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Potential applications of waste lignin from the paper and pulp industry in Viet Nam

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ABSTRACT

The conversion of waste lignin from the paper and pulp industry is a potential process to produce chemicals and materials in the industry. With the development and the demand for the pulp and paper industry, the amount of waste lignin will increase remarkably. In Vietnam, the forest tree for the pulp industry is abundant, and the pulp industry has increased in recent years. In parallel, the government planned to develop the material resource and high-tech factories for this industry. In this work, we summarized the pulp and paper industry in Vietnam, then suggest the potential applications of waste lignin in several valuable products.

Key words: Lignin, paper industry, agriculture, construction, carbon fiber, fuel

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INTRODUCTION

Lignin is a by-product of the pulp and paper industry, and it can be considered as a renewable material coming from lignocellulosic feedstock. In the industry, technical lignin can be classified by the lignin production processes (Table 1). The current technology to produce lignin can be classified into sulfurcontaining lignin and sulfur-free lignin technology¹. Sulfur-containing lignin contains Kraft lignin, sulfite lignin, and hydrolyzed lignin from the Kraft process, lignosulfonates process, and enzymatic hydrolysis process, respectively. While sulfur-free lignin comes from Organosolv and soda processes¹. The global lignin market size is around USD 954.5 million in 2019 and grows up 2% each year from 2020 to 2027². For the production, global lignin production is around 100 million tonnes/year in 2015 and expected to increase to 225 million tonnes per year in 2030¹. Lignosulphonate is the most global lignin production. Additionally, lignin from the paper and pulp industry has to treat to remove the pollutants before using, and there are several techniques that have been used for the removal and recovery of lignin³. The technology for the recovery of lignin carried out on physicochemical and biological methods. The physicochemical methods include coagulation and precipitation, adsorption, membrane technologies, ozonation, and advanced oxidation processes³.

Looking at the chemical view, lignins have aromatic backbones⁴, making them an ideal renewable feedstock of aromatic compounds for a range of applications, including automotive brakes, wood

panel products, surfactants, phenolic resins, phenolic foams, biodispersants polyurethane foams, and epoxy resins⁵⁻¹⁰. In fact, native lignin is a heterogeneous polymer with the phenylpropane unit (C9-unit) of the p-hydroxyphenyl (H), guaiacyl (G) and syringyl (S) types^{4,11}. The C9 units are linked to form lignin with C-O-C and C-C linkages (Figure 1), and the most abundant linkage of lignin is β -O-4 linkage (45 – 65%)¹². Based on the structure of lignin, it has many advanced properties such as biodegradability, antiaging to asphalt (in lignin-epoxy resins), antioxidant and UV-protection¹, super strength (for cement), adhesive binding¹¹, good thermal property, watersoluble of lignosulfonates lignin (apply for binders, dispersing agent, surfactant, adhesive and cement additives)¹³. However, lignin is considered as waste and primarily burned for recovering energy, and utilizes only less than 2% to produce chemical products 13,14. Even though the application of lignin increases due to the demand for lignin in animal feed and natural products²; also, lignin is used in the production of bitumen, biofuels, bio-refinery catalysts, concrete admixtures, adhesives, and binders. The application of lignin will be presented in detail in the next section. In Vietnam, the paper industry is an old industry. Before the 20th century, the paper was made in a manual method. In 1912, the first paper factory was established with a capacity of 4000 tonnes/year in Viet Tri city. After that, several factories were built, and the Bai Bang paper factory was the biggest paper factory with 53,000 tonnes of pulp/year and 55,000 tonnes pa-

per/year¹⁷. In recent years, the paper industry has

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Table 1: Classification	or technical lightlis			
Lignin type	Scale	Chemistry	Sulphur content	Purity
Kraft	Industrial	Alkaline	Low	High
Lignosulphonate	Industrial	Acidic	High	Low
Organosolv	Industrial/Pilot	Acidic	Free	High
Soda	Industrial/Pilot	Alkaline	Free	Moderate
Hydrolytic	Industrial/Pilot	Acidic	Low/Free	Moderate

increased production capacity. In the period 2016 -2019, paper production increased by around 31% each year¹⁸, the production output also increased by 25.7% annually. Additionally, the consumption and export of paper went up to 12.3% and 65.1%, respectively¹⁸. In fact, the total paper consumption and export are estimated at 5.432 million tonnes and 1 million tonnes in 2019, respectively¹⁹. In which, Vietnam produced ca. 4.43 million tonnes of pulp and paper in 2019, which was higher than 2018 around 20.6%, based on the data of the Pulp and Paper Associate²⁰. Therefore, a large amount of by-product, lignin, will become available to transform into valuable products. In this work, we summarize the pulp and paper industry in Vietnam and suggest the potential applications of waste lignin in Vietnam.

Table 1: Classification of technical ligning 15,16

REVIEW

The pulp and paper industry in Vietnam *Materials*

Vietnam is a tropical country and has the mountain account for ca. 40% of the country's land area. Vietnam has a huge amount of forest and plantation trees for the pulp and paper industry. The main material region is located in the North and Central of Vietnam, while the big pulp and paper factories are located in the South of Vietnam¹⁷. Therefore, the government decided to plan the regions for the development of paper raw material plants, as shown in **Table 2**.

With this plan, wood chips – material for pulp and paper – have been increased and become abundant. At present, wood chips are enough for the pulp and paper production in Vietnam and export as well. Additionally, China is the biggest pulp market in the world, and they need around 21.44 million tonnes of pulp/year¹⁸. China is our neighbor making a reduction in transportation cost. Therefore, it is great to develop material for the pulp and paper industry due to the demand for our own country and export as well. Currently, the export of wood chips has been increased year by year and reached more than USD 1.3 billion per year, as shown in **Table 3**.

The pulp and paper industries

Though Vietnam has a huge amount of materials for the pulp and paper industries, the most product produced is packaging paper (82%) from the waste paper (**Figure 2**)¹⁸. In which, we have to import waste paper for the packaging paper production due to the low recovery waste paper (< 40%) in Vietnam²². For the writing paper, the production responses for 55% of the national demand and it needs to import 45% of the demand, around 483,000 tonnes/year¹⁸. It is due to the low-tech factories in Vietnam¹⁷. Therefore, we need to invest the fund for the development of the hitech pulp and paper industry to increase the value of this industry and reduce the environmental effect.

In order to enhance the national production of pulp and paper, the government planned to invest in the hi-tech pulp and paper industries (**Table 4**) to increase production capacity and reduce the cost and pollution effect²¹. The investment plan helped to improve the pulp and paper industries of Vietnam and reduce the dependence of importation. The enhancement of this industry also boosts an increase in the plantation industry, making the increase in the economy in rural areas. Additionally, the spread of the pulp and paper factories near the area of materials reduces the transportation cost of materials and products as well.

Interestingly, several companies recently invested in the high-tech pulp/paper factories in Vietnam to upgrade the paper sector. Indeed, Lee & Man Vietnam invested USD 650 million to build a paper factory with a production capacity of 420,000 tonnes/year in Hau Giang province²³. This factory mainly produces packaging paper from more than 95% of wastepaper. However, this factory applies high technology to produce high-quality paper from waste paper and reduces water pollution. In parallel, the government also encourages investment in the pulp industry and allows only the high technology plant to avoid any risk of the environment. The joint-stock company between Thai Binh Xanh and Povry declared to invest VND 11,650 billion in a pulp factory in Quang Tri province²⁴. This pulp factory planned to produce

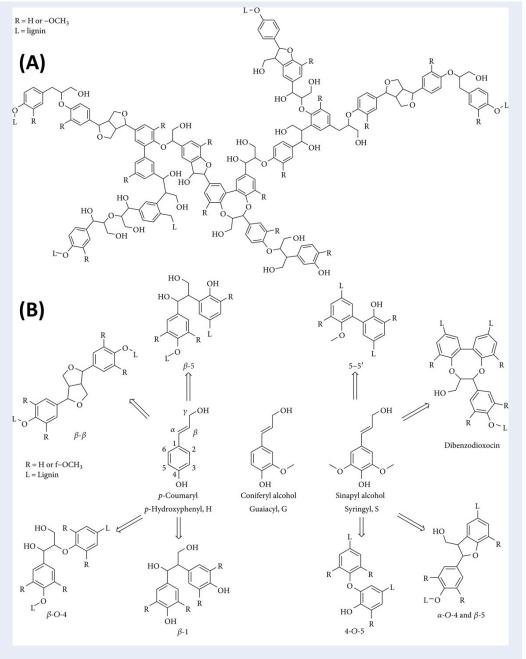


Figure 1: (a) A typical structural model of lignin; and (b) the linkages in lignin. Reproduced from Open-access ref.¹²

Period	Region (Unit: ha)							
	Northwest	Northeast	Red River Delta	North Central	South Central Coast	North Central High- lands	South Central High- lands	Mekong River Delta
2011-2015								
Forest area in 2015	123,500	104,550	142,800	136,992	159,501	86,781	75,950	42,100
Afforestation after exploitation	51,000	63,500	82,500	63,000	80,000	32,500	38,500	34,500
Planting new forests	61,100	27,500	34,800	43,500	64,000	47,500	29,500	800
2016 - 2020								
Forest area in 2020	145,500	110,550	164,300	157,492	173,001	104,781	85,450	42,100
Afforestation after exploitation	121,247	92,122	136,917	131,243	144,168	72,318	71,208	35,083
Planting new forests	22,000	6,500	21,500	20,500	13,500	18,000	9,500	0
2021 - 2025								
Forest area in 2025	145,500	110,550	164,300	157,492	173,001	104,781	85,450	42,100
Afforestation after exploitation	132,269	100,496	149,364	143,175	157,274	95,255	77,682	38,273

 Table 2: Regions and areas for the development of paper raw material plants²¹

Table 3: Value of Vietnam's woodchip export till the end of April 2019^{22,23}

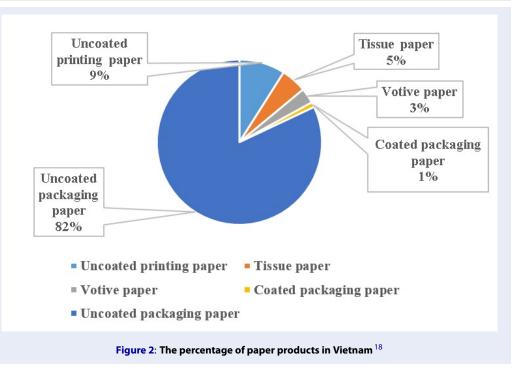
Year	Quantity (dry tons)	Value (USD)
2010	3,996,381	437,021,153
2011	5,179,093	761,871,879
2012	5,820,885	796,351,503
2013	7,063,461	983,390,245
2014	6,971,740	958,044,609
2015	8,062,563	1,166,400,705
2016	7,221,613	986,850,338
2017	8,201,298	1,072,656,296
2018	10,375,720	1,340,083,064
The end of April 2019	3,973,554	557,273,279

300,000 – 700,000 tonnes/year with friendly environmental technology. Those investments along with other investments from Vietnam and foreign companies, will boost the pulp and paper industry in Vietnam, making an increase in the number of laborers, economy, and the following industry as well, such as the industry of lignin, by-product from the pulp and paper industry.

The applications of lignin

As mentioned, lignin is considered as a waste, but its application is rising in recent years. Indeed, lignin has a broad application in many fields, and it has many specific benefits in each field. The summary of the lignin application is illustrated in **Table 5**.

Besides those applications with the technologies developed by Borregaard, several technologies have been developed focusing on the transformation of



lignin by chemical processes. The new technologies focus on lignin depolymerization (hydrogenolysis, pyrolysis, oxidation, hydrolysis, and gasification), functionalization of hydroxyl groups (alkylation, phenolation, urethanization, and etherification), synthesis of chemically active sites (hydroxyalkylation, amination, nitration, and sulfonation), and production of lignin graft copolymers¹. Moreover, there are some technologies showing novel applications of lignin in the production of lignin-based carbon fibers, BTX, phenol, oxidized products, energy storage devices, nanocomposites, drug delivery systems, tissue engineering, filtration, and heavy metal capturing devices¹. Figure 3 shows several potential conversions of lignin into useful products; it can be converted into phenolic compounds applying many industries or copolymerize to form a copolymer in the plastic industry.

The potential applications of lignin in Vietnam

Based on the current industry in Vietnam and the demand for the industry, lignin has many potential applications in Vietnam. These potential applications rely on the current condition in Vietnam. Vietnam is in the process of transforming from an agricultural to industrial country; therefore, the demand for both agriculture and industry has been increased in recent years. The increase of industry boosts the de-

velopment of the economy and the demand for development of house and building, materials as well as bulk chemicals and fuels. There are several potential applications of lignin such as dispersants, paper sizing, industrial cleaners and water treatment, concrete admixtures, unbaked bricks, and plant nutrition (Figure 4). Those products have many advantages, e.g., the advantages of dispersants are milling economy, a wide range of heat stability, controlled fiber staining, no azo reduction, lower paste viscosity in the formulation, improved cost/performance after standardization of dye strength, and environmentally friendly²⁹. While the advantages of plant nutrition are Improving formulations and plant nutrition, stable solutions, custom formulations³⁰; of concrete admixtures are improved strength and durability of the cured concrete, reduced formulation cost, increased workability of the concrete mix, cost effective water reducers³¹; of Industrial Cleaners and water treatment are cost-effective additives for industrial cleaning to disperse dirt particles, in water treatment formulations to reduce the fouling of cooling water as well as a low-cost conditioning agent for boiler water sludges, enhance the metal cleaning and water treatment abilities of other components typically present in these types of formulations and are compatible with sodium citrate, gluconates, EDTA, NTA, and triethanolamine³²; of papersizing provide a simple and ready to use green solution for increasing the

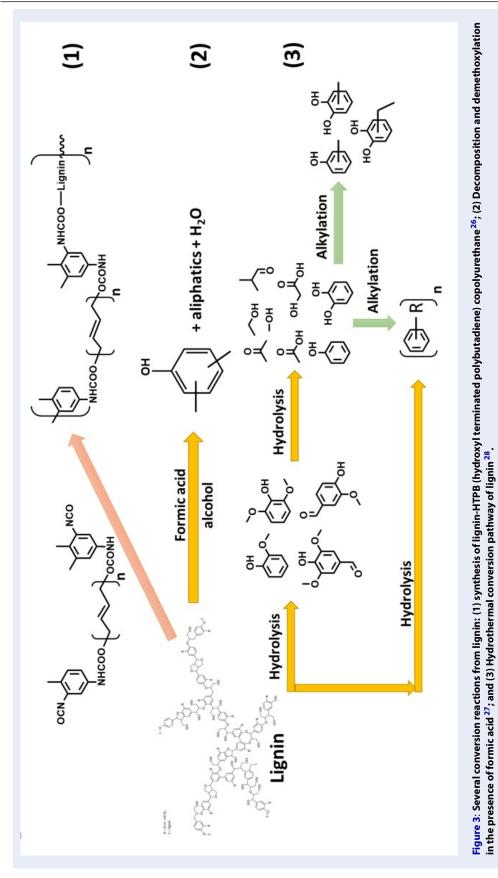




Table 4: The list of investment projects by each stage²¹

Seq.	Company/Factory Region Production capacity (tons/year)			Period	
			Pulp	Paper	
1	Vietnam pulp and paper company	Phu Tho	-	150,000	2011-2015
2	Phuong Nam pulp factory	Long An	100,000	-	2012-2020
3	An Hoa company	Tuyen Quang	130,000	-	Produced
				140,000	2011-2015
4	Pulp factory	Southern		650,000	2011-2015
5	Pulp and paper factory	North Central	100,000	180,000	2011-2015
6	Pulp factory	South Central Coast	250,000	-	2012-2015
7	Pulp factory	Red River Delta	50,000	-	2010-2015
8	Paper factory	Market area		1,080,000	2011-2015
9	Pulp and paper factory	Central Highlands	130,000	200,000	2010-2020
10	Pulp and paper factory	South Central Coast	130,000	400,000	2010-2020
11	Pulp and paper factory	Mekong River Delta	330,000	420,000	2011-2020
12	Pulp and paper factory	Northeast	250,000	200,000	2016-2020
13	Pulp and paper factory	North Central	150,000	200,000	2016-2020
14	Paper factory	Market area	-	1,130,000	2016-2020
15	Paper factory	North Central	-	200,000	2021-2025
16	Expanding pulp and paper factory	Red River Delta	300,000	200,000	2021-2025
17	Pulp and paper factory	South Central Coast	300,000	250,000	2021-2025
18	Expanding pulp and paper factory	Central Highlands	130,000	200,000	2021-2025
19	Other projects	Market area		2,855,000	2021-2025
Total			2,350,000	8,455,000	

strength of recycled based corrugated mediums on size-film press or spray sizer, increased CMT, SCT, RCT, reduced energy consumption and steam energy savings, improved runability, enhanced optical properties ³³; and of unbaked bricks are achieving the standard in the compressive and yield strength, low-cost material, reducing the environment effect ³⁴.

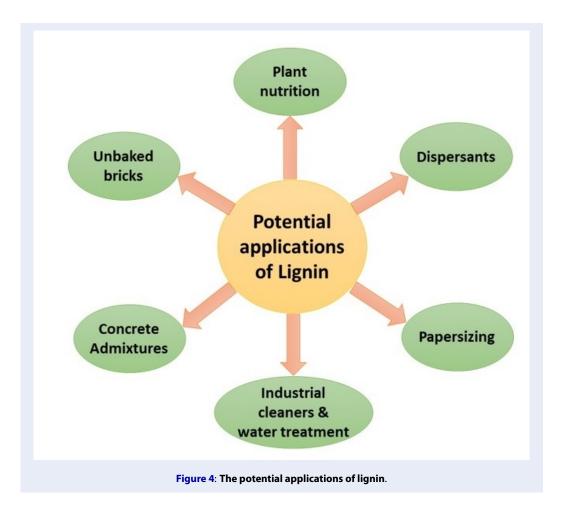
Among the potential products, concrete admixtures, unbaked bricks, plant nutrition, carbon fiber, and bulk chemicals are the most potential products in Vietnam. In fact, the concrete mixtures, unbaked bricks, and plant nutrition are the most viable applications of lignin in Vietnam due to the high demand for raw materials in these industries as well as easy production processes. Moreover, the production of bulk chemicals from lignin is also a promising process to produce chemicals from biomass, enhancing the environment quality (**Figure 3**). For unbaked bricks, they have currently been developing in Vietnam to reduce the environmental effect, and these products can also enhance the use of waste materials such as waste lignin from the pulp industry. Dr. Bui from the University of Transport and Communications developed unbaked bricks from inorganic solid waste of the pulp industry³⁴. The unbaked bricks synthesized showed an increase in water absorption and a decrease in compressive and yield strengths with an increase in the percentage of the pulp's solid waste (Table 6). Interestingly, the synthesis bricks reached the Vietnam standard TCVN 6477:2016 with the use of 40 and 50% of the pulp's solid waste. Besides that, the use of 60% of solid waste was also a promising candidate though having lower yield strength and higher water absorption in comparison to the standard (M150, Table 7). The high amount of using solid waste can be tailored to enhance the physical properties and reduce water

Table 5: The applications of lignin²⁵

Table 5: The	e applications of lignin ²⁵	
Seq.	Fields	Application and benefits
1	Agriculture	 Lignin-based dispersants, processing aids, and binding agents to the agro- chemical industry. Natural plant nutrition
2	Animal Feed Additives	- Aquafeed and Fishery - Pig feeds - Poultry feeds - Ruminant feeds
3	Battery Additives	- Organic additives for lead acid batteries
4	Carbon Black Disper- sions	- High performance dispersants for aqueous carbon black dispersions
5	Ceramics	 Increasing mechanical strength Reducing cracks and breakages Decreasing energy costs and breakage rates Achieving good plasticity with less water Reducing final costs
6	Concrete Admixtures	- Improved strength and durability of the cured concrete - Reduced formulation cost - Increased workability of the concrete mix
7	Dyestuff Dispersants	- Sustainable primary and secondary dispersants for use in disperse, vat, reactive and acid dyes.
8	Emulsions	- Lignin-derived emulsion stabilisers
9	Gypsum Board	- Use as water reducers and processing aids in gypsum board manufacturing.
10	Industrial Binders	- Sustainable & environmentally friendly organic binders for the agglomeration of valuable metallurgical fines
11	Industrial Cleaners & water treatment	 Use as cost-effective additives for industrial cleaning to disperse dirt particles Water treatment: formulations to reduce the fouling of cooling water A low-cost conditioning agent for boiler water sludges.
12	Oil and gas	- Innovative high-performance solutions for petroleum drilling applications,
13	Papersizing	- Green solution for increasing the strength of recycled based corrugated medi- ums on the size-film press or spray sizer
14	Resin extension	- Extend finished resins or to replace the phenol in such resins fully or partially.
15	Road & soil dust control	- Highly effective dust suppressants for unpaved roads and other areas such as airstrips, road shoulders, and racetracks,

Table 6: Compressive and yield strengths and water absorption ³⁴

Name of prod- ucts.	Cement (%)	Ashstone (%)	Inorganic waste (%)	H ₂ O (%)	Compressive strength (MPa)	Yield strength (MPa)	Water absorption (%)
M1	10	10	80	15	4.26	0.66	18.50
M2	20	10	70	15	8.93	2.03	16.05
M3	30	10	60	15	15.33	2.67	12.69
M4	40	10	50	15	17.80	4.70	11.26
M5	50	10	40	15	22.30	5.57	10.42



Standard bricks (M)	Compressive strength (MPa)	Yield strength (MPa)	Water absorption (%)
M35	3.5	-	< 14
M50	5.0	1.6	
M75	7.5	1.8	< 12
M100	10.0	2.2	
M125	12.5	2.5	
M150	15.0	2.8	
M200	20.0	3.4	

Table 7: Compressive and yield strengths, and water absorption of concrete following the TCVN 6477:2016
standard ³⁴ .

absorption to reduce the price of unbaked bricks. As mentioned, lignin can be applied to produce highvalue materials such as carbon fiber precursor and bulk chemicals (Figure 3). Lignin has high advantages in using carbon fibers' precursors due to high carbon yield and negligible toxic through the carbonization process³⁵. According to the literature, lignin is the best carbon-fiber precursor³⁵. Currently, electrospinning is a promising process to produce carbon nanofibers^{36,37}; Lallave et al.³⁶ produced carbon fibers with diameters less than 200 nm. Additionally, lignin can be converted into chemicals and fuels through the hydrodeoxygenation (HDO) process³⁸. The HDO process transforms lignin into phenolic compounds for surfactants and paint industries, while further HDO process produces hydrocarbons as fuels from lignin. Currently, Hossain et al. 39 proved that the depolymerization and deoxygenation of the lignin model could be carried out without the addition of hydrogen, opening a new way to develop and produce low-cost chemicals and fuels from lignin.

CONCLUSIONS

In this work, we summarized and suggested potential products and processes from waste lignin of the pulp and paper industry for the Vietnam market. The industry of lignin is a promising field to enhance the contribution of the wood industry. In parallel, the increase of demand for paper and the investment as well as the good policy of the government, will improve the lignin market and material for further processes. For the Vietnam market, the products from lignin were used for agriculture, construction and basic dispersants are the best to develop due to the low-cost and huge demands. Besides, the development of high-tech materials such as carbon fiber and high valuable raw materials are also considered to gain the industry in Vietnam for the future.

LIST OF ABBREVIATIONS

BTX: Benzene – Toluene - Xylene CMT: Corrugating Medium Test EDTA: Ethylenediaminetetraacetic acid HDO: the hydrodeoxygenation NTA: Nitrilotriacetic acid RCT: Ring Crush Test SCT: Compression Test TCVN: the Vietnam standard

COMPETING INTERESTS

The author(s) declare that they have no competing interests.

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