Research Status and Analysis of Biomass Pyrolysis Gasification Technology

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Abstract: This paper analyzes the main methods and principles of biomass thermochemical conversion worldwide, introduces common equipment for biomass pyrolysis and gasification, and discusses the current research and numerical simulation of biomass pyrolysis gasification experiments. This paper also analyzes the application status of biomass pyrolysis gasification technology and summarizes the bottleneck problems encountered so as to preliminarily discuss the ways and methods of using biomass pyrolysis gasification to solve tar problems in the future.

1. Introduction

A useful measure to curb global warming is to use renewable energy to replace fossil fuels. Biomass is a unique renewable energy source as it can be stored and converted into fuel or chemical raw materials. In the use of biomass energy, biomass gasification technology can convert solid biomass into gaseous fuel used for power generation, gas supply, and central heating through thermochemical reaction.

From a biological point of view, the main components of biomass are cellulose, hemicellulose, lignin, and a small amount of extracts and ash, as shown in Table 1. Woody plants with a dense fibrous structure and high lignin content grow relatively slowly, while herbaceous plants with loose fibers, low lignin content and high cellulose content are generally annual plants. Different biomass materials differ in composition, and the relative ratio of cellulose to lignin is a factor in the determination of biomass type and the way to handle biomass. The pyrolysis products of cellulose include non-condensable gas, coke and a large amount of aldehydes, ketones and organic acids. In comparison, hemicellulose produces less tar but more non-condensable gas during pyrolysis. Wood produces much more coke than cellulose and hemicellulose during pyrolysis. The pyrolysis process of biomass can be seen as a linear superposition of the pyrolysis process of cellulose, hemicellulose, and lignin [1].

| Type     | Cellulose | Hemicellulose | Lignin | Extract | Ash |
|----------|-----------|---------------|--------|---------|-----|
| Cork     | 41        | 24            | 28     | 2       | 0.4 |
| Hardwood | 39        | 35            | 20     | 3       | 0.3 |
| Pine bark| 34        | 16            | 34     | 14      | 2   |
The biomass is converted into biomass energy through two main pathways, namely, thermochemical conversion and biochemical conversion. Generally speaking, thermochemical conversion is more efficient in terms of reaction time and is stronger in converting organic matter. For example, lignin is a typical non-fermenting material; therefore it cannot be thermochemically but biochemically degraded. The thermochemical conversion mainly includes the steps of direct combustion, pyrolysis, gasification and liquidation [1], as shown in Figure 1.

![Thermo-chemical processes for bioenergy production and the corresponding products](image)

Fig.1 Thermo-chemical processes for bioenergy production and the corresponding products

Combustion is the most widely used in biomass conversion. Biomass combustion equipment mainly includes fixed bed, moving bed, fluidized bed and rotary kiln [2]. Pyrolysis refers to the thermal decomposition process of converting biomass into solid carbon, liquid (biomass oil and tar) and non-condensable gases under high temperature anoxic or anaerobic conditions. Nowadays, pyrolysis research is becoming increasingly important, which is because pyrolysis technique is considered to be an industrially achievable biomass conversion process and pyrolysis is the most important first step in gasification and combustion [3]. Biomass gasification is a process in which low oxygen or a suitable oxidant (such as water vapor and CO₂) converts carbon into a kind of combustible gas. When air or oxygen is used as the gasification medium, the process of gasification is similar to that of combustion. However, gasification is the process of local biomass combustion, which mainly uses fixed bed/moving bed, fluidized bed, circulating fluidized bed, etc. [4].

Direct liquidation is a thermochemical process carried out at high pressure and low temperature conditions, in which the biomass is cracked into small fragments and dissolved in water or other suitable solvent. Lignocellulosic materials are rich in hydroxyl groups, thus able to be liquefied into intermediate products for biopolymer production, such as epoxy resin, polyurethane foam, plywood adhesives, etc. [5].

2. Research status of biomass pyrolysis gasification
The downdraft gasifier is the most widely used in current fixed bed gasification systems. His et al. [6] studied the biomass gasifier using the downdraft fixed bed gasifier with air as the gasification agent, aiming to explore the effect of operating parameters on biomass gas production. But the experiment did
not mention the production rate of tar. Garcì'a-Bacaicoa et al. [7] studied the gasification characteristics of biomass and high-density polyethylene in a downdraft gasifier, aiming to investigate the feasibility of gasification using the mix of the two substances. In 2009, Sharma [8] studied 75 kW downdraft biomass gasifier system, and investigated the effect of pressure decrease on the combustion of the latter turbine. The whole system adopted water washing and sand bed to purify biomass gas, but failed to give the content of tar in the biomass gas. Ratnadhiroya et al. [9] studied the molar distribution of CO/CO₂ and CO/H₂ along the length of the downdraft gasifier, suggesting that the molar distribution equations of CO/CO₂ and CO/H₂ can be used to develop the actual gasification status in the downdraft gasifier. Compared with the fixed bed gasifier, the biggest advantage of the fluidized bed gasifier is that the uniform temperature distribution in the window and larger particle size. Dual fluidized bed gasifier is the current research focus. This kind of gasifier was pioneered by Professor Kunii [10], followed by Bartelle-Columbus and FERCO in the United States, TNEE in France, and AVSA in Belgium. The current research is concentrated in Europe (Austria and the Netherlands), Japan, and China [11]. Studies suggested that high tar content in biomass gas remains the main problem for this gasifier. Capucine Dupont et al. [12] completed the experimental study of biomass pyrolysis gas on a fluidized bed reactor with a temperature range of 800–1000 ℃ and particle diameters of 0.4 mm and 1.1 mm. The experimental results is closely related to the size of the biomass particles. Biomass pretreatment and feedstock are the biggest difficulties facing the application of fluidized bed. Recently, the University of Western Kentucky in the US developed a new biomass gasification gas production technology, which can reduce tar content and improve carbon conversion rate and gasification efficiency. The US National Renewable Energy Laboratory conducted studies on high-pressure combined gasification using coal biomass fluidized bed, and partial gas recirculation combustion was used to supplement energy and reduce tar content [13, 14].

Domestic research on biomass gas production has a late start but a rapid development. Wei [15] selected 13 kinds of biomass of agricultural and forestry wastes and plants such as straw, rice husk, corn cob, sawdust, the branches and leaves of salix mongolica and salix matsudana, alfalfa, reed, and alkali grass to make the biomass mixture and uses thermal gravimetric analyzer to study the co-pyrolysis characteristics of the biomass mixture and a typical lignite and the temperature of the lignite precipitation. Mi et al. [16] used a thermogravimetric analyzer to study the reactivity of biomass semi-coke in a CO₂ atmosphere. The reactivity experiment of peanut shell coke under different partial pressures of CO₂ shows that the reactivity of the coke sample is positively related to the concentration of the reaction gas. Cao et al. [17] conducted a thermal experimental study of a high-temperature air generator and obtained the its performance parameters, thereby preparing for the further study of high-temperature air gasification system. Wei et al. [18] studied biomass gasification by using a fixed bed updraft gasifier to explore the effects of operating conditions such as furnace temperature, raw material characteristics and fuel layer thickness. Zhou et al. [19] studied the effects of rice husk, Korean pine and fraxinus mandshurica ongas production under different parameters of oxygen, temperature, biomass ratio. The result shows that the gasification temperature is the main influencing factor for biomass entrained-flow bed. Zhao et al. [20] conducted a rapid pyrolysis gasification study of biomass under high-temperature and high-energy conditions generated by thermal plasma. Hu et al. [21] conducted a comparative analysis on the difference in tar yield rate, carbon conversion rate and gasification efficiency between coupled gasification and conventional coupled gasification by adopting upper and lower reactors of the fixed bed and taking vinasse as the gasification fuel. Duan et al. [22] carried out an experimental study on the NO reduction using simulated biomass gas, thereby obtaining the reburning characteristics of biomass gas.

3. Biomass pyrolysis gasification application status
Foreign biomass gasification research mainly focuses on the fields including district heating using biomass gas, cement plant gas supply and power generation, power generation using biomass gasification, ammonia production using biomass gasification, and dimethyl ether or methanol production using biomass gasification. Compared with equipment used in China, the biomass
gasification equipment used at abroad is generally larger with higher automation degree. Its design raw material is often wood waste and is mostly used for power generation and heating. According to statistics, the ratio of commercial gasification equipment types manufactured in Europe and America is: updraft fixed bed gasifier: 2.5%; downdraft fixed bed gasifier: 75%; fluidized bed: 20%; others: 2.5% [23]. Updraft fixed bed gasification unit designed and developed by Moore Corporation of Canada can accept biomass feedstock with a higher water content (up to 60%); however, since the tar formed during pyrolysis does not pass through the combustion zone, the equipment will produce gas with a higher tar content. The production of most of the updraft gasifiers has been suspended due to its pollution (water pollution caused by tar residues). The gasification efficiency of the fluidized bed gasification unit designed and manufactured by Canada General Fuel Gasification Equipment Co., Ltd, the carbonization-gasification gas generation system manufactured by American Standard Solid Fuel Company, and the downdraft gasification gas generation system manufactured by German Army Belt Energy Co., Ltd is 60%-70%. The USA has invested much in biomass gasification research and development, such as the US Bettelle (63MW) and Hawaii (6MW) projects, which are mainly based on the biomass gasification-steam combined cycle power generation (IGCC) with high power generation efficiency. Tokyo Institute of Technology in Japan has developed a biomass high-temperature air gasification system for municipal waste treatment [24, 25]. Sweden's pressurized biomass gasification power generation project (4MW) adopts high-temperature air and water vapor as a medium for gasification study of high-density biofuels, suggesting that both gas production and low calorific value increase with increasing temperature [26]. The power generation efficiency of Italy's 12MW biomass gasification power generation BIGCC project is about 31.7%. But its construction cost reached 25,000 yuan/kwh, so the power generation cost is about 1.2 yuan/(kwh), making it difficult to be applied on a large scale [27]. In Europe, the UK (8 MW), Finland (6 MW) and the EU established three 7-12MW biomass gasification power generation BIGCC demonstration project, and France, Germany, as well as the Netherlands also carried out research on biomass gasification [28-30].

Domestic biomass gasification technology has developed rapidly since the 1980s. Guangzhou Institute of Energy, Chinese Academy of Sciences has made progress in the field of circulating fluidized bed gasification power generation, and has built and operated several gasification power generation systems [31, 32]. China Agricultural Machinery Research Institute has developed and promoted a wood drying technology using fixed-bed gasifiers [33]. Shandong Energy Research Institute successfully developed key equipment in straw gasification unit and centralized gas supply system during the Seventh Five-Year Plan and Eighth Five-Year Plan, thus helping to complete the set of technologies concerning gas generation, transmission and distribution. After the completion of the first pilot project for centralized gas supply in 1994, China has established a number of biomass gasification centralized gas supply projects nationwide [34, 35], for example, Liaoning Energy Research Institute has developed a downdraft biomass gasifier [36].

The most prominent problems restricting the commercialization of biomass pyrolysis gasification technology in China are high tar content in biomass gas and low automation degree [37]. Professor Beenackers of the Netherlands pointed out in his article commenting on European moving bed biomass gasification technology that the two problems above are common to see in the amplification design of current updraft and downdraft gasifiers, which undoubtedly hinders the promotion and application of biomass gasification process [38]. The advantages and disadvantages of various gasifiers are shown in Table 2.

| Table 2. Advantages and disadvantages of selected types of gasifiers |
|-------------------------------------------------------------|
| **Advantage** | **Disadvantage** |
| Updraft fixed bed/moving bed | Simple, low price; outlet temperature of about 250 °C; good operation under pressure; high carbon conversion efficiency; low dust content in gas; high thermal efficiency | High tar content; easy to burn empty, be overhead, and produce slag; small size of feedstock; much time and effort for packing |


### Downdraft fixed bed / moving bed
- Simple process; tar only exists in the produced gas

### Fluidized bed
- Flexible feeding speed and feedstock composition; can accept high ash fuel; can undertake pressure; high CH₄ content in gas; scale can be large or small; temperature is easy to control
- Operating temperature is limited by ash sintering; high gas temperature; high tar and particulate content in gas; high carbon content in fly ash

### Circulating fluidized bed
- Flexible process; operating temperature up to 850 °C
- Corrosion and wear; poor control of operation with biomass as fuel; tar problem still exists

#### 4. Research status of numerical simulation of biomass pyrolysis

In 1989 and 1991, Koufopanos et al. established the pyrolysis kinetic model of biomass and its components and the pyrolysis model based on the biological particles affected by kinetics, heat and heat transfer [39, 40]. The chemical reaction kinetics model mainly studies the pyrolysis kinetics of lignocellulosic materials, aiming to provide a simple kinetic model for engineering application. The particle size is below 1 mm, thus the mass and heat transfer can be neglected and the reaction is considered to be controlled by pyrolysis kinetics. The pyrolysis rate of a kind of biomass can be described by the accumulation of the corresponding pyrolysis rates of cellulose, lignin, and hemicellulose. The kinetics of each component is modeled by a kinetic flow diagram that can predict the pyrolysis rate and ultimate weight loss, with a wide range of pyrolysis parameters (including heating conditions). The main disadvantage of this model is the failure to consider the transfer phenomenon (heat transfer and mass transfer) that plays a controlling role in the pyrolysis process of large particles.

In 1995, Srivastava et al. [41] predicted the content of biomass materials during the pyrolysis process by selecting different stages of the first-stage reaction and the second-order reaction under isothermal and non-isothermal conditions, respectively. The results show that this model can be used to predict the concentration curve of various biomasses, but the main disadvantage is the failure to combine with the transfer phenomenon. In 1999, Jalan et al. [42] studied the kinetics and heat transfer effects of pyrolysis of a single biological particle in a cylindrical coordinate system. The model of the single biological particle is based on physical and chemical changes that are both controlled by heat transfer phenomena. In addition, chemical changes include primary and secondary pyrolysis reactions. The proposed energy balance model equation considers the non-isothermal reaction of biological particles.

In 2000, Janse et al. [43] established a model for the flash pyrolysis of individual wood particles. This model makes the following assumptions: the model is one-dimensional; the ideal gas criterion is applied; local heat balance exists inside the particles between the gas phase and the solid phase; the macroscopic size of particles do not change during the reaction; wood and charcoal have changeable densities; cellulose and hemicellulose include macroscopic pores, and the diameter of impervious microscopic pores is less than 100 angstroms; porosity remains constant during conversion; Darcy gas model is used to describe the process during which the steam passes through the porous matrix of wood and charcoal to the particle surface; there is no restriction of external mass transfer; water evaporation of is not considered. The simulation results show that there is no need for the detailed description of the internal mass transfer phenomenon in the flash pyrolysis model, while correct information on chemical kinetics and heat transfer parameters is essential. This model only targets at one wood species, and makes no special explanation of tar and biomass oil conversion. In 2000, Larfeldt et al. [44, 45] conducted model and measurement studies on heat transfer of charcoal pyrolyzed from large-particle wood. The result shows that: the pyrolysis rate of large-particle biomass
restricts heat transfer through the surface charcoal layer; thermal diffusion of charcoal is constant (approximately 0.7 mm²/s). The main problem of the model is that it fails to consider the effects of kinetics.

From 2003 to 2004, Babu et al. [46, 47] studied the pyrolysis model of solid particles based on chemical reaction kinetics and heat transfer effects. Later, Babu simulated the biomass pyrolysis process on a fixed bed [48]. The main disadvantage of the above models is that they are only applicable to a single wood species without taking into account the effects of porosity [49]. The simulation result shows that the current model can deliver better-matched results with the experiment than the earlier models. However, Babu’s model shows no improvement in the aspect of physical and chemical mechanism compared to Koufopanos’ model, but only applies different numerical solutions.

In 2007, Zabaniotou et al. [50] studied the model of internal particle transfer and chemical reactions based on rapid pyrolysis of olive residues. The study selects spherical particles with a radius of 175 μm, the temperature range of 573 to 873 K, and the heating rate of 200 °C/s. The model considers both chemical reaction kinetics and heat transfer, and the simulation results are consistent with the test results (temperature, conversion process, and product distribution) obtained from the wire mesh reactor. However, the model does not consider shrinkage, so it can only be applied to a single material with woody physical parameters. In 2008, Gerun et al. [50] studied the local oxidation process of CFD simulation of two-stage downdraft gasifier, aiming to investigate the tar cracking situation of each reaction zone. This model is more inclined to simulate the combustion process of tar than its secondary thermal cracking. In 2009, Papadikis et al. [51] studied the rapid pyrolysis of biomass using CFD targeting the entrained-flow bed, using the two-stage hemispherical model.

In addition, Ratnadharaya et al. [52] studied the thermodynamic equilibrium and kinetic free model of the new biomass gasifier. Sharma et al. [7] also studied the biomass pyrolysis model based on thermodynamic equilibrium. Di Blasi et al. [53-55] have worked on the simulation of biomass pyrolysis gasification since 1992, but he focused more on the biomass pyrolysis and gasification process from the aspects of heat transfer, mass transfer and flow, but less on chemical reaction kinetic models. Based on the definition of tar and biomass oil, the liquid produced by biomass pyrolysis can be divided into two categories: tar and biomass oil. This classification can simulate not only the biomass pyrolysis reaction for oil production, but the relationship between tar production and temperature during biomass pyrolysis gasification under high temperature conditions [56]. The details are shown in Figure 2.

![Chemical kinetic scheme of biomass pyrolysis](image)

Biomass pyrolysis simulation is mostly carried out under low temperature conditions. The kinetic models of chemical reaction are mostly competitive models. Biomass after pyrolysis produces non-condensable gas products, liquid products (condensable gas products) and solid products. However, the liquid products produce biomass oil (below 500 °C) and tar (above 500 °C). The composition of the two condensable gas products is greatly different, so their excitation of pyrolysis under different temperature conditions is quite different, meaning that the application of a uniform excitation may affect simulation accuracy.
5. Conclusion

(1) At present, the main problems in biomass gasification include high tar content in biomass gas, large amount of waste water pollution due to biomass gas purification, low biomass calorific value and low energy utilization efficiency. High tar content is the most prominent problem, which causes pipe blocking and equipment shutdown. The removal of tar by water washing leads to a waste of energy (about 3% of total biomass energy) and environmental pollution due to the difficulty of waste water treatment. The catalytic cracking method can pyrolyze most of the tar using a catalyst under certain temperature conditions, but it brings the trouble of replacing the deactivated catalysts such as nickel-based catalysts, novel catalysts, dolomite and CaO. More importantly, the tar is often pyrolyzed to hard tar, which is more difficult to handle, thereby causing greater difficulty to subsequent tar processing. In addition, catalytic cracking requires complicated techniques and large-scale equipment. High-pressure super-critical gasification is mainly used for hydrogen production, while the technique for syngas production is more complicated. Therefore, it is difficult to realize industrial production for dispersed biomass resources. The high-temperature thermal cracking method of tar is of engineering application prospects and is simple and easy to operate. However, the conventional biomass gasifier has low gasification temperature and short residence time, which is not conducive to the full cracking of tar.

(2) The current research on high-temperature thermal cracking of biomass tar focuses mainly on the pyrolysis using aromatic compounds such as naphthalene to simulate tar. The composition of biomass tar is extremely complicated. So far, the researchers have not been able to provide sufficient pyrolysis data under high-temperature gas conditions.

(3) Due to the complexity of the physical and chemical properties of biomass, some current models ignore the internal temperature gradients of the particles or only simulate the pyrolysis of one chemical component. In addition, most models stimulate the high-temperature pyrolysis under low heating intensity and slow temperature rising conditions, and the model of biomass pyrolysis is not good enough under high temperature conditions. From the engineering point of view, it is great significance to understand the secondary cracking of tar, temperature distribution, degree of conversion, amount of gaseous products, solid product accumulation during high-temperature particle pyrolysis for the design and operation of the reactor. However, direct measurement of the above data is quite difficult. Therefore, it is necessary to establish a model that can be used under high temperature conditions to reflect the high-temperature pyrolysis of biomass.

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