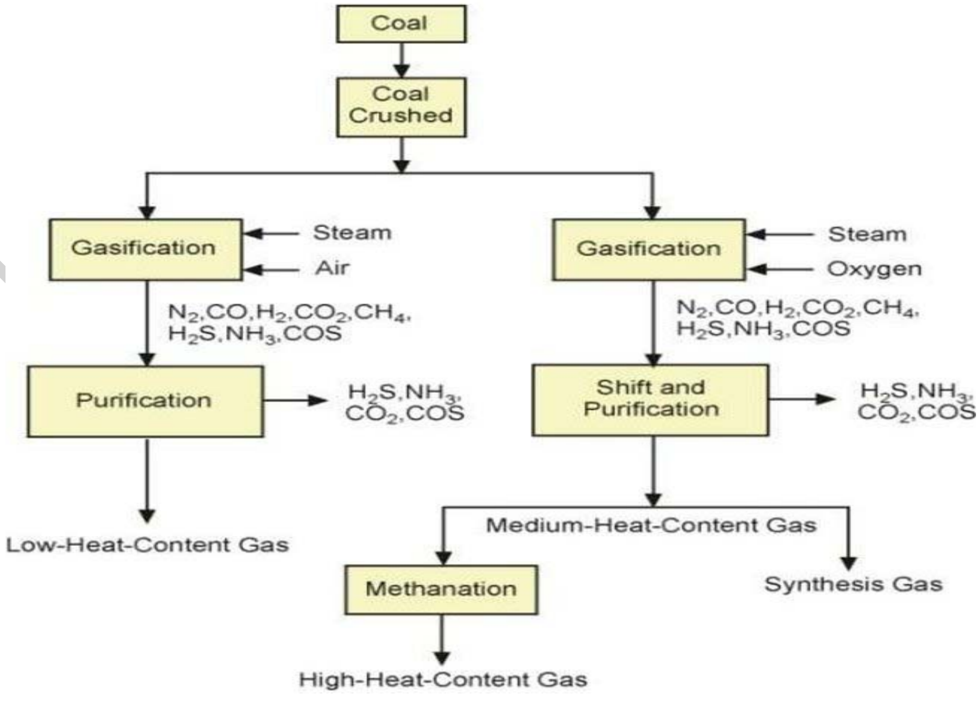

Figure1. Coal gasification process

The production of each of these gases is shown in Figure 1.

Coal is first crushed and sometimes dried, then fed into gasifier, in which coal reacts with steam and either air or oxygen. The gasification reaction usually occurs at high temperatures from 800 to 1900°C and high pressure up to 10 MPa. When coal is burned with less than a stoichiometric quantity of air, with or without steam, the product is low-heat-content gas, which after purification can be used as fuel gas. Utilizing oxygen in place of air produces medium-heat-content gas. The gas produced is used as synthetic gas; some of CO in gas must be reacted with steam to get additional hydrogen. This step is called shift conversion, which sets up the proper ratio of gaseous components depending on the requirements of different synthetic gases for producing liquid fuels, SNG, ammonia, or methanol.

2. Chemistry of Coal Gasification

Coal gasification involves two distinct stages, i.e. devolatilization followed by char gasification. Devolatilization occurs quite rapidly as the coal is heated above 400°C. During this period, the coal structure is altered, producing solid char, tars, condensable liquids, and light gases. The distributions of devolatilization product under various heating conditions had been determined. It is found that devolatilization products in an inert gas atmosphere are very different from those in an atmosphere containing hydrogen at elevated pressure. After devolatilization char then gasifies at a lower rate, the specific reactions that take place during this second stage depending on the gasification medium.

2.1 Rapid Devolatilization and Hydrogasification

In an inert-gas atmosphere, coal pyrolysis is divided into three overall processes. Initially, the coal undergoes a kind of depolymerization reaction that leads to the formation of a metastable intermediate product. The product then undergoes cracking and recondensation to result in the evolution of primary gases and oils, yielding semichar. After the rate of devolatilization has passed a maximum, the third reaction, in which the semichar is converted to char primarily through the evolution of hydrogen, becomes important.

In a hydrogen atmosphere at elevated pressure, additional yields of methane or other light gaseous hydrocarbon can result during the initial coal gasification stage from: (a) direct hydrogenation of coal or semichar because of active intermediate formed in coal structure after coal pyrolysis. The direct hydrogenation should also result in an increase in the amount of coal carbon that is gasified, (b) the hydrogenation of other gaseous hydrocarbons, oils, tars, and carbon oxides.

The kinetics of the rapid-rate reaction between gaseous hydrogen and the active intermediate has been studied. It is shown that direct hydrogenation depends on hydrogen partial pressure (P_{H2}). Greatly increased gaseous hydrocarbons produced during the initial coal gasification stage are extremely important in processes to convert coal into SNG. Several coal hydrogasification processes had been designed and tested and had shown significant increase in overall process efficiency.

2.2 Coal Gasification Reactions

When coal is gasified under practical conditions of coal gasification, following reactions will take place during gasification:

$$Coal \xrightarrow{Pyrolysis} Char(C) + \text{Coal volatiles(VM)} \quad (1)$$

$$VM + H_2 \xrightarrow{Hydrocracking} CH_4 - \Delta\text{H} \quad (2)$$

$$VM + H_2O \xrightarrow{Gasification} CO + H_2 + \Delta\text{H} \quad (3)$$

$$C + 2H_2 \xrightarrow{Hydrogasification} CH_4 - \Delta\text{H} \quad (4)$$

$$C + H_2O \xrightarrow{Gasification} CO + H_2 + \Delta\text{H} \quad (5)$$

$$C + CO_2 \xrightarrow{Gasification} 2CO + \Delta\text{H} \quad (6)$$

$$CO + H_2O \xrightarrow{\text{Shift Conversion}} CO_2 + H_2 - \Delta\text{H} \quad (7)$$

$$C + O_2 \xrightarrow{Combustion} CO_2 - \Delta\text{H} \quad (8)$$

Coal volatiles include all gases, tar, and light gaseous hydrocarbon. The pyrolysis reaction occurs under all conditions of gasification. The tar undergoes hydrocracking and gasification reactions producing CH_4, H_2, and CO. The char undergoes hydrogasification and gasification reactions producing CH_4, H_2, and CO.

The shift conversion reaction takes place under all conditions of gasification. Sulfur, nitrogen, and oxygen present in coal are converted to H_2S, NH_3, and organic compounds containing sulfur and nitrogen and H_2O respectively. The extent of coal conversion depends on thermodynamics and kinetics of these reactions.

2.3 Thermodynamics

The gasification reactions 5 and 6 are endothermic with an enthalpy ΔH of about 120-160 KJ mol^{-1}. They are favored at high temperature above 1000°K. The shift reaction and the hydrogasification reaction are moderately exothermic with an enthalpy ΔH of about 32-88 KJ mol^{-1}. They are favored at low temperatures below 1000°K. The combustion reaction 8 is a strong exothermic reaction with an enthalpy ΔH of about 376KJ mol^{-1}.

The equilibrium constants of reaction 8 show that the reaction has no significant thermodynamic limitations at temperature up to 2500°K. Under practical conditions of coal gasification, the combustion reaction proceeds to completion while the gasification reactions and shift reaction approach pseudoequilibrium.

2.4 Coal–Char–Gasification Kinetics

Although the initial coal-gasification stage is completed in seconds or even less at elevated temperatures, the subsequent gasification of the coal chars produced at the initial coal-gasification stage is much slower, requiring minutes or hours to obtain significant conversions under practical conditions. Since reactor designs for commercial gasifiers are largely dependent on coal char, i.e. coal char reactivity, the kinetics of coal char gasification systems has been extensively investigated. Some results about coal char gasification reaction 4, 5, and 6 will be introduced respectively here.

2.4.1 Coal–Char–Hydrogen Reaction

For the gasification of coal chars in pure hydrogen, gasification rates have been given by equation (1):

$$\mathrm{w} = \frac{\mathrm{dx/dt}}{1-\mathrm{x}} = \frac{\mathrm{k_H P_{H_2}^2}}{1+\mathrm{K_H P_{H_2}}} \tag{9}$$

where k_H, K_H =kinetic parameters that depend on temperature and the reactivity of the char; P_{H_2} = hydrogen partial pressure; and w = specific carbon gasification rate (min^{-1}). Based on equation (1) gasification rates increase significantly with increasing hydrogen partial pressure. At sufficiently high hydrogen partial pressures, the reaction rates increase almost linearly with pressures.

2.4.2 Coal–Char–Carbon Dioxide Reaction

Reaction 6 is significantly different from reaction 4. At atmospheric conditions, for example, reaction rate in pure carbon dioxide is generally greater than rate in pure hydrogen by a factor of 10^2 to 10^3. The reaction rate depends mainly on temperature and pressure. The reaction rates increase with temperature. Arrhenius activation energies of char-CO_2 reaction for different chars have been found to vary from 80 to 200 KJ mol^{-1}. Many researchers have used Langmuir type adsorption equations to represent gasification rate as follows:

$$w = \frac{dx/dt}{1-x} = \frac{k_C P_{CO_2}}{1+\mathrm{K}_{C,1}P_{CO} + \mathrm{K}_{C,2}P_{CO_2}} \tag{10}$$

where k_C, $\mathrm{K}_{C,1}$, $\mathrm{K}_{c,2}$ = kinetic parameters that depend on temperature and the reactivity of char, and P_{CO}, P_{CO_2} = carbon monoxide and carbon dioxide partial pressures. Based on this equation, it is seen that reaction rates increase with P_{CO2} only at relatively lower pressure levels and then to plateau at pressures less than approximately 1 MPa. Carbon monoxide is the principal product of the reaction and its concentration may have inhibiting effect on the rate.

2.4.3 Coal Char–Steam Reaction

The reaction is very similar to char-carbon dioxide reaction in many respects. The reaction rate is faster than coal char-carbon dioxide reaction. The rate of this reaction also depends mainly on temperature and pressure. Specific rates in steam of different coal chars increase with temperature. Arrhenius activation energies of char-steam reaction for different chars have been found to vary from 100 to 200 KJ mol^{-1}. The rates increase with steam pressure but become independent at pressures higher than 1 MPa. In general, a similar Langmuir type equation is used to represent gasification rate for this reaction. Therefore carbon monoxide and hydrogen as reaction products may have retarding effect on the reaction rate.

2.4.4 Some Factors Related to Char's Reactivity

The reactivity of char produced in the pyrolysis step depends on nature of parent coal. The reactivity of char increases with oxygen content of parent coal but decreases with carbon content. In general, coal chars produced from low rank coals are more reactive than chars from high rank coals. The reactivity of char from low rank coal may be influenced by catalytic effect of mineral matter in char. The reactivity of char decreases as the carbon content of parent coal increases. As the carbon content of coal increases, the reactive functional groups present in coal decrease and the coal substance becomes more aromatic and cross-linked in nature. Therefore chars obtained from high rank coals contain less amount functional groups and larger amount of aromatic and cross-linked structures, which reduce reactivity.

The reactivity of char also depends upon thermal treatment it receives during its formation from the parent coal. It is found that specific gasification rate of char decreases as the char preparation temperature increases. That is due to the decrease in active surface areas of char. Therefore a change of char preparation temperature may change the chemical nature of char, which in turn may change the gasification rate.

Chars have higher surface areas when compared with parent coals. The surface area changes as char undergoes gasification. The surface area increases with carbon conversion, reaches maximum and then decreases. These changes in turn affect gasification rates. In general, reactivity increases with the increase in surface area. The initial increase in surface area appears to be caused by cleanup and widening of pores. The decrease in surface area at high carbon conversion may be due to coalescence of pores, which ultimately leads to collapse of the pore structure.

The reactivity of char may be influenced by catalytic effect of mineral matter in char. It is found that the reactivities of lignite chars obtained from lignites initially treated in acid were in general much lower than the corresponding reactivities exhibited by lignite chars derived from untreated lignite. But this phenomenon was not observed with several bituminous and subbituminous coal chars. This behavior apparently resulted from a catalytic effect of sodium or calcium combined with carboxyl functional groups in the organic structure of lignite. Since the concentration of carboxyl functional groups decreases significantly with increasing coal rank, this catalytic effect would predominate in lignite and would decrease rapidly with increasing coal rank.

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Biographical Sketch

Xingzhong Sha was a graduate student of ECUST, Shanghai, China. Majoring in Chemical Technology he was the visiting scholar at Tohoku University, Japan. He has published about 90 papers in Chinese and English. His book *Coal Gasification and its Utilization* was published in Chinese in 1995.