
Bio-oil production from agricultural crop residues – a review

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Abstract: India generates approximately 500 million tons of agricultural crop residues per year. This crop residue (waste) can be converted into fuel by using thermo-chemical and bio-chemical conversion routes. Thermo-chemical conversion routes mainly include gasification, pyrolysis, and liquefaction. In recent years, the pyrolysis technology for the production of bio-oil is gaining attention globally, due to its simple and efficient conversion techniques. This paper provides the updated review of bio-oil production mainly from agricultural crop residues and focuses on the different bio-oil production techniques such as fast pyrolysis and hydrothermal liquefaction; the reactor used for bio-oil production, properties, and application of bio-oil. The bio-oil produced from agricultural residues has a heating value of between 17–21 MJ/kg, exactly half of the diesel fuel. The properties of bio-oil showed the potential ability to be used as fuel in many applications such as boiler, furnaces, and diesel engines.

Keywords: bio-oil; crop residues; fluidised bed reactor; fast pyrolysis; hydrothermal liquefaction; HTL.

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1 Introduction

The consumption of crude oil is increasing at a very alarming rate and it leads to a problem like the oil crisis as well as greenhouse gas emission and climate change. India ranks third largest consumer of crude oil. Renewable energy sources receive remarkable attention worldwide to replace these conventional fuels (Panwar et al., 2010). The biomass is one of the promising sources of renewable energy and has been used as fuel in applications such as cooking and lighting (Panwar, 2010; Paul et al., 2016). The global annual potential of biomass is predicted as 1.08×10^{11} toe (tons of oil equivalent), which is ten times the requirement of present world energy (Kan et al., 2016). The biomass as fuel includes wood, agricultural waste, forest residues, municipal solid waste, and animal waste, among these sources agricultural waste possesses a higher energy value because it is comprised of an organic constituent such as cellulose, hemicellulose, and lignin (Tsai et al., 2006). The wide range of crops is cultivated across the globe, with an enormous quantity of crop residues that remain unutilised on the field. Annually, 140 billion MT of biomass is generated globally from agriculture (Wankhade and Bhattacharya, 2017) and approximately 500–550 million tonnes (Mt) of biomass (crop residue) is generated annually in India from agriculture waste (Devi et al., 2017). Generally, only less amount of crop residue has been used by farmers as feed for animals and the remaining is being burned into the open field to clear the field for the growing of next crops. Whereas, it is observed that the crop residue such as wheat, rice, corn, barely, cotton, sugarcane, soybeans, rapeseed, and sunflower have the significant potential to produce biofuel (Alavijeh and Yaghmaei, 2016). In addition, Hiloidhari et al. (2014) estimated the future energy from surplus crop residue biomass is 4.15 EJ and which is equal to 17% of India's total energy consumption. It is observed that the burning of crop residue not only pollutes the environment but it is a loss of vital energy resources.

From the last few years, the thermochemical conversion of biomass (crop residue) into fuel is becoming an interesting topic and it has received great attention because it is the simplest way to convert biomass into liquid fuel. Pyrolysis is a thermochemical process in which the cellulose, hemicellulose, and lignin of biomass are decomposed into solid, liquid, and gas products when heated at temperature about 450 to 600°C (Della Rocca et al., 1999; Yaman, 2004). Depending upon the residence time and temperature, pyrolysis is further categorised mainly into two type's slow and fast or flash pyrolysis. The slow pyrolysis is characterised by lower heating rates resulting in a carbon rich solid residue called as charcoal, formed by heating the biomass at a temperature

around 300°C to 400°C for period of few minutes to few hour or day (Demirbaş, 2001; Demirbaş et al., 2016). On the other hand, the fast pyrolysis (FP) is characterised by higher heating rates to yield a liquid, formed by the heating of biomass at a temperature of about 500°C but only for a short period, one second or few seconds. Apart from this two pyrolysis, there is flash pyrolysis process. It requires a higher temperature rate and very low residence time as compared to FP. The flash pyrolysis involves a higher heating rate (more than 1,000°C/s) than the FP (10–200°C/s) (Sowmya Dhanalakshmi and Madhu, 2019). Although flash pyrolysis gives higher bio-oil yield, it is found uneconomical due to the requirement of high temperature and higher heating rate. The produced bio-oil has some properties equivalent to diesel fuel, as well as it contains very less sulphur, hence the oil produced from crop residue can be used as an alternative to conventional fuels (Bridgwater, 2012; Mura et al., 2013). The surplus advantage of bio-oil is higher volumetric heat content, easy to transport, and can be used as an efficient energy carrier (Czernik and Bridgwater, 2004). Different modes of pyrolysis and its corresponding yield are presented in Table 1.

It is crystal clear that India has an ample amount of crop residue availability and these crop residues are either leftover the field or being burned to clear the field. However, the research community is thoroughly searching the best conversion route of such organic waste into green fuel. The bio-oil production from different thermo-chemical methods is becoming a promising route. The propagation of such technology is essential to maintain the social, economic, and environmental sustainability. Therefore this article aimed to review the state of the art of bio-oil production methods such as FP and hydrothermal liquefaction (HTL), mainly from crop residues. The fuel properties and different applications of bio-oil are also covered in this article.

Table 1 Typical modes of pyrolysis and its corresponding yield

Types of pyrolysis	Residence time	Temperature (°C)	Heating rate (°C/s)	Yield (%)			References
				Liquid	Char	Gas	
Slow	Hours/day	300–400	0.1–1	30	35	35	Bridgwater (2006, 2012) and Brennan and Owende (2010)
Fast	1 to 2 s	500–600	200–500	75	12	13	Bridgwater (2006, 2012) and Brennan and Owende (2010)
Flash	Less than 1 s	500–600	More than 1,000	70–75	12–13	13–12	No (2014) and Onay and Kockar (2003)

2 Bio-oil production from crop residues

This paper comprises of different bio-oil production techniques. Currently, there are two methods used mostly for the conversion of crop residue into bio-oil, FP and HTL (Ross et al., 2010; Gollakota et al., 2018).

2.1 FP process

Now it's been three decades that FP is being reviewed for the production of bio-oil from biomass. In recent years, this technology has gained wide attention and found to be an

economical way to produce bio-oil from the biomass like agricultural wastes, loose biomass and solid waste (Bridgwater, 2012; Mohan et al., 2006; Mura et al., 2013). The FP process required conditions such as rapid heating of biomass in the temperature range of about 450–550°C, dry feedstock (less than 10%), residence time of few seconds and rapid quenching of formed vapour to yield about 75–80% of liquid (dry basis of feed) along with some char and gases. The produced liquid is called as bio-oil or pyrolytic oil and it is dark brown in colour and appears to be same elemental composition of biomass (Bridgwater and Peacocke, 2000; Bridgwater, 2012; Islam et al., 2008; Özbay et al., 2008). The FP process conditions such as temperature, heating rate, feed size, feed rate and sweep gas flow rate plays a profound effect on the quality and quantity of bio-oil yield. The temperature and heating rate are the most important parameters for the conversion of biomass into liquid fuel. Generally, the suitable temperature for bio-oil production reported in between 450 to 550°C (Ali et al., 2015, 2016; Mullen et al., 2010; Pattiya and Suttibak, 2012).

The high heating rate in FP is the mandatory for rapid thermal decomposition of biomass to yield maximum volatiles. Generally, for FP the heating rate was reported best in between 10–200°C/s (Barik, 2019). Similarly, Demiral et al. (2012) reported that 40°C/min was found optimum for the effective bio-oil production. However, Wang et al. (2017) reported that as the temperature increases from 400 to 500°C the bio-oil yield increases and reached to maximum 55.09 at 500°C under inert atmosphere. However, the bio-oil yield starts to decrease as the temperature increased above 600°C. It is due to the fact that the most volatiles particles are activated at 500°C temperature. Moreover, it is also concluded that the yield of char decreases and gas increases as the pyrolysis temperature increases from 400 to 600°C. It is evident that the yield of bio-oil is highly dependent on the pyrolysis condition such as temperature. However, our previous study revealed that the maximum yield of bio-oil 42 wt% from wheat straw was achieved at a temperature of 500°C, biomass particle size of 1 mm and N₂ flow rate of 4.0 m³/h (Paul et al., 2020). Generally, it is observed that the yield of bio-oil obtained from crop residues varies from 30 to 70 wt%. The cotton and maize straw at pyrolysis temperature of 490°C delivered 36 wt% and 42 wt% of bio-oil respectively (Ali et al., 2015, 2016).

The particle size of the feedstock affects the heat transfer efficiency of pyrolysis process. The large size particles requires high amount of heat energy, on the other hand, small particles size feedstock gives rapid and uniform heating. It is also observed that large size feedstock gives lower oil yield. The particle size from 250 µm to 1.0 mm was found optimum for maximum bio-oil production (Ali et al., 2015; Pattiya and Suttibak, 2012). Whereas, the carrier gas (sweep gas) is used to carry the clean gas swiftly from reactor into quencher. The carrier gas prevents secondary cracking of vapours, polymerisation and condensation of vapour in reactors. The carrier gasses which can be used in FP are N₂, CH₄, H₂, CO and CO₂ as well as produced gas from FP can also be used, generally inert gas is used mostly (Zhang et al., 2011). Among the inert gas, N₂ gas as carrier gas was used in many studies (Atsonios et al., 2015; Chen et al., 2010; Zheng, 2007; Luo et al., 2004). Furthermore, Wang et al. (2018) studied the effect of H₂ and N₂ as a sweep gas on the FP process of lignocellulosic biomass in a fixed bed reactor. It was observed that the H₂ was consumed by the vapours, during the pyrolysis process. Hence, the yield of non-aqueous oil and non-condensable gases were found improved in the presence of H₂ over N₂. However, the crop residues for example wheat straw and rice straw has high ash content. The high ash content and mainly potassium content from the ash, lowers the bio-oil yield and increase char and gas yield (Isahak et al., 2012;

Luo et al., 2004). It is observed that the presence of high ash promotes the char formation during the pyrolysis process (Jendoubi et al., 2011).

Many types of reactor were developed for FP process till now. The reactor is the heart of FP because rapid heating of biomass is taking place in this chamber. Bridgwater (2012) and Isahak et al. (2012) studied the following types of reactor, bubbling fluidised bed, circulating fluidised bed, rotating cone pyrolyser, vacuum pyrolysis, auger reactor, ablative reactor and ablative pyrolysis.

The details of each type of reactor including its pros and cons were studied and reviewed by Isahak et al. (2012). Whereas, Fadhilah et al. (2020) and Luo et al. (2004) found that bubbling fluidised bed reactor is the most effective reactor for the FP process. This reactor possess advantages such as high heat transfer rate (high efficiency), rate of devolatilisation, easy to operate and trouble-free char collection. The catalytic FP is technique adopted to produce high quality bio-oil from different biomass (Pattiya et al., 2010). The qualitative problem of crude bio-oil such as high oxygen content, low heating value and instability were removed by catalytic FP (Chen et al., 2019). The catalyst can be employed with biomass or supplied externally through secondary reactor for the FP process. The use of zeolite as catalyst found very promising for the FP of crop residues because it produces bio-oil with less percentage of acid and desirable phenolic compounds (Akhtar and Amin, 2012; Zhang et al., 2014). However, Chen et al. (2017) studied the catalytic pyrolysis of biomass, mainly to increase furfural compound percentage in bio-oil. The furfural is an aldehyde, and used as solvent for refining lubricating oils, as well as fungicide and weed killer. Furthermore, the authors also suggested that the agricultural biomass such as wheat straw, corn cob and cotton stalk has the great potential to produce furfural compound using heterogeneous catalyst.

However, the recent studies on FP of agricultural crop residues such as wheat straw (Paul et al., 2020), sugarcane bagasse (Treedet and Suntivarakorn, 2018), cotton stalk (Ali et al., 2015), rice husk (Cai and Liu, 2016), substantiate that the crop residues has the noble potential to produce a green fuel, i.e., bio-oil. The comparison of past research work of FP of agricultural residue is presented in Table 3.

2.2 *Hydrothermal liquefaction*

HTL is a new and at an early development stage process and found to be great potential (Gollakota et al., 2018; Xiu and Shahbazi, 2012). HTL the name itself indicates that processes which involves thermal (temperature) and hydro (water) condition with elevated pressure. The HTL process involves the degradation of the biomass at temperature between 200°C to 400°C and pressure between 5 to 25 MPa with minimum residence time of 0.2 to 1 hour (Biswas et al., 2019). The HTL gives main product as bio-oil and solid residue, bio-char and aqueous organic compound as by-products. The bio-oil produced from HTL has lower oxygen content and higher heating value than the produced bio-oil from FP (Peterson et al., 2008; Savage et al., 2010). The HTL process mainly carried out with the help of solvent such as water. The water under subcritical condition has the property such as low dielectric current and high ion content, which help to increases the catalytic reaction of acid base catalyst during the liquefaction process (Kruse and Dinjus, 2007). The water as solvent has major impact on the viscosity and oxygen content of produced bio-oil. Therefore, the organic solvent such as ethanol, methanol, acetone, 2-propanol, methanol, toluene and glycerol were used to improve the quality and quantity of bio-oil (Cao et al., 2016; Demirbaş, 2008; Huang et al., 2011;

Xu and Etcheverry, 2008; Yang et al., 2009; Yuan et al., 2011). The important parameters for HTL of crop residues are temperature, residence time, and pressure and catalyst effect. Zhang et al. (2018) investigated the HTL of tomato waste in autoclave reactor, the study shows that the effect of water as only solvent, addition of H₂SO₄, and mixture of water and ethanol as solvent on the yield of bio-oil. Authors concluded that yield of bio-oil increased from 30–40% by the addition of H₂SO₄ catalyst and the yield of 45 wt% was obtained from the mixture of water and ethanol (17/3, v/v) used as solvent at the temperature of 250 and pressure of 6.5 MPa. The residence time was 30 min and it was also found that the residence time has not a significant effect on the yield of bio-oil.

Zhu et al. (2015a) investigated HTL of barely straw. The maximum bio-oil yield of 34.9% was obtained at temperature range between 280–300°C. The increase in temperature up to 300°C gives maximum bio-oil, as the temperature starts to increase above 300°C to 400°C, the gas and solid residue yield increased, and however, the same effect of temperature has observed by Cao et al. (2016). The study on the HTL of rice straw revealed that the maximum bio-oil yield of 50.31 wt% was achieved at temperature of 260°C with 1 h retention time when glycerol used as solvent (Cao et al., 2016). However, Tian et al. (2020) compared the yield of bio-oil produced from HTL of different feedstock such as peanut straw, soybean straw, maize straw and rice straw. The maximum yield of bio-oil 16.0 wt% was obtained from soybean straw at a temperature of 320°C. The study revealed that biomass composition like cellulose and hemicellulose majorly affect the yield of bio-oil.

Table 2 Comparison of FP and HTL

<i>Conversion methods</i>	<i>Experimental conditions</i>	<i>Process descriptions</i>	<i>Advantages</i>	<i>Disadvantages</i>
Fast pyrolysis	Temperature required between 450–500°C Vapour residence time required between 1 to 2 s, and atmospheric pressure	Decomposition of organic compounds takes place and volatile compounds gives off and condensed into liquid	Easy and simple working Nearly gives 80% bio-oil yield Lower capital cost	Poor fuel quality
Hydrothermal liquefaction	Temperature required between 300–400°C, residence time required 0.5 to 1.0 h and high pressure (5–25 MPa)	The process takes place in aqueous medium followed by complex sequence of reactions	High heating value bio-oil is obtained with low moisture content	Comparatively gives low bio-oil yield Required higher capital cost

Introduction of catalyst in the HTL of crop residues improve the quality and quantity of bio-oil by increasing the heating value due to decrease in the oxygen and nitrogen content of bio-oil. The study on the catalytic HTL was studied many research scientist. The homogeneous such as alkali catalyst or organic solvents and heterogeneous catalyst such as zeolite or supported metal catalyst generally used. Tiwari et al. (2018) reported that yield of bio-oil produced from the rice husk HTL was increased by 6.7% when ethanol used as catalyst for the process as well as the carbon, hydrogen content was found increased and hence the heating value was found increased by 7.79%. Moreover, it is reported that the alkali, heterogeneous catalyst and solvent catalyst has great impact on

the yield, properties as well as on the product distribution of liquefaction process (Duan and Savage, 2010; Nazari et al., 2015; Wang et al., 2013; Zhu et al., 2015b). Further research on the optimum temperature and critical pressure for conversion of biomass into bio-oil is in progress. As well as the cost of HTL is also a significant issue (Gollakota et al., 2018). The comparison of FP and HTL of crop residues is presented in Tables 2 and 3.

Table 3 Comparison of past research on FP and HTL

<i>Temp.</i>	<i>Biomass</i>	<i>Capacity</i>	<i>Type of reactor</i>	<i>Bio-oil yield, wt. %</i>	<i>Heating value of bio-oil, MJ/kg</i>	<i>Reference</i>
<i>Fast pyrolysis</i>						
500°C	Wheat straw	5 kg/h	Fluidised bed	42.0	19.59	Paul et al. (2020)
480°C	Sugarcane bagasse	18 kg/h	Circulating fluidised bed	78.07	18.48	Treedet and Suntivarakorn (2018)
490°C	Maize stalk	2 kg/h	Fluidised bed	42.0	17.8	Ali et al. (2016)
490°C	Cotton stalk	2 kg/h	Fluidised bed	36.0	16.75	Ali et al. (2015)
450°C	Rice husk	1–3 T/h	Fluidised bed	48.1	17.2	Cai and Liu (2016)
<i>Hydrothermal liquefaction</i>						
320°C	Soybean straw	150 g	Autoclave reactor	16.0	32.98	Tian et al. (2020)
250°C	Tomato waste	10 g	Autoclave reactor	45.0	12.3	Zhang et al. (2018)
260°C	Rice straw	15 g	Autoclave reactor	42.12	23.74	Cao et al. (2016)
300°C	Barely straw	60 g	Autoclave reactor	34.9	28.4	Zhu et al. (2015a)

3 Properties of bio-oil

The bio-oil is a liquid produced from the depolymerisation of biomass compounds, having dark brown colour. The bio-oil is basically a mixture of water and oily phase. The percentage of water reported in between 40–50% (Yang et al., 2014). Bio-oil has same elemental composition of biomass and its physio-chemical characteristics varies according to the feedstock and process condition adopted. However, the properties of bio-oil produced from different agriculture feedstock were compared with diesel fuel, as shown in Table 4. The high water and high oxygen content in bio-oil is responsible for the low heating value of bio-oil, i.e., 17–21 MJ/kg. The high viscosity and low pH obstructs the direct application of bio-oil as fuel in diesel engines and turbines (Lu et al., 2008). These poor fuel properties make bio oil only possible to be used in boilers and furnaces (Li et al., 2014). During the storage of bio-oil, viscosity also increases with time. The presence of char or ash in the bio-oil slows the polymerisation reaction and affects the storage stability of bio-oil (Zheng, 2007). The bio-oil produced from the thermal cracking cellulose, hemicellulose and lignin contains number of chemical compounds. Mainly the compounds identified in bio-oil are acids, alcohols, aldehydes, ketones, phenols, sugar, ester, furans, guaiacols and multifunctional components (Ahmad et al., 2019; Gollakota et al., 2018; Nanda et al., 2014).

Table 4 Properties of bio-oils and diesel

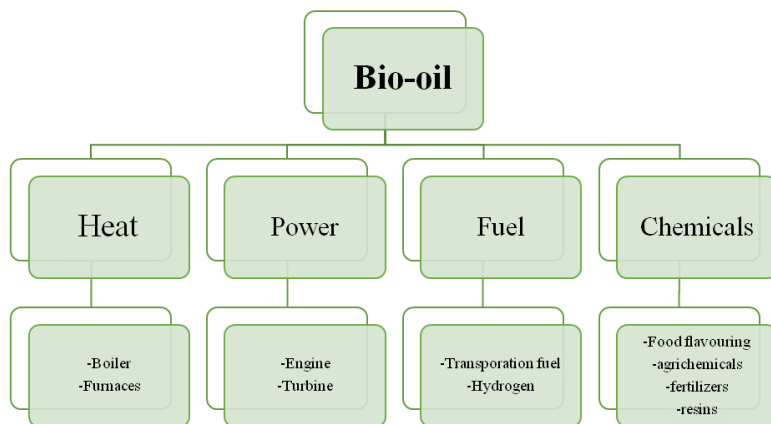
Feedstock	C	H	N	S	O	Water (%)	Density (kg/m ³)	pH	Viscosity (mPa.s @ 40°C)	LHV (MJ/kg)	Reference
Woody biomass	55–65	5–7	0.1–0.4	0.0–0.05	40–50	15–30	1,050–1,250	2.8–3.8	25–100	16–19	Venderbosch and Prins (2010)
Sugarcane bagasse	45.24	7.47	0.36	0	46.93	40.91	1,274	2.4	24.54	18.4	Treedet and Sunitvarakorn (2018)
Rice husk	31.63	8.7	0.5	1.8	48.12	35	1,167	3.22	9.8	17.20	Cai and Liu (2016)
Coconut shell	59.14	5.47	4.21	0.3	30.84	32	1,053.6	3.1	1.47	19.75	Rout et al. (2016)
Peanut shell	50.60	7.01	1.75	0.18	40.46	33.3	1,111	2.88	13.3	14.0	Wang et al. (2009)
Maize stalk	0.59	7.37	1.60	0.01	40.43	35.2	1,066	2.85	9.0	12.6	Wang et al. (2009)
Cherry seeds	58.71	8.24	3.01	0.02	30.02	4.91	-	-	-	26.50	Duman et al. (2011)
Cherry seed shell	56.78	7.36	2.46	0.05	33.25	5.52	-	-	-	23.38	Duman et al. (2011)
Cassava rhizome	70.9	6.3	0.4	-	22.1	16.8	1,100	3.3	18	23.3	Pattiya and Sutibak (2012)
Cassava stalk	70.1	6.6	1.2	-	21.7	19.0	1,100	3.4	15.6	22.6	Pattiya and Sutibak (2012)
Pine wood	45.5	5.8	0.1	-	48.8	25.4	1,170	2.9	13	18.8	No (2014)
Soyabean oil cake	67.89	7.77	10.84	-	13.50	-	1,107	-	72.38 (50°C)	22	No (2014)
Coffee bean residue (oily phase)	42.56	4.32	0.62	-	46.19	22	-	3.6	221	21.59	Yang et al. (2014)
Rice straw (oily phase)	39.76	4.51	0.08	-	49.85	25	-	3.4	210	20.41	Yang et al. (2014)
Diesel	86.58	13.2	65 ppm	0.3	0.01	-	840	-	2.1 (@ 50°C)	40	Rout et al. (2016)

4 Applications of bio-oil

The pyrolysis oil produced from agricultural waste is used to generate heat and electricity. However, the use of it as a fuel in the engine is the emerging topic of research. Because bio-oil has some properties which are very distinct from diesel fuel. The high water content, oxygen content, and viscosity are the main barriers of bio-oil application as fuel in the engine. The energy value of bio-oil was found between 17–21 MJ/kg, which is nearly half of the diesel fuel (Mohan et al., 2006). The bio-oil has been used as fuel in boilers, engines, and turbines as a partial or complete replacement, with some modification in the design of particular or upgrading the bio-oil (Czernik and Bridgwater, 2004). The application of bio-oil as low grade fuel in the boiler was studied at the industrial level by Sippula et al. (2019). Furthermore, the smoke emission analysis of bio-oil for the boiler revealed that the total suspended particulate matter concentration was lower or nearly equal to the wood fired boilers.

The emulsified bio-oil and diesel were used for running the diesel engine (Yuan et al., 2018). The study revealed that the performance of emulsified bio-oil was found better than the diesel, in terms of brake specific fuel consumption (BSFC) and brake specific energy consumption (BSEC). The emission of smoke and CO were reduced while using emulsified bio-oil and CO₂ emission was found increased. Nevertheless, bio-oils are mixtures of water and various organic compounds, mainly including acids, alcohols, aldehydes, phenols ketones, sugars, furans, esters, hydrocarbons, as well as large molecular oligomers. So, it can be used for the production of food flavourings, aromatic, olefins, resins, adhesives, agrichemicals, and fertilisers in the chemical industry (Czernik and Bridgwater, 2004). However, the other end product of the FP like char and gas can be used as fuel for combustion. The bio-oil have wider application as shown in Figure 1.

Figure 1 Different applications of bio-oil (see online version for colours)



5 Conclusions

The burning of crop residues is creating a huge problem such as air pollution and greenhouse gas emission. Conversion of crop residue into fuel is a new and emerging topic of research. Therefore, in this paper two thermo-chemical conversion methods were

reviewed. FP is already successful technology and used for biofuel production from biomass but it has some limitations: poor quality of bio-oil and energy consumption is more. However, HTL used wet biomass for the liquefaction because it is conducted under an aqueous environment. The produced bio-oil has high heating value but the requirement of high pressure during the HTL process makes it uneconomical and hence it is not commercialised. Some major conclusions drawn from this paper are presented below:

- 1 for the conversion of agricultural crop residue into bio-oil, the FP process found more convenient and easy and efficient than HTL
- 2 moreover, HTL is at the early stage development process, and research on lowering the operating pressure could make it commercialised
- 3 the fluidised bed technology is the most used and successful technology for the FP of crop residues
- 4 the study of bio-oil properties showed that it has a heating value between 17–21 MJ/kg, density between 1,000–1,250 kg/m³, and a pH of 2–4
- 5 the bio-oil produced from crop residues has great potential to be used as fuel in boiler, turbine, and engines to produce power and heat
- 6 furthermore, due to the distinctive properties of bio-oil, it needs to be upgraded by techniques such as filtration, solvent addition, emulsification, esterification, catalytic cracking, and supercritical fluids, etc.

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