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REVIEW ON RENEWABLE ENERGY SOURCES BASED ON THERMAL CONVERSION IN THE PULP AND PAPER INDUSTRY

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Abstract

Pulp and paper industries have great potential and prospect for resources of renewable energy. These resources are generated from almost all stages of the pulp and paper manufacturing process. Energy-rich biomass in pulp mills includes bark, sawdust, wood waste, pins, fines, knots, black liquor, and sludge from wastewater treatment plant (WWTP). Energy-rich rejects at the paper mill includes fiber and plastics from the hydropulper unit, and sludge from WWTP. Among these biomasses, bark has the greatest quantity while hydropulper rejects has the highest calorific value. The amount of bark reaches 100-300 kg/t pulp or 60-90% of wood waste in pulp mills, while hydropulper rejects although only reach 5-10% of used paper quantity, has a heating value of 28.81 MJ/kg (air dried basis). Biomass and reject can be converted into renewable energy with a higher energy density through thermal-based conversion technologies such as hydrothermal processing, torrefaction, pyrolysis, and gasification. The renewable energy products include hydrated sludge, biomass pellets, torrefied biomass, bio-oil, and syngas. Several technologies such as gasification are already operating commercially, while several other technologies such as hydrothermal and pyrolysis are still being improved.

Keywords: *biomass, pulp and paper, renewable energy, thermal conversion*

1. Introduction

The potential for renewable energy in Indonesia is quite high but has not been optimally utilized so that it has not been able to achieve the energy mix target as mandated in the national energy policy [1]. The National Energy Policy (KEN) which is a government program as stated in Presidential Regulation No. 79/2014 mandates the use of renewable energy in 2025 with 23% share of the primary energy mix and increases to 31% in 2050 [2]. Currently, final energy consumption is still dominated by the use of fuel oil with a share of 50% with the largest energy users being the transportation sector (43%), the industrial sector (35%), and the household sector (14%) [2]. Indonesia's oil reserves in 2018 amounted to 7.51 billion barrels. This reserve has decreased 0.27% compared to 2017. Similar to petroleum, natural gas reserves in 2018 were 135.55 TSCF (trillions of standard cubic feet). This reserve also decreased by 5.02% against 2017 [2].

Limited fossil fuel reserves encourage the pulp and paper industry to conserve and diversify renewable energy alternatives that are more environmentally friendly and cheaper. Pulp and paper mills have great energy potential because they process large amounts of lignocellulosic materials which contain varying amounts of cellulose, hemicellulose, lignin and

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small amounts of extractives [3]. These resources are produced in all stages of the pulp and paper manufacturing process, namely wood preparation, pulp and paper manufacturing, chemical recovery, recycled paper processing and wastewater treatment. The pulp mill is an energy intensive industry, but it must be able to meet its own energy needs through the integration of various biomass energy conservation technologies in the process. Currently, the pulp mill meets its energy needs by burning heavy black liquor in the recovery boiler and bark and other wood waste in the power boiler.

2. Renewable Energy Resources at the Pulp Mills

The main types of renewable energy resources produced from pulp mills can be classified as biomass reject, black liquor, and sludge from wastewater treatment.

a. Biomass Reject

Biomass reject produced from the pulp mills includes bark from the debarking process, sawdust from the wood cutting process, wood chips from wood storage, pins and fines from chip filtering, knots from the deknoting process, pulp rejects from the screening process, black liquor from the cooking process, and sludge from the wastewater treatment process (Table 1) [4], [5]. These rejects generally have a relatively low moisture content, medium heating value, easy to be dried and processed by burning in a boiler for energy recovery. The amount of bark reaches 100-300 kg/t pulp or 60-90% of wood waste in pulp mills and has a calorific value of about 20 MJ/kg, pins and fines reaches 50-100 kg/t dry pulp, knots reach 2-6% unfiltered pulp or 25-70 kg/t pulp (dry basis) [4], [5], and pulp rejects up to 30 kg/t o.d. pulp.

Table 1. Source of biomass reject at Kraft mill (dry basis) [4], [5]

No.	Biomass reject	Amount (kg/t dried pulp)	Calorific Value (MJ/kg)
1.	Sawdust coming from the slasher deck	10 – 30	
2.	Bark from debarking drum	100 – 300	20.00
3.	Pins and fines from chip screening	50 – 100	
4.	Wood waste from woodyard	0 – 20	
5.	Knots from pulp deknotting	25 – 70	15.44
6.	Pulp reject	30	15.30

b. Black Liquor

Black liquor is spent cooking liquor that contains organics from the wood (mostly lignin) and the inorganic chemicals used for the delignification. In the Kraft pulp process, the addition of sodium hydroxide (NaOH) and sodium sulfide (Na_2S) to wood chips results in the extraction of lignins and hemicellulose from the insoluble cellulose fraction [6]. Black liquor comes out of the cooking process with 15-18% dry solids content. Coming out of the evaporation plant, the black liquor has a dry solid content of about 70-83% [7]. The inorganic part (about 45% dry mass of the black liquor) mainly consists of sodium carbonate and sodium sulfate. Due to presence of the inorganic materials, the lower heating value (LHV) per tonne of black liquor solids is relatively low (LHV, 12.3 MJ/kg) [8]. A pulp mill produces 1.7–1.8 tonnes dry solids of black liquor per tonne of pulp and represents a potential energy of 250–500 MW per mill producing 1000–2000 ADt per day of pulp [9]. Energy content of spent liquor obtained from different type of feedstock and different pulping process are shown in Table 2.

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c. Sludge from Wastewater Treatment

The wastewater treatment process produces sludge, consisting of primary sludge collected in primary clarifier and biological sludge collected in secondary clarifier. Primary sludge comprises short fibers (fines) that cannot be retained on the paper machine. Secondary sludge is a high-protein by-product (biosolids) from the secondary (biological) wastewater treatment that contains bacterial cells, minerals, polysaccharides, nucleic acids, enzymes, etc. [10]. The primary sludge can be dried relatively easily. The primary sludge can be dried to about 70% moisture by belt press, or up to 50% by screw press. Compared to primary sludge, secondary sludge is more difficult to drain. The presence of intracellular water makes secondary sludge difficult to be dried by conventional mechanical dewatering. Mechanical drying can only reduce moisture content by about 85%. This sludge is generally mixed with polymers and dried together to become 25–40% dry solids content [5]. The calorific value of the pulp mill sludge is around 20-24 MJ/kg (dry basis and ash free). The total sludge cake production per ton of product varies, depending on the production process and wastewater treatment process. The kraft pulp mill produces sludge about 58 kg/ton product, sulfite pulp mill about 102 kg/ton product [11].

Table 2. Energy content of spent liquor obtained from different type of feedstock and different pulping process [12]

Pulping process	Feedstock	Proximate analysis (%)			Ultimate analysis (%)				HHV (MJ/kg)
		Ash	Volatile matter	Fixed carbon	C	H	N	S	
Kraft	Pine & spruce	35.93	56.92	7.15	30.67	3.74	0.67	0.00	14.51
Kraft	Reed	24.39	50.00	25.61	33.76	4.15	0.38	0.95	13.35
Soda	Wheat straw	26.74	54.43	11.65	39.84	3.03	0.31	0.64	9.70
Soda	Wheat straw	20.63	65.98	13.39	39.05	4.54	1.00	0.78	14.43
NSSC	Broadleaf wood	24.17	50.62	25.21	36.32	3.43	0.04	5.45	14.98
NSSC	Recycle paper and wood	23.27	66.19	10.54	38.30	4.74	0.39	0.00	15.71

3. Renewable Energy Resources at the Paper Mills

The main types of solid waste generated from paper mills can be classified as rejects from recycled paper, sludge from deinking, and sludge from wastewater treatment [13]. The use of recovered paper results in large volumes of wastes, such as rejected plastics, fibers, and other coarse materials; "stickies" (adhesive residues); and in the case of some mills, deinking sludge (fibers, minerals, and ink). Sludge can be categorized into four sections: (a) primary sludge, which is produced in the primary clarifier and contains mainly fines and fillers; (b) de-inking paper sludge, which is produced during the floatation process for recycled papers; (c) secondary sludge or biological sludge, which is the effluent of the microbial wastewater treatment system and is made of microbial masses; and (d) combined primary and secondary sludge, which is a mixture of primary and secondary sludges from the same plant [14].

a. Hydropulper Reject

Hydropulper reject is waste that is discharged from the repulping process at the initial stage of the pulping process from recovered paper. Plastic-fibre reject are additional wastes that originate primarily from paper recycling processes and contain both fibrous wastes and plastic contaminants [15]. The rejects from recovered paper are impurities and consist of bundle of fibres, foil, staples and metals from ring binders, sand, glass and plastics and paper constituents as fillers, sizing agents and other chemicals with the amount depending on the quality of recovered paper as raw material. Rejects have a relatively low moisture content, high heating values, and easily dewatered [16]. At present, there are forty-five paper industries in Indonesia that operate using recovered paper as raw material. The Indonesian Pulp and Paper Association (IPPA) reports that the consumption of recovered paper for paper production reaches 6,598,464 tons per year [17]. Most of the Indonesian paper industry produces hydropulper reject in the amount of 5-10% of recovered paper, or around 329,923 to 659,846 tons of air dried hydropulper reject per year [18]. The waste component consists of mostly 51% fiber and 49% plastic [19]. Reject generation form different recovered paper grade are shown in Table 3.

Table 3. Reject generation form different recovered paper grade [5], [18]

Paper grade	Recovered paper grade	Rejects (% by dry mass)	
		Heavy-mass and coarse	Light-mass and fine
Market DIP	Office paper	<1	4-5
Graphic paper	News, magazines	1-2	3-5
	High grades	<1	≤3
Sanitary paper	News, magazines, office paper, medium grades	1-2	3-5
Liner, fluting	Old corrugated containers, Kraft papers	1-2	3-6
Board	Sorted mixed recovered paper, old corrugated containers	1-2	3-6

b. Sludge from Deinking

Deinking sludge is produced at a paper mill that produces recycled fiber from recycled paper. This residue contains mainly short fibres or fines, coatings, fillers, ink particles, extractive substances and deinking additives [16]. The amount of deinking paper mill sludge on a dry mass basis generates in the range of 170-600 kg for production of one tonne newsprint to tissue paper [16]. Total solids suspended in the deinking sludge can be categorized into organic materials (such as short or fine fibers) and inorganic materials (such as kaolin, clay, calcium carbonate, titanium dioxide produced from coating materials, ink particles, ink removal additives, dyes, contaminants other pigment-based, and other chemicals used for paper production) [5]. The deinking sludge has a high moisture content in the range of 35-60% with a typical gross calorific value (HHV) of 6-7 MJ/kg on a dry basis [20] and high ash content in the range of 45-65% depending on the quality of the fibre initially brought into the mill [21].

c. Sludge from Wastewater Treatment

About 40–50 kg of sludge (dry) is generated in the production of 1 tonne of paper and of that approximately 70% is primary sludge and 30% secondary sludge [16]. Balwaik and Raut [22] have reported that around 300 kg of sludge is produced for every tonne of recycled paper. Primary sludge consists mainly of cellulose fine fibers and a papermaking filler (such as calcium carbonate) that are lost due to incomplete solid-liquid separations in the production process and is relatively easy to be dried. Secondary sludge consists of high microbial protein content, making it difficult to be dried. The volume of secondary sludge is lower than the volume of primary sludge, as most of the heavy, fibrous or inorganic solids are discharged in the primary clarifier. Secondary sludge often needs to be mixed with primary sludge or other filtration aids to allow adequate drying [16].

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Paper mill sludge can be efficiently utilized to produce fuels and chemicals [23]. Paper sludge has a high ash content and low calorific value so it is necessary to improve the temperature of the combustion chamber to allow the co-combustion of sludge with other fuels, to maintain combustion stability and reduce toxic emission [24]. Comparation of proximate, ultimate and heating values of biomass waste from pulp and paper mills are shown in Table 4.

Table 4. Proximate, ultimate and heating values of biomass waste from pulp and paper mills

No.	Analysis	Pulp sludge [25]	Pulp reject [25]	Knot	Black liquor	Recycled paper sludge	Paper reject [19]	Coal
1.	Proximate analysis (adb)							
	a. MC	9.08	9.42	13.74	28.0	4.83	3.93	12.21
	b. VM	57.53	68.16	60.30	29.5	42.60	81.93	41.86
	c. FC	8.72	17.00	18.81	8.5	4.96	6.98	39.18
	d. Ash	24.67	5.42	7.15	34.0	47.61	7.10	6.75
2.	Ultimate analysis (adb)							
	a. C	31.21	39.43	40.12	44.5	23.09	n.a	55.78
	b. H	5.10	6.50	6.02	4.3	2.73	n.a	6.32
	c. O	37.33	47.79	45.73	44.7	25.81	n.a	30.24
	d. N	1.29	0.35	0.18	0.1	0.49	n.a	0.69
	e. S	0.40	0.51	0.80	6.4	0.27	n.a	0.22
3.	LHV, MJ/kg	12.26	15.30	15.44	13.05	3.56	29.30	22.43

Notes: MC = moisture content
VM = volatile matter
FC = fixed carbon
LHV = lower heating value
C = carbon; H = hydrogen; O = oxygen; N = nitrogen; S = sulfur.

4. Renewable Energy Production Based on Thermal Conversion in the Pulp and Paper Mills

Renewable energy with a higher energy content can be produced from the pulp and paper mills waste through thermal-based technology. Thermal conversion based technologies include hydrothermal treatment, torefaction, pyrolysis, and gasification.

a. Hydrothermal Treatment

Hydrothermal treatment processes can convert waste into value-added resources such as solid fuels such as coal or organic fertilizers. Hydrothermal processing technologies have significant potential for biomass with high moisture content. Hydrothermal liquefaction (HTL) involves the thermochemical conversion of biomass in the presence of subcritical water into a liquid product known as bio-oil. HTL requires an operating temperature of 300–350°C at 5–20 MPa for 5–60 min, wherein water is in the liquid phase. Hydrothermal carbonization (HTC) converts biomass into solid fuel at a lower temperature (180–250°C) and saturated pressure (2–10 MPa). The solid product has carbon content similar to lignite with mass yields varying about 35-60% [26].

HTC of paper mill sludge has been extensively studied in the last few years. Areeprasert et al. [27] performed HTC of paper sludge in the lab-scale experiment at 180–240°C for 30 min and resulted the highest HHV can achieve about 13.6 MJ/kg at 240°C. Afterwards, HTC was performed at the optimized condition (197°C at 1.9 MPa for 30 min) in the pilot-scale experiment and resulted the HHV of 14.7 MJ/kg. The combustion behavior of the products showed that the activation energy of the treated paper sludge was lower than the original material in range of 113–147 kJ/mol [28]. Makela et al. [29] investigated the effect of acid and base additions to the properties of hydrochar produced through hydrothermal carbonization of mixed sludge from the pulp and paper industry. Saha et al. [14] evaluate the fuel properties of the hydrochars produced from various types of paper mill sludges. HHV_{daf} of hydrochar produced at 260°C varied between 11.4 MJ/kg and 31.5 MJ/kg depending on the feedstock. Carbonization of various sludge residues from the pulp and paper industry have also been studied for fuel properties and process energetics, combustion characteristics, and potential use as adsorbents for environmental applications [29].

The hydrothermal conversion of black liquor solid was investigated for producing phenolics and hydrochar [30]. The maximum bio-oil yield of 53.3% was achieved at 50 wt% ethyl acetate/water cosolvent under 300°C for 30 min where phenolics were the main products in bio-oil. Chu et al. [31] investigated the HTC of black liquor acid sediment into phenolics performed in a batch-type reactor at a temperature range of 260-340°C for 0-120 min, the total bio-oil yield was 16-26 wt% based on black liquor solid.

According to Kumar et al. [26], hydroprocessing technology has not been commercialized due to a number of technological and economic crises, namely technology related to process development and optimization to achieve high thermal efficiency, challenges in the field of catalyst recycling and regeneration to increase service life and hydrothermal efficiency processes, and the technology uses high pressure equipment so that the process has a high capital investment.

b. Torrefaction

Torrefaction is a thermochemical conversion process that occurs in a temperature range of 220–320°C, with oxygen limited under atmospheric pressure. The process is based on the breakdown of hemicellulose molecules, with minor changes in the structure of cellulose and lignin. During the process, there is a partial devolatilization that leads to a decrease in mass, whereas the initial energy content is mostly preserved, lead higher energy density [32]. About 70% of the mass is retained as a solid product, and retains 90% of the initial energy content. The torrefied product can then shaped into pellets or briquettes which contain more energy density than ordinary wood pellets [33].

Huang et al. [34] used a laboratory scale batch reactor to perform the torrefaction of waste from pulp industries at 300-320°C with 20 min. The energy densification ratios of the torrefied products of pulp sludge were enhanced as high as 1.50 and 1.26, respectively. The corresponding dry-basis heating values increased to high values of 27.49 and 19.74 MJ/kg. Severy et al. [35] demonstrate a pilot-scale plant for torrefaction of tanoak logs for pulp and paper production and produce torrefied biomass with a higher heating value of 21.2 to 23.0 MJ/kg (dry basis) compared to 19.6 MJ/kg for the original biomass.

c. Pyrolysis

Pyrolysis is the thermal decomposition of organic components in biomass starting at 350–550°C and rising to 700–800°C in the absence of air/oxygen by releasing volatile matter from the biomass [36]. The long chain of carbon, hydrogen and oxygen compounds in biomass breaks down into smaller molecules in the form of gases, condensable vapor, and solid char [37]. Pyrolysis is attracting more interest in producing liquid fuel products due to its advantages in storage, transport and versatility in applications such as combustion engines, boilers, turbines, etc. [37].

Mondal et al. [38] studied the pyrolysis behaviors of various lignin samples from Kraft pulping black liquor of Loblolly pine at 600°C under N_2 atmosphere. The yields of char were 30-

46%. Chutia et al. [39] pyrolyzed dry black liquor solids at 500°C with the heating rate of 40°C/min and produced bio-oil with a calorific value of 29.86 MJ/kg.

Tarelho et al. [40] had studied the production of biochar by pyrolysis of biological sludge from wastewater treatment in the pulp and paper industry in the peak temperature range of 300-600°C which produce biochar in the range 0.40-0.73 kg/kg dry sludge with the organic matter content in the range 55.9 to 75.3%wt. (dry basis).

Pyrolysis of fibre recycling mill contaminated with plastic wastes at 350°C resulting char in calorifc values 20.9-35.1 MJ/kg which are comparable to common bituminous coal used for power generation. Pyrolysis at 550°C produced condensable volatiles with yields of up to 54 wt.%, with the calorifc value up to 41.7 MJ/kg comparable to heavy fuel oil. The estimated net energy yield for the waste streams about 10-25 MJ/kg dry feedstock [41]. Salan et al. [42] investigated catalytic pyrolysis of pulper rejects with catalysts from activated clinoptilolite and meerschaum. The highest liquid (61.4%) and char (32.19%) yields were obtained using 15% clinoptilolite and 5% meerschaum, while the highest gas yield (21.44%) was obtained via the non-catalytic process.

d. Gasification

Gasification is the thermochemical conversion of solid fuels with a gasification agent to produce syngas or synthesis gas, which can be used for the production of energy (heat and/or electricity generation), chemicals, and biofuels [43], [44], [45]. Syngas consists of a mixture of CO, H_2, CO_2, CH_4 (primary components) and H_2O, H_2S, NH_3, tar, and other trace species (secondary components), with a composition dependent on feedstock type and characteristics, operating conditions, and gasification technology [44], [45]. The gasification agent can be water, oxygen, steam, or CO_2 [44], [45], [46].

Gasification of blend composed of 95% of paper rejects and 5% of de-inking sludge produce syngas with net calorific value of 5 MJ/Nm^3 and a total tar content of 11.44 g/Nm^3 at an equivalence ratio of 0.3 [47].

In the pulp and paper mills, black liquor is considered as a major energy source that can be gasified to produce synthesis gas producing various valuable energy products (electricity, dimethyl ether, synthetic natural gas, methanol, hydrogen, or synthetic diesel) and has potential to replace conventional black liquor recovery system with the recovery boiler [8]. Black liquor gasification technologies are distinguished in two major classes: (1) Low temperature gasification, and (2) High temperature gasification. Low temperature gasifier operates at 600–850°C, below the melting point of inorganics, thus avoiding smelt-water explosions, whereas high temperature gasifier operates in the 900–1000°C range and produce a molten smelt [48]. Black liquor gasification can also be integrated with combined cycle technology which has the potential to generate more electricity, or the syngas can be used for the synthesis of bio-methanol and bio-dimethyl ether. Black liquor gasification on a commercial scale has begun to be developed, among others SCA-Billerud process, MTCI fluidized bed gasification, direct alkali regeneration system (DARS) process, BLG with direct causticization, Chemrec gasification, and catalytic hydrothermal gasification [48].

Most of the lime kilns at the pulp mills are heated up with the oil or gas, being biggest users of the fossil fuels. The conversion of the bark and screening fines into product gas using the gasification process enables the replacement of fossil fuels in the lime kiln. The circulating fluidized bed (CFB) gasification process is well suited to bark and screening fines, due to the high share of volatiles and the relatively high reactivity of the solid compounds in the fuel. Valmet has lime kiln bark gasification plants at PT. OKI Pulp & Paper Mills in South Sumatera, Indonesia with capacity of 2 x 110 MW. The fuel for gasification included Eucalyptus bark and screening fines**.** The bark gasification plant could reduce 85% of fossil energy consumption in lime kiln [49].

5. Conclusion

The pulp and paper industry has renewable energy resources that are produced in all stages of the pulp and paper manufacturing process. Currently, the pulp mill meets its energy needs by burning heavy black liquor in the recovery boiler and bark and other wood waste in the power boiler. Research results have shown that biomass waste could be converted into renewable energy with higher energy density through thermal-based conversion technologies such as hydrothermal processing, torrefaction, pyrolysis, and gasification. The renewable energy products including hydrated sludge, biomass pellets, torrefied biomass, bio-oil, and syngas. Several technologies such as gasification are already operating commercially, while several other technologies such as hydrothermal and pyrolysis are still being improved.

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