



A Review on the Thermochemical Pretreatment of Lignocellulosic Biomass for the Production of Coal-grade Solid Biofuel

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Abstract

Thermochemical pretreatment or torrefaction of biomass is rapidly gaining attention because of simple operation, efficacy, and potential to produce valuable solid fuel. Torrefaction of lignocellulosic biomass is reviewed with the purpose of understanding the present state-of-the-art and future prospects. The following aspects have been discussed: biomass selection, optimization of engineering parameters, characterization of solid, liquid, and gaseous products and finally comparison with coal so that torrefied biomass may be blended with coal and utilized in a pulverized coal-fired furnace to produce electricity.

Keywords: Biomass torrefaction; Sub-bituminous coal; Thermochemical pretreatment; Energy yield

Introduction

Political uncertainty greatly determines the fluctuation in the price of crude oil in the range of USD 30-110 per barrel. Though the reserve of crude oil is limited, adequate supply by Saudi Arabia, Russia, and USA (for shale oil) put pressure on its cost which is now hovering around 25-35 USD per barrel bracket. A high crude price, greenhouse gas (GHG) emission concerns and unpleasant output during processing of coal necessitate research on renewable energy e.g., solar, wind, biomass etc. Solar energy is not available during the night whereas wind energy is available mostly in coastal area. On the other hand, biomass is available in most part of the world and the rate of production of biomass is much faster than the rate of production of coal, crude oil, etc., which usually takes millions of years.

It is well understood that the network of liquid and gaseous transportation fuels is well established throughout the world. Hence, there is a thrust to produce similar fuels from biomass. Pyrolysis and gasification are proven technologies to produce bio-oil and syngas (a mixture of carbon monoxide and hydrogen gas) from biomass. However,

a few countries like India, China, Australia, Germany etc. are utilizing coal for the production of electricity as they have a high reserve of coal and available infrastructure for the exploitation of coal. To mitigate environmental pollution and reduce the emission of GHG, there is considerable pressure on these countries to shut or reduce pulverized coal-fired thermal power plants.

India has a very limited reserve of crude oil and natural gas but very good availability of coal. However, Indian coal has low heating value, high ash content and unacceptable sulfur content. The availability of biomass (both agricultural and forest residue) in India is appreciable. The main drawback of biomass is high moisture, high oxygen content, low bulk density and low heating value. If these demerits are improved by means of some suitable process, the modified biomass may be co-combusted with coal in thermal power plants and make the process more environment-friendly, at the same time reduce dependence on imported crude and save money of the Federal government.

Torrefaction is such a process with good potential to convert biomass to value-added solid biofuel.

The Objectives of this Review Paper are Mentioned below:

1. Define torrefaction
2. Summarize experimental and modelling studies on torrefaction and important findings
3. Study the prospects of blending torrefied biomass with coal

Torrefaction

Torrefaction, also known as mild pyrolysis, is a thermochemical pretreatment step wherein a carbonaceous material is heated in a temperature range of 200-300°C in absence of oxygen. A retention time of 30-60 minute is usually adequate; besides, heating rate is varied in the range

of 5-20°C per minute. A few other variables are particle size, sweeping gas flow rate, biomass type etc. A large sized particle offers mass and heat transfer resistance. Hence, particle size of the order of 1 mm or less is suitable for torrefaction. Published works suggest that sweeping gas flow rate does not have much influence on the yield or quality of torrefied biomass. Majority of studies employ a fixed-bed reactor for torrefaction; however, a fluidized-bed will ensure better heat transfer characteristics and consequently torrefaction product with uniform quality. Strictly speaking, the range of temperature is decided from the thermogravimetric analysis (TGA) of the specific biomass under inert atmosphere.

The following (Table 1) summarises experimental and modelling studies on torrefaction carried out by different researchers on different biomass species.

S. No.	Biomass	Operating Conditions	Important Findings	Reference
1	Sawdust briquette	T: 250-270°C; retention time: 2-4 hr; a mild steel chamber was used for torrefaction with circulation of hot gas from exchanger	Faster rate and less smoking during combustion of torrefied wood; Energy yield: 76.5-89.6%; H/C and O/C decreased	[1]
2	Briquette from wood residue	T: 220-270°C; retention time: 0.5-1.5 hr; inert medium	Temperature has more influence than retention time; Energy yield ~50-97%; HHV increased by 15%; 73% decrease in equilibrium moisture content	[2]
3	Briquette from wood residue	Economic analysis of incorporating torrefaction within wood briquetting factory	Improvement in the quality of briquettes; Break-even point becomes lower; Multi-product manufacturing facility possible	[3]
4	Beech and willow	T: 200-300°C; retention time: 10-30 min; gasification of wood and torrefied wood was considered in different reactor configurations	Thermodynamic losses during gasification may be reduced by using torrefaction pretreatment; H/C and O/C decreased but HHV increased	[4]
5	Willow and straw	T: 225-300°C; particle size: 0.7-2.0 mm for hardwood, <5 mm for straw	Two-step reaction model proposed; activation energy 76 and 151.7 kJ/mol; preexponential factor 2.48×10^4 and 1.10×10^{10} kg/kg s Majority of hemicellulose degraded	[5]
6	Beech, willow, straw, and larch	T: 220-300°C; atmospheric pressure; retention time: 10-60 min; quartz fixed-bed reactor	Xylan-containing beech, willow and straw are more reactive than larch; Acetic acid and methanol are formed	[6]
7	Eucalyptus	T: 240-280°C; inert atmosphere (N_2); retention time: 0-3 hr; <5 mm particle size; heating rate: 10°C/min	Grindability improved; TGA was carried out; kinetic parameters were estimated; torrefaction influenced decomposition of hemicellulose	[7]

8	Reed canary grass, short rotation willow coppice, wheat straw	T: 503-563 K; inert atmosphere; retention time: 10-50 min; heating rate: 20 K/min	Torrefied biomass resembles properties of low rank coal; DTA was carried out on combustion of raw and torrefied biomass	[8]
9	Reviews torrefaction of different biomass	Efficiency of torrefaction, pelletization and pyrolysis discussed	Techno-economic evaluation of torrefaction, fast pyrolysis and pelletization	[9]
10	Leucaena, woody biomass	T: 200-250°C; inert atmosphere; retention time: 30 min; <75 micron particle size; P: up to 4 MPa	Solid yield increased with pressure; HHV increased significantly; H/C and O/C decreased	[10]
11	Norway spruce wood chips	T: 533-583 K; inert (N ₂) and air atmosphere; retention time: 8-25 min; heating rate: 5-20 K/min; 4-12.5 kg/h mass flow rate of wood chips	Significant decomposition of hemicellulose; Appreciable increase in HHV	[11]
12	Scots pine pellets	T: 230-270°C; inert atmosphere; retention time: 1 hr;	HHV increased from 18.37 to 24.34 MJ/kg; Grinding energy requirements decreased for torrefied pellets; More particles were in the smaller size range	[12]
13	Leucaena	T: 240-320°C; inert atmosphere (N ₂); retention time: 0-1140 min; heating rate: 10°C/min; nitrogen flow rate 150 mL/min	HHV, ultimate analyses were compared at similar yields; Torrefaction at 320°C altered cellulose structure	[13]
14	Corn cob	T: 175-195°C; wet and dry torrefaction carried out; inert atmosphere (N ₂); high-pressure batch reactor; corncob:water equals 1:9 w/w in WT; 600 RPM; heating rate: 10°C/min;	Most of hemicellulose got removed; Dry torrefaction degraded cellulose as well; Wet torrefaction improved yield of levoglucosan	[14]
15	Eucalyptus globulus and Eucalyptus nitens	TGA carried out: 20-600°C; heating rate: 10°C/min; nitrogen atmosphere, 70 mL/min	Activation energy of hemicellulose decomposition was ~121-170 kJ/mol; 295-310°C appeared to be the most feasible temperature for torrefaction	[15]
16	Rice husk	T: 210-270°C; inert atmosphere (N ₂), 200 mL/min; retention time: 1 hr; heating rate: 10°C/min;	Torrefaction and organic acid-leaching pretreatment followed by pyrolysis was carried out	[16]
17	Rice straw	T: 200-300°C; inert atmosphere (N ₂); retention time: 10-70 min;	Torrefaction improved HHV, but decreased solid and energy yields; Temperature had more pronounced influence than retention time	[17]
18	Acacia nilotica	T: 220-280°C; inert atmosphere (N ₂), 40 mL/min; retention time: 20-60 min; heating rate: 5-15°C/min; fixed-bed reactor	Max. HHV and energy yield obtained at 252°C, 60 min retention time and 5°C/min heating rate; H/C and O/C both decreased; HHV increased by 18.62% Fuel and flow properties improved considerably	[18]

19	Pigeon pea stalk (cajanus cajan)	T: 200-300°C; inert atmosphere (N ₂), 40 mL/min; retention time: 0-60 min; heating rate: 5-20°C/min; cylindrical-shaped quartz reactor	Temperature had highest impact on torrefaction; Optimum condition for maximum HHV and energy yield was 248°C, 60 min, 16°C/min; HHV became 21.15 MJ/kg; Crystallinity index decreased	[19]
20	Acacia nilotica	T: 220-280°C; inert atmosphere (N ₂), 40 mL/min; retention time: 20-60 min; heating rate: 15°C/min; fixed-bed reactor; particle size: 0.7-1.15 mm	HHV increased from 19.31 to 24.76 MJ/kg; fixed carbon improved from 11.35 to 60.4 wt.%, whereas the fuel ratio improved from 0.13 to 1.63; torrefied biomass was rich in sodium, potassium, calcium etc.; Moisture absorbed by torrefied biomass was only 6.61% as compared to 35.44% by raw biomass	[20]
21	Pigeon pea stalk (cajanus cajan)	T: 220-280°C; inert atmosphere (N ₂), 40 mL/min; retention time: 20-60 min; heating rate: 15°C/min; cylindrical-shaped quartz reactor	Maximum HHV and energy yield were obtained at 280°C and 60 min retention time; HHV increased by 37.1%; Flowability improved after torrefaction; Combustibility index increased by 76.2%	[21]
22	Pigeon pea stalk (cajanus cajan)	T: 225-275°C; inert atmosphere (N ₂), 40 mL/min; retention time: 15-45 min; heating rate: 15°C/min; cylindrical-shaped quartz reactor	HHV increased by 28.6% at 275°C and 45 min retention time; Energy density also improved with severity of torrefaction; Torrefied biomass exhibited better combustion properties	[22]

Table 1: A summary of torrefaction studies in the past few years.

Suitability of Biomass as a Feedstock for Torrefaction

Biomass consists of hemicellulose, cellulose, lignin, and extractives. Thermogravimetric analysis (TGA) of biomass dictates that within a temperature range of 200-300°C, majority of hemicellulose gets degraded, thereby making biomass more brittle. Hence, this temperature range is usually considered very much suitable for torrefaction. There is rupture of C-O, O-O, O-H bonds etc., hence formation of CO₂, CO, and moisture. A van Krevelen diagram suggests reduction of both H/C and O/C atomic ratio due to torrefaction, which brings it closer to sub-bituminous coal. Waste biomass, after torrefaction, presents a pathway for waste minimization, waste-to-wealth creation or waste valorization. Hence, biomass appears to be a very much suitable candidate for torrefaction. It may be added that biomass with moisture less than 10 wt% and moderate ash content is very much suitable for upgrading as solid fuel via torrefaction.

Optimization of Process Variables

The outcome of any process depends on a set of independent variables. Similarly, torrefaction also depends

on independent variables like temperature, heating rate, retention time, particle size, sweeping gas flow rate, biomass type etc. in general, energy yield is a very important parameter which needs to be maximized during torrefaction.

Solid yield = (amount of torrefied biomass/amount of raw biomass)*100%

Energy yield = (solid yield)*(HHV of torrefied biomass/HHV of raw biomass)

HHV: higher heating value (MJ/kg)

Singh, et al. [18] optimized torrefaction of *Acacia nilotica* in a fixed-bed reactor using response surface methodology. Maximum higher heating value and energy yield were obtained at 252°C, 60 min retention time and 5°C/min heating rate. H/C and O/C atomic ratios in torrefied biomass decreased by 75.54 and 18.62%, respectively, as compared to raw biomass. Singh, et al. [21] carried out optimization of torrefaction for eucalyptus biomass in a tubular quartz reactor. Maximum HHV was obtained at 280°C and 60 min retention time. Carr Compressibility Index (CCI) and Hausner Ratio (HR) decreased indicating better flow behaviour. The fuel ratio increased significantly, thereby indicating higher

fixed carbon and hence better fuel properties. The overall activation energy decreased from 179.1 to 81.7 kJ/mol, due to torrefaction, indicating lower thermal energy requirement for conversion of torrefied biomass. FT-IR analysis suggested reduction of peak intensity of O-H bonds, thus indicating decomposition of hemicellulose.

Characterization of Products

Torrefied biomass is the main product in torrefaction. It should be characterized for bulk density, HHV, proximate and ultimate analysis, and various fuel and flow properties. Singh, et al. [22] studied torrefaction of pigeon pea stalk, an agricultural residue widely available in India and other Asian countries, in a fixed-bed reactor. They have reported that energy density and HHV increased upon torrefaction. The HHV of torrefied biomass increased by 28.6% when reaction was carried out at 275°C and 45 min retention time. Moisture reabsorption decreased significantly indicating that torrefied biomass may be stored for a long time yet there would be no or minimum degradation. The activation energies of hemicellulose and cellulose decreased to a good extent. Singh, et al. [20] performed torrefaction of *Acacia nilotica* in a tubular quartz reactor. The HHV and fixed carbon increased from 19.31 to 24.76 MJ/kg and 11.35 to 60.4 wt%, respectively, when raw biomass was torrefied at 280°C for 40 min retention time. ICP-MS analysis revealed that the torrefied biomass was rich in sodium, potassium, calcium, magnesium etc. Moisture absorbed by torrefied biomass was 6.61% only, as opposed to 35.44% for raw biomass. Besides, the van Krevelen diagram also clearly suggests that O/C and H/C ratios of torrefied biomass approach very close to sub-bituminous coal.

The liquid product from torrefaction does not have much heating value hence cannot be utilized as a transportation fuel. However, several value-added chemicals like acetic acid, furfural, etc. may be extracted from it. The gaseous product consists of CO₂, CO, small amount of H₂ and lighter hydrocarbons. This gas stream may be recycled and combusted for thermal energy which, in turn, may be utilized to preheat biomass before it is sent to the torrefaction reactor.

Prospects of Blending Torrefied Biomass with Coal

Fuel and flow properties of torrefied biomass, as discussed earlier, suggest that it may be blended with sub-bituminous coal and utilized in a pulverized coal-fired thermal power plant, the blended fuel will have less ash and sulfur but acceptable flow properties as well as heating value. Not many published works are available on the commercial application of torrefied biomass in power generation applications. Research in this direction is therefore required.

Summary and Future Prospects

Torrefaction appears to be an excellent pretreatment step to convert low-value biomass into value-added solid biofuel. Torrefied biomass has high fixed-carbon and calorific value whereas very low ash and sulfur content. It also exhibits improved flow and fuel properties. These features make it an ideal candidate for blending with coal. Hence, before we can phase out coal, it may be blended with torrefied biomass for producing electricity in thermal plants. It may also be predicted that torrefied biomass alone will be capable of producing electricity via combustion in power plants. It has been observed that there is a need to carry out research on detailed characterization of liquid product obtained from torrefaction and separately catalytic torrefaction.

Nomenclature

TGA	Thermogravimetric analysis
HHV	Higher heating value (MJ/kg)
FTIR	Fourier-Transform Infrared
GC-MS	Gas chromatography-mass spectrometry
ICP-MS	Inductively coupled plasma-mass spectrometry
EY	Energy yield
SY	Solid yield
H/C	Hydrogen/carbon atomic ratio
O/C	oxygen/carbon atomic ratio
CCI	Carr compressibility index
HR	Hausner ratio

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