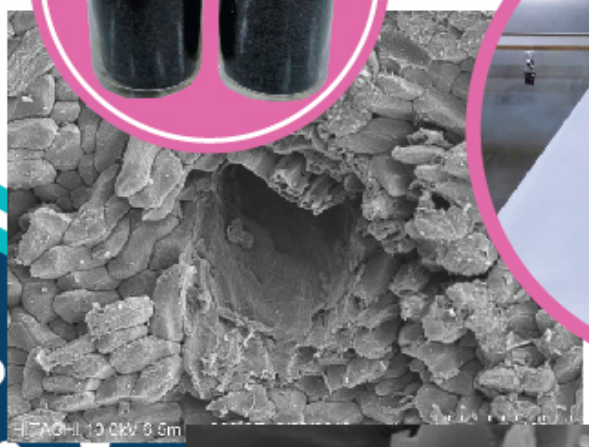
Issue 20. Jan-June 2020
ISSN No. 1985-4951

INTROPica

INSTITUTE OF TROPICAL FORESTRY AND FOREST PRODUCTS

Centre of R&D in Tropical Biocomposite and Bioresource Management

Borderless Potential of Lignocellulosic Biomass



Assalamualaikum and greetings.

Welcome to the issue 20 (Jan – June 2020) of INTROPica. This issue focuses on "Borderless Potential of Lignocellulosic Biomass". What a wonderful way to acknowledge the wealth of insight and lessons learned and shared amongst our peers, essential to continuous improvement.

In the midst of closing this issue, our entire workforce has implemented the concept of working from home as the COVID-19 pandemic affects the entire world including Malaysia. We know that most of our readers are now working from home, and our magazines typically are distributed to our collaborators and guests during their visits to our office in INTROP (Institute of Tropical Forestry and Forest Products). To help reduce waste and prevent stacks of magazines piling up in the office where no one will read them, we are producing this magazine exclusively as a digital edition. Every issue of the magazine dated back to its founding year, in 2008, is available at introp.upm.edu.my.

Borderless potential of lignocellulosic biomass in creating versatile materials with highly tuneable properties and a broad range of applications dominates this issue of the magazine; with article after article making the case for an approach that will enable us to overcome the global crisis in a sustainable and eco-friendly manner. Owing to the importance of lignocellulosic biomass being the most abundant and bio-renewable biomass on earth, this issue provides insights into the potential of lignocellulosic biomass as an alternative platform to fossil resources.

Developments in the valorisation of lignocellulosic biomass still remains a big challenge together with many opportunities. Thus, extensive research is currently being undertaken all over the world to convert lignocellulosic biomass to value-added products at high selectivity, and yields at economical costs. One of the most important goals is to fractionate lignocellulose into its three major components; cellulose, hemicelluloses and lignin. Various pre-treatment approaches have been developed to increase the accessibility and biodegradability of these components for enzymatic or chemical action. Once these components are isolated, target compounds can be obtained through either chemo-catalytic or microbial production processes. That is why future developments in the valorisation of lignocellulosic biomass are directly correlated to improvements in the fields of chemical and microbial synthesis.

Owing to the recent advancements in these fields, the number and diversity of lignocellulosic biomass-based product and specialty chemicals have been rapidly increasing. Furthermore, biorefinery technologies have been developed to refine lignocellulosic biomass in similarity to petrochemistry for producing biofuels, biochemicals and biopolymers. Through research and development, many more lignocellulosic biomass derivatives are leaning towards the realization of commercial production.

Concept of 'Lignocellulosic biorefinery' that creates values from lignin is increasingly coming to the forefront, an area that has been impeded by the heterogeneity of lignin breakdown products. Recent development in lignocellulosic biomass research indicated that it is possible to produce modified lignin comprised largely of a single monomer and with consequently more simplified structure. In addition, the use of engineered zip-lignin will probably pave the way to increasing the usability of lignin-breakdown products. Combining modifications to polysaccharides and lignin using synthetic biology approaches may provide a long-term route to developing tailored-biomass feedstock to completely replace petroleum at the heart of the fuel, materials and chemical industries.

As our behavioural patterns change due to the COVID-19 crisis, our impact on nature and the environment changes too. Most people have now developed a renewed sense of optimism that damage done to the environment is reversible. The pandemic has given us an opportunity to press reset on our relationship with nature and get on track to a more sustainable future. We are hopeful that the pandemic will ease its condition worldwide soon, due to the brilliant work of scientists and medics so that the society can return to something approaching normality before the pandemic.

Stay safe and healthy! We'll get through this together.

INTROPica
HIGHLIGHTS

TRANSFORMATION OF LIGNOCELLULOSIC BIOMASS INTO INDUSTRIALLY DRIVEN PULP AND PAPER PRODUCTS: TIME TO CHANGE

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For centuries, wood has been used as a primary resource for pulp and papermaking production. A perceptible change of applying other than wood resources can be brought into pulp and paper industry by looking at its massive demand for raw materials. In principle, raw materials for papermaking are categorised into 2 groups: (1) virgin and (2) recycled fibres which can be derived from (1) woody, non-woody plants and agricultural wastes and (2) recycled paper or paperboards respectively. Due to environmental concerns on the depletion of natural forest resources in most global regions, the utilisation of lignocellulosic biomass (LCB) such as non-woods and agricultural waste fibres is expected to become an important fibre resource.

LCB is identified as the most abundant, economical, inexpensive, terrestrial biomass portion and a highly renewable natural resource worldwide that could be possibly replenished in a sustainable manner. Its utilisation is well known in bioproducts, bioethanol, power generation or even heat production processes as well as pulp and papermaking. Cellulose and hemicellulose are called polysaccharides while lignin is a polyphenolic polymer. In papermaking, the fibres need to be prepared into individual fibres before forming them into a sheet of paper in order to create the best bonding among the individual fibres network. The initial challenges of using LCB in pulp and papermaking are (1) to develop efficient and cost-effective methods in separating the targeted cellulose component from lignin and hemicellulose and (2) to convert fractionated cellulose individual fibres from LCB into paper sheets. In order to achieve the objective, the selection of pulping condition or combinations of chemical and mechanical approaches have to be critically studied to address the fact that different sources of LCB have different specific lignin, hemicellulose, and cellulose contents. Besides, more approaches have arisen to improvise the process via pre-treatment, enzymatic hydrolysis or fermentation methodology to be more feasible, practical and accept by the industry. The most challenging stage is pre-treatment that functions to partly destroy the lignin matrix hence to ease the releasing process of cellulose into individual fibres in the following processes. Thus, there are a number of approaches that have arisen to improve the overall prospect of LCB-derived industries. It is expected that LCB like bamboo will be one of emerged raw material resource in the world (Chen et al. 2019, Kaur et al. 2016).

The attributes of LCB make it attractive as a bulk raw material for chemical and biotechnology industries; however, there are a variety of technical challenges to the industrial use of LCB. Some obstacles in manufacturing LCB fibres at commercial scales are expected in each processing step beginning with mode of harvesting, collecting system, way of handling, appropriate transportation, technique of storing, types and level of washing, bleaching sequences, papermaking process, chemical recovering, sustainable supply chain and in fact the finishing section. High technology and innovation place LCB to be more economical as a raw material for pulp and papermaking. Hence, the future of LCB as a raw material for pulp and papermaking looks brighter day by day. In pulp and papermaking history and performance, China is leading the pulp and paper industries in using non-wood raw materials. It is reported that China already produced two thirds of the non-wood pulps worldwide in the past 20 years (Chandra 1998). The reasons for the need to use LCB in pulp and paper production are because of (1) shortage of fibre (2) increasing demand (3) availability of LCB fibres (Hammet et al. 2001) and the uniqueness of LCB fibres anatomy for niche and specific specialty paper besides writing or printing grade paper. The challenges that industry might face are (1) energy usage and, (2) waste generation and management that need to be critically carried out (Rullifank et al. 2020). This related to type of raw material, pulping condition and grade of paper products which matters to a great extent as shown in Table 1.

Table 1 Category of LCB comprised a part of non-wood and agricultural waste raw materials with different types of processing and paper products.

Category of LCB	Raw material from LCB	Condition of pulping	Types of pulp or paper	Country	References
Non-wood	Bamboo Semantan *(Gigantochloa scortechinii)*	Pre-treatment : 2 % NaOH, 6 h, 60°C. Pulping: Kraft pulping at 14 % and 18 %.	Unbleached pulps.	Malaysia	Ainun et al. (2018)
		Pulping: Alkaline sulfite anthraquinone (AQ) and methanol (ASAM) pulping process, 16% NaOH, 90 min.	Unbleached pulps.	Malaysia	Moradbak et al. (2016a)
			High durable paper and paperboard.	Malaysia	Moradbak et al. (2016a)
	Bamboo (i) Mayan *(Gigantochloa robusta)* (ii) Sembilang *(Dendrocalamus giganteus).*	Pre-hydrolysis kraft Bleaching: D0ED1D2 and D0EpD1 D2	Dissolving pulp	Indonesia	Purwita & Sugesty (2018)
	Hemp Mauritian hemp	Pulping: Soda cooking, 12% NaOH, 90°C, 90 min. i. 100% Mauritian hemp fibres ii. mixtures of Mauritian hemp fibres with Elephant grass fibres and wastepaper.	A4 sized papers.	Mauritius	Amode & Jeetah (2020)
	Kenaf	Pulping: Soda-AQ 2 stage, 19% NaOH, 90°C, 90 min.	Writing and printing grade paper.	Malaysia	Mahmudin et al. (2012)
Agricultural waste	Oil palm empty fruit bunch (EFB)	Pulping: 2% NaOH, 2 h pre-treatment time, 121°C.	(i) Corrugating medium. (ii) Fibreboard.	Indonesia	Harsono 2020
	Pineapple leaf	Pulping: Soda pulping 10% NaOH, 180 min.	biodegradable plates with biocoating	Thailand	(Iewkittayakorn et al., 2020)
	Bagasse Ethiopian sugarcane bagasse	Pulping: Sulfur free soda pulping process, 130°C, 10% NaOH, 60 min. Bleaching: Single-stage hydrogen peroxide process.	A4 Printing Paper	Ethiopia	Mamaye et al. 2020
	Wheat straw	Pretreatment: Crude xylano-pectinolytic enzymes. Pulping: Soda-AQ pulping 160°C.	A4 printing paper	India	(Varghese et al., 2020)
	Sugarcane bagasse and oil palm empty fruit bunches.	Pulping: Chemical pretreatments, 121°C, pressurized refining under steaming conditions (thermomechanical condition).	Paperboards	Indonesia	Mulyantara et al. (2017)

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INTROPica
HIGHLIGHTS

QUATERNIZED LIGNOCELLULOSIC FIBRES AS ADSORBENTS FOR TEXTILE DYE REMOVAL

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Introduction

Cellulose and its derivatives are low-cost, widely resourced, non-toxic, biodegradable, and renewable raw materials. In addition, they can remove coloured matter from dye wastewater, metal ions and some organic compounds from wastewater. Thus, if these waste byproducts are converted into adsorbent materials, it would add a considerable economic value, decrease the waste disposal cost, and also provide an inexpensive option to the currently available commercial adsorbents. The agricultural waste is seen to be very promising raw materials for the production of the adsorbents with good adsorption capacity, satisfactory mechanical strength and lower ash content (Savova et al., 2001).

Several attempts have been made to find easily available and inexpensive adsorbent materials which can help in the removal of pollutants using agricultural solid by-products (Rafatullah et al., 2010). Many researchers have published studies investigating various adsorbents made of agricultural solid wastes and have tested them for dye removal from effluent waste (Mafra et al., 2013).

Quaternization on lignocellulosic fibres

Although the use of agricultural biomass as an adsorbent for dye removal is encouraging for reducing the material costs. However, most of this agricultural biomass has a low adsorption capacity. Hence, the biomass has to be physically or chemically modified for improving its selectivity and adsorption capacity. The chemical modification process like quaternization on the lignocellulosic biomass is more advantageous as compared to thermal modification, due to its low process costs. The physical treatment processes like carbonisation or thermal activation need a higher temperature (300 to 900 ºC) in the furnaces; therefore, this process consumes a lot of electricity and needs costly equipment like furnaces. Hence, by chemically treating the biomass, higher energy consuming steps like thermal activation and carbonization could be substituted by simplified chemical reactions like quaternization and mercerization. This reduces the production costs for chemically modified adsorbents by 60% in comparison to the thermally activated adsorbents (Koay, 2013).

The chemical process of quaternization adds the $R\text{-}NH^{3+}$ groups (wherein R is the structure of the natural fibre, like cellulose, lignin and hemicellulose) on the lignocellulosic fibre surface using the epoxy substitution reaction, thus increasing their affinity for anionic substances and promoting the ion-exchange adsorption process (Bhatnagar et al., 2011; Malakootian et al., 2011). are a natural, straight-chain polymer, consisting of β-D-glucopyranose monomers which are covalently linked together by the β-1, 4-glycocidic bond. The cellulose molecule consists of several hydroxyl groups; however, the chemical substitutions only occur at the hydroxyl group present at the C-6 conformation, due to the stearic hindrance (Perez and Samain, 2010). Several quaternizing agents are available, depending on the type of chemical reaction needed. In the studies, the researchers have used trimethylamine and (3-Chloro-2-Hydroxy Propyl) TrimethylAmmonium Chloride (CHPTAC). The quaternizing reactions which use triethylamine need pyridine as the main catalyst and N, N-DiMethylFormamide (DMF) as the solvent; however, these chemicals are very expensive and also carcinogenic in nature. Hence, CHPTAC was a more preferred quaternizing agent, as it required only sodium hydroxide, which is a cheap and a relatively safe chemical in the chemical (De Lima et al., 2012).

The quaternization cellulosic reaction is carried out in two steps. In step one, the chlorohydrin form of CHPTAC is converted to its quaternary epoxy intermediate (i.e., 2,3-epoxypropyltrimethyl-ammonium chloride) as described in Figure 1a The second step of the quaternization reactions involves the reaction of the quaternary epoxy intermediate

with the –OH groups present at the C-6 position of the cellulosic polymer to yield a quaternized cellulosic fibre as described in Figure 1b (Wang, 2005).

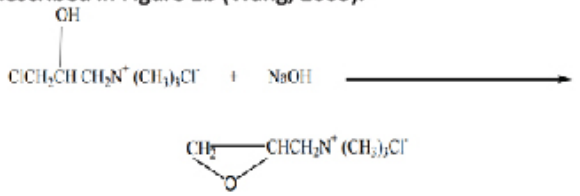

Figure 1a: Conversion of the quaternary chlorohydrin reagent to its epoxy form

The resultant quaternized lignocellulosic fibres showed a higher affinity and adsorption capacity for the anionic substrates since quaternization improved the surface area, porosity and the chelating functional groups of the fibre. In Figure 2, the probable electrostatic interaction between the anionic dyes and the quaternized cellulosic fibre has been described (Anjaneyulu et al., 2005). Table 1 has summarized the studies which have been carried out using the quaternized lignocellulosic fibres as the adsorbing material for removing the anionic substrates.

Conclusion

Quaternized fibres are potential adsorbents to capture textile dyes. The advantages of quaternized fibre adsorbents are due to their minimum processing cost (utilization of abundant availability of fiber as agrowaste) and energy saving as no thermal treatment is needed during processing as compared to activation process for activated carbons.

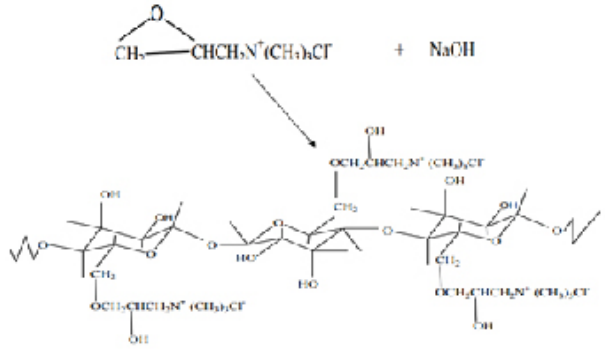

Figure 1b: Reaction of the quaternary epoxy with the cellulosic fibre

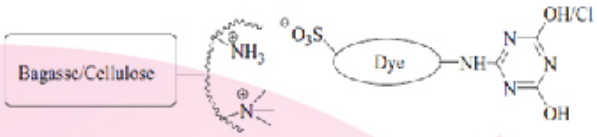

Figure 2: The probable electrostatic interaction between the quaternized cellulosic fibres with the anionic dyes (Anjaneyulu et al., 2005).e

Table 1 Researches that were conducted using quaternized lignocellulosic biomass for the removal of anionic dyes

Quaternized biomass	Dyes	References
Cotton fibres	Congo red	(Xiong et al., 2015)
Palm kernel shell	Methylene blue	(Koay et al., 2014)
Flax shive	Reactive black 5	(Wang and Li, 2013)
Wheat straw	Reactive red 228	(Zhang et al., 2012)
Wheat straw	Methyl orange	(Xu et al., 2010)
Sugar cane bagasse	Acid green 25	(Wong et al., 2009)
Maize cobs	Acid Red 73	(Elizalde-Gonzalez et al., 2008)
Sawdust	Reactive red 24	(Lim et al., 2003)
Rice husk	Reactive Orange 16	(Low and Lee, 1997)
	Basic Blue 3	
	Methyl orange	
	Reactive Blue 19	
	Reactive Red 4	
	Reactive blue 2	

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HIGHLIGHTS

ARISING APPLICATIONS OF BIOCHAR-BASED MATERIALS DERIVED FROM LIGNOCELLULOSIC BIOMASS

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Introduction

Recent considerable attention has been given to biomass conversion as a platform for economically producing energy from renewable resources. All these processes of conversion will produce a solid co-product which is called the biochar. Biochar is a carbon-rich and porous material solid product which can be used for a wide range of applications, including soil improvement, remediation and pollution control. The variety of biomasses, including agricultural residues, wood waste resources and forestry residues, and animal manure are considered raw material for biochar production (Amin *et al.*, 2016). Among these, lignocellulosic biomass comprises a large portion of the terrestrial biomass found on the surface of the earth (Das *et al.*, 2020). However, a large fraction of all these lignocellulosic biomasses are often disposed of by burning, which is not restricted to developing countries alone (Anwar *et al.*, 2014). The transformation of lignocellulosic biomass into biochar can provide an alternative way to reuse these waste materials, reduce the carbon emissions and contribute significantly to mitigating climate change (Speratti *et al.*, 2018). Hence, lignocellulosic biomass derived biochar was proclaimed as a miracle cure for global warming.

Biochar has the ability to generate renewable energy from lignocellulosic biomass in an environmentally sustainable manner. Generally, the biochar was applied as fertilizer for improving the soil quality, retaining nutrients and enhancing plant production (Bonanomi *et al.*, 2017; Rawat *et al.*, 2019). The organic matter and nutrients of the biochar can increase the total amount of nitrogen, soil pH, organic carbon and available phosphorus (Bayu *et al.*, 2016; Rawat *et al.*, 2019). The existence of plant nutrients in the biochar and its porous nature as a medium for microorganisms helps to improve soil properties and increase nutrient absorption which were the key reasons to apply biochar in soil. Moreover, biochar carbon is resistant to degradation and thus biochar can retain carbon in soil for thousands of years (Hagab *et al.*, 2016). On that account, biochar is recommended as a soil amendment for the long-term restoration of carbon sinks.

Currently, the use of biochar in the urban landscape and green construction sector is also being explored. Biochar are used for the production of lightweight biochar-based concrete frames, biochar-lime tiles, tile adhesives and plaster. Biochar based composite provides excellent insulation and is capable of maintaining the desired indoor humidity. The composite may prevent the indoor air from being too dry, which is one of the major causes of respiratory problems; it may also prevent condensation on the exterior walls, which could otherwise cause mold formation. The low thermal conductivity, high chemical stability, and low flammability are three of the most desirable properties of biochar that make it suitable as a building material. Low thermal conductivity is caused by the presence on the surface of biochar particles of a wide variety of various nanopores, mesopores, and micropores (Gupta and Kua, 2017; Schmidt, 2013). In addition, biochar may also introduce into polymeric materials, such as epoxy, to modify their properties and to develop composite materials with improved thermal and mechanical properties. Biochar-filled polymer composites are lightweight materials, readily available in various structural types and are known to improve the thermal, mechanical and electrical properties and the ease with which these composites are made by compression molding (Abdul Khalil *et al.*, 2014).

Biomass provides an avenue for bioeconomic sector growth through the biochar production research and development (R&D) activities. The ongoing R&D commitments and increasing of biomass varieties have correspondingly increased the pace of technological change for biochar industry. Malaysia's biomass sector has the ability to fulfill

the three main pillars of sustainable growth, that is, economic, business and social benefits. According to Hassan *et al.* (2019), the world oil production increased by 40%, and palm oil had the largest share of 38.7%, accounting for approximately 179.6 million tons of production annually. In 2019, the total export of palm oil and palm based products in Malaysia was 25.2 million tonnes and generating export earnings of RM67.5 billion. The expansion of the plantation area is expected to go hand in hand to meet the ever growing demand for oil palm products. Malaysia's oil palm plantation areas was capped at about 6.5 million hectares by 2023 (Yusof, 2019). Looking at the current horizon of the palm oil industry, it can be said that there is a great deal of emphasis on sustainability for biochar production in Malaysia.

Conclusions

Biochar is undoubtedly one of the most exciting research areas of the decade, with discoveries and their realistic application rising exponentially year after year. The production of biochar is not limited to merely replace the burning agricultural fields in the developing world. In the modern world, the use of biochar is multiplying: soil enhancer, adsorbent for wastewater, composite material for building sector, as carbon electrodes in energy storage, food packaging, silage or feed supplements. All these applications could be part of more complex cascades. Hence, a precise understanding of the relationships between the biochar's structure and assessment is required in order to achieve the greatest possible for the biochar application.

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INTROPica HIGHLIGHTS

LIGNOCELLULOSIC BIOMASSES BONDED WITH CITRIC ACID

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Introduction

Malaysia is one of the leading agricultural commodity producers in the South-East Asian region and therefore the agricultural wastes are in abundance and readily available (Ozturk *et al.* 2017). The main agri-based wastes that existing in the country are oil palm biomass (in the form of fronds, trunks and fibers), paddy straw, rice husk, banana residues, sugarcane bagasse, coconut husk and pineapple waste (Goh *et al.* 2010). The main agricultural wastes come from the oil palm biomass which accounted for 46,000 kilotonne in the form of fronds and 11,000 kilotonne in the form of trunks. According to Goh *et al.* (2010), in the year of 2007, approximately 880 kilotonne paddy straw and 484 kilotonne rice husks were produced from the replanting of paddy. Banana residues, sugarcane bagasse and coconut husks were accounted for 530 kilotonne, 234 kilotonne and 171 kilotonne, respectively. Pineapple farm, after the extraction of the fruit, has generated 48 kilotonne of pineapple wastes (Goh *et al.* 2010).

Potential of lignocellulosic biomasses

Agricultural residues have displayed great potential in the production of particleboard as they possess suitable anatomical and chemical structures that are needed for particleboard fabrication (Papadopoulous *et al.* 2015). A review by Madurwar (2013) explored the potential application of agricultural waste for construction and building materials such as particleboards, masonry composites, bricks and thermal insulator. In the review, various types agricultural wastes and various technologies have been used in the production of particleboard. The application of agricultural wastes has been verified as a feasible sustainable alternative material for particleboard production and could help to reduce environmental impact. On the other hand, a review compiled by Klimek and Wimmer (2017) revealed that particleboard with acceptable and comparable properties to that of wood-based particleboard could be produced from wood pruning, straw and leaves as well as plant's stalks. Klimek et al. (2016) replaced the conventional used UF resin with methylene diphenyl diisocyanate (MDI) and found that particleboard fabricated using sunflower, topinambour and cup-plant stalks as raw materials have successfully complied with the European standard (EN 312). However, some improvement is needed for straw-based, bagasse-based and husks-based particleboard. Melo et al. (2014) produced particleboard with wood, rice husk and bamboo and reported that bamboo offers good properties to the resultant particleboard while rice husk showed the opposite results. Incorporation of certain proportion of other woody materials is necessary once rise husks are being used. Hafezi et al. (2016) reported that amino silane coupling agent (SiNH) is required to improve the mechanical properties of the UF-bonded wheat straw particleboard.

Citric acid as green binder

On the other hand, declining of non-renewable fossil resources is anticipated to restricted the usage of conventional synthetic resins in the near future. In addition, conventional synthetic resins such as urea formaldehyde (UF) resin release formaldehyde that are detrimental to human health. Several countries in the world have already imposed stringent regulation in limiting the formaldehyde emission from the wood-based panels. Non-formaldehyde based resin or green binder are therefore in high demand. On account to that, citric acid (CA), also called 2-hydroxy-1,2,3-propanetricarboxylic acid, is potentially to be applied as a green binding agent to produce particleboard with no formaldehyde emission.

Citric acid (CA) is an organic acid that exists naturally in fruits and vegetables. The formula of CA is $C_6H_8O_7$. Citrus fruits such as lemons and limes are known to contain higher amount of CA (Penniston et al. 2008). CA is a versatile substance that are being actively used in many areas (Cirminna et al. 2017). In the recent years, it has been identified to be able to serve as modifying agent and green binder for wood and wood composites (Lee et al. 2020). The formation of ester linkages as a result of the reaction between carboxyl groups of CA and hydroxyl groups of wood constituents are said to bring adhesivity and good bonding properties (Umemura et al. 2012). Furthermore, in a study by Del Menezzi et al. 2018, it was confirmed that the hydroxyl group on the aliphatic chains of lignin units could be react with CA too. Therefore, application of CA as wood binding agent is feasible.

Composite bonded with citric acid

Lignocellulosic biomasses have great potential to be converted into value-added composites. Four types of wood composites including wood-based molding, particleboard, fibreboard and veneer-based panels has been fabricated using CA as main binding agent. The type of lignocellulosic materials used and the composites products bonded with CA are listed in Table 1.

Conclusion

CA has been proven to be able to serve as binding agent for various types of lignocellulosic biomasses. However, future study should be focused on reducing the pressing temperature and time to increase its competency compared to the conventional binder. In addition, a more economical substrate or processing pathway is needed for the synthesis of CA in order to counter the fluctuate price of CA in the global market.

Table 1. Types materials source and products produced by using citric acid as bonding agent (Lee *et al.* 2020)

Type of lignocellulosic materials	Composite products	Reference
Acacia mangium bark powder	Wood based molding	Umemura *et al.* 2012
Bamboo materials	Particleboard	Widyorini *et al.* 2016a
Teak	Particleboard	Widyorini *et al.* 2016b
Petung *(Dendrocalamus asper)*	Particleboard	Widyorini *et al.* 2017
Sweet sorghum bagasse	Particleboard	Kusumah *et al.* 2016
Nipa fronds	Particleboard	Santoso *et al.* 2019
alang-alang *(Imperata cylindrica)*	Particleboard	Syamani *et al.* 2018
New Giant Reed *(Arundo Donax L.)*	Particleboard	Ferrandez-Garcia *et al.* 2019
Sugarcane bagasse	Particleboard	Liao *et al.* 2016
Pineapple *(Ananas comosus* (L.) Merr.) leaves	Medium density fiberboard	Indrayani *et al.* 2015
Elephant dung fibers	Fiberboard	Widyorini *et al.* 2018
Poplar *(Populus tomentosa Carr)*	Plywood	Zhao *et al.* 2019
Poplar Veneer	Wood veneer panels	Del Menezzi *et al.* 2018

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INTROPica
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LIGNOCELLULOSIC MATERIAL: SUSTAINABLE MATERIAL FOR FUTURE DEVELOPMENT

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Introduction

In the era of sustainable development, green material becomes an essential topic. Green materials are environmentally responsible material. Their characteristic is environmentally friendly, biodegradable, and renewable. Lignocellulosic material is one of the green materials that can be found on the earth. It is a sustainable and carbon neutral material compare with the materials derived from petrochemical or refined from ore (Nanda et al. 2015). Lignocellulosic material could be strange for non-science people, but it is also commonly known as plant biomass. Lignocellulosic material is divided into the wood and non-wood materials. Wood materials are derived from the wood tree and non-wood materials are derived from the non-woody plant such as bamboo, rattan, kenaf and others.

Conventionally, lignocellulosic materials were widely used in structural products such as furniture, composite board, building beam, paperboard and others. Besides than structural products, lignocellulosic materials were also traditionally used as raw materials to produce charcoal and later the pellet fuel. Lignocellulosic materials always are one of the significant economic sectors in many countries, including Malaysia. The export of Malaysia wood-based product is reached to Rm22.5 billion in the year 2019 (MTC, 2020) and the industries are still growing.

Nowadays, the application of lignocellulosic material has expanded beyond than conventional wood-based industries. The application of lignocellulosic materials has been leveraged into many other high technology industries (Olatunji et al, 2020). For example, maize is currently widely used as feedstock for biofuel production (Choudhary et al. 2019). The lignocellulosic materials majorly consist of cellulose and hemicelluloses, and some starch compounds. These carbohydrate compounds can be turned down into reducing sugar through the hydrolysis process and fermented into bioethanol. Besides, the unique morphological characteristic of lignocellulosic materials surface allows the biomass such as bamboo turn into activated carbon through pyrolysis (Nor et al, 2013). The application of activated can be found in pharmaceutical industries. Also, the fiber from wood or agricultural products can be dissolved and spun into rayon for textile industries (Chen et al, 2016).

Environment concern

Inconsolably, the public often relates the lignocellulosic materials as the root cause of greenhouse gases. For example, wood is one of the essential lignocellulosic materials. Timber harvesting from forest always received the condemnation from media and organization as one of the main factors to the climate change (Lordan et al, 2018). Consequently, public turn to over reacts and against the wood-based product. The public is afraid that the support of the wood product will eventually lead to more deforestation or forest degrading. This concern is partially correct. This disaster will happen if forest harvesting in an uncontrolled situation.

However, lignocellulosic materials, including wood, is known as a carbon sink (Bosch & Hazen, 2013). They are the carbon pools that we can reproduce in a short time (short crop rotation) compared to other carbon sinks such as fossil fuel. Technically, the lignocellulosic material will remain as a carbon sink as long as the material did not deteriorate or burn to turn into carbon sources. The mature tree has limited capacity in absorbing carbon in atmospheric (Jiang et al, 2020). The sustainable forest harvesting will remove the mature tree, and the vacated space will be reintroduced with a juvenile tree through replantation. This sustainable practice will allow this juvenile tree to absorb a large quantity of carbon gases from the atmosphere through the photosynthesis. This system can help in reducing greenhouse gases available in the atmosphere.

For other applications that require to turn carbon sink to carbon source in order to obtain the output, lignocellulosic materials could probably still one of the best options as it is a neutral carbon material. The carbon cycle of utilizing

lignocellulosic materials in such applications is shorter than many other carbon sink materials such as oil and gases (Prasad et al, 2016). The beauty of lignocellulosic materials as neutral carbon materials should receive more attention.

Future

Lignocellulosic materials have been used as raw materials/-feedstock for many industries. However, the potential of lignocellulosic material still not fully revealed. With the rise of the awareness and the acceptance of lignocellulosic materials as sustainable materials, the application and the demand on lignocellulosic material expecting to be rose. There will be more lignocellulosic conversion value-added products in the future.

However, with the growth of the world population, the lignocellulosic materials supply from the sustainable forest could be insufficient in future (Payn et al, 2015). The agricultural biomass literally will become the future of the lignocellulosic materials. The agricultural-based lignocellulosic material, especially the byproducts from agricultural, can be foreseen as the mainstream lignocellulosic materials for future application without compromise the global food demand from the agricultural sector (Pelletier et al. 2011).

Conclusions

For sustainable development, lignocellulosic materials as green materials should be highlighted. The public perspective of lignocellulosic materials towards the environment needs to be changed. Industries should give the lignocellulosic materials more attention, and the public should widely accept lignocellulosic-based products.

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INTROPica
HIGHLIGHTS

MECHANOCHEMICAL REACTIONS IN LIGNOCELLULOSIC MATERIAL: A REVIEW

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Introduction

Mechanochemistry is a discipline that examines mechanical energy-induced chemical changes in a substance (Beyer and Clausen-Schaumann 2005). However, the mechanochemical reactions of lignocellulosic entities have always been disregarded in the application of mechanical treatments, e.g. grinding, milling and ultrasound, even though it has already been widely acknowledged as means of chemical activation in solid-state physics and polymer chemistry since the past decade (Boldyreva et al. 2013). Therefore, alteration of morphology and chemistry composition should be addressed to which mechanical treatment may influence the formation of mechano-radicals that engage in multiple secondary reactions with one another. Theoretically at least, providing the ability to establish simplistic and sustainable chemical processes, mechanochemistry has the potential to encourage minimal use, if any, potentially harmful chemicals in cellulose fibrillation. This review offers an overview as to how mechanochemistry affects the fibrillation, ensuring a far broader understanding of the mechanism and its effects entailed.

Mechano-radical formations in cellulose

The formation of mechano-radicals in lignocellulosic materials is affirmed using Electron Spin Resonance (ESR) technique. The reactions are focused primarily on the dissociation of carbon-oxygen, carbon-carbon, as well as other chemical bonding of which is attributed to the exposure of applied mechanical stress on the semi-crystalline samples to go through non-homogeneous deformation. Only if shear force (mechanical energy) exceeds the intramolecular forces that keeping the celluloses glued together, it causes homolytic and/or heterolytic rupture to the cellulose chain to create free radical sites, especially in amorphous regions (Solala 2015). Until any mechanical operation, no ESR signals should be observed. These radicals are highly reactive that engage in several chemical transformations, including disproportionation, hydrogen abstraction, and radical rearrangements (Solala et al. 2012).

The formation of mechano-radicals causes the chain breakage of the cellulose backbone, as illustrated in **Fig. 1**, and this is likely to take place in the mid-chain of cellulose than at the chain ends (Solala 2015). Crosslinks or joints between the cellulose molecules are known for high energy sites for chain rupture. The radicals behavior in mechanically degraded cellulose molecules relies upon the ambient conditions in which they are formed, especially temperature and oxygen attendance. Mechanochemical activation can facilitate various forms of bond cleavage; radical formation, ring breaking, and chain-breaking, as shown in **Fig. 2.** The mechanical activation is expected to initiate ring and chain bond cleavages, while the radicals-induced bond cleavage is attributed to the interaction between the free radicals with the cellulose molecules.

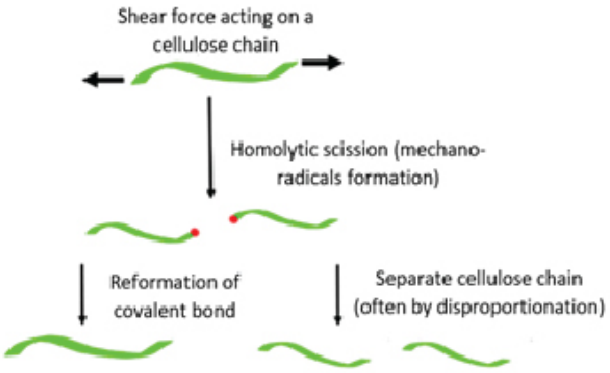

Figure 1: Schematic presentation of mechano-radicals formation of the cellulose chain once subjected to shear force. The mechano-radicals are formed by homolytic chain scission. They typically react fast, either forming new covalent bonds or by undergoing other reactions, e.g. disproportionation that leaves the fractured cellulose chains separate (Solala et al. 2019).

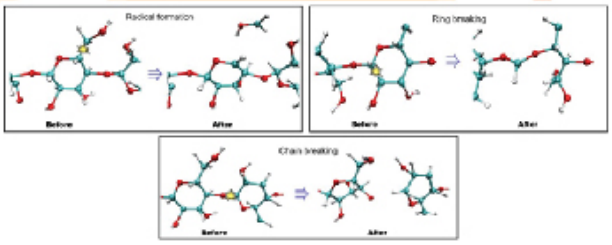

Figure 2: Bond cleavages in the cellulose chain by mechanical activation (Solala 2015).

The dissociation of glycosidic bonds (depolymerization) creates two radical sites in the cellulose molecules. The previous report indicates, during the mechanical treatment, milling, in particular, at least three mechano-radicals have been produced in cellulose fiber; singlet, doublet, and triplet signals. In the ESR results, a singlet signal was produced by alkoxy radicals ($R_{cell}—C_4—O•$), whereas double and triple signals were created by carbon radicals positioned respectively in the C_1 and $C_2—C_3$ locations respectively. Both doublet and triplet signals were capable of reacting with oxygen molecules to produce peroxy radicals ($R_{cell}—C_1—O—O•$), which the singlet signal was incapable of reacting with. The glycoside connexions can be broken in two varying ways, under the impact of mechanical stresses, as seen in **Eq. (1)** and **(2)**, resulting in alkoxyl and alkyl radicals. The alkoxy radicals of $•C_4—Rcell$ is expected to interfere with protons at C4, C3, and C5 positions of a fresh cellulose segment. As denoted in **Eq. (2)**, $•O—C_4—R_{cell}$ and $R_{cell}—C_1•$ are considered to represent the singlet and doublet signals respectively. These reactive radicals induce breakage of the cellulose chain as it may interfere with carbon-carbon bonds along the cellulose backbone to cause chain scission. During a reaction, this unwanted side-reaction is constantly regenerated, referred to as chain propagation, to form new radicals by hydrogen abstraction. Whenever the $C_2—C_3$ bonds are ruptured, the resulting mechano-radicals would exhibit as the triplet signals. There is also the likelihood of dehydrogenation at C_2, C_3, C_4, and C_6 positions to generate free radicals as triplet signals, yet it is unlikely to take place under mechanical stress.

$R_{cell}—C_1—O—C_4—R_{cell} \rightarrow R_{cell}—C_1—O• + •C_4—R_{cell}$ **(Eq. 1)**

$R_{cell}—C_1—O—C_4—R_{cell} \rightarrow R_{cell}—C_1• + •O—C_4—R_{cell}$ **(Eq. 2)**

*Where, R_{cell} is a cellulose molecule

Effect of mechano-radicals formation in cellulose

The fractionation results found that the unbleached pulp has been more fibrillated than its bleached equivalent and its suspension resulted in less fibrous aggregation (Solala *et al*. 2012). This correlates to the density of charge, where the unbleached pulp was about twice as high as the fully-bleached pulp attributed to the loss of hemicelluloses, glucuronoxylans in particular, whose carboxyl groups are the main contributor to the charge in the pulps (Bhardwaj *et al*. 2004). The charge density is attributable to the swelling and resilience of the fibers in the suspension and often influences the ease of mechanical processing (Laine and Stenius 1997). It was also affirmed when the cellulose fiber was milled in the presence of nitrogen or oxygen, the degree of polymerization reduction was noticed to be higher in the presence of oxygen compared to nitrogen (Solala *et al*. 2012). The probable explanation would be that the derived mechano-radicals were instantly scavenged by oxygen molecules to minimize the risk of radical recombination, thereby elevating the depolymerization activity.

As for the unrefined (non-fibrillated) sample, the radical content is explained by their different chemical histories; phenoxy radicals are formed in the bleaching stage with chlorine dioxide (Dence 1996; Tarvo *et al*. 2009) and hydrogen peroxide (Lanchenal 1996). For fibrillated sample, the free radical content goes hand in hand with the increasing degree of fibrillation and thermal activation. That being said, during mechanical fibrillation the maximum temperature was about 60-80 ^{o}C, so the radical formation is unlikely to take place considering the thermal alteration of cellulose in wood takes place over many hours at about 180-250 ^{o}C (Solala *et al*. 2012).

To determine the concentration of the free radicals in samples was Electron Spin Resonance (ESR) spectroscopy, where it detects those unpaired electrons (radicals) through magnetic field interaction. EPR tests are usually taken around a week after mechanical treatment to allow stabilization of the free radicals formed as a result of initial rapid decay. This is important to not contribute any significant extent to the radical content (Iller *et al*. 2002). Both bleaching and mechanical degrees subjected to the pulp influence the free radical content in the samples. The spectra of unbleached pulp revealed a significant singlet signal, which signifies alkoxy radical (R–O•) formed as a

result of glycosidic bonds rupture between C_1 and C_4. Usually, those cellulose-derived free radicals may react with the residual lignin to form very stable phenoxy radicals. In the absence of lignin, these free radicals are more likely to react rapidly with oxygen molecules to form stabilized phenoxy radicals ($R_{cell}—C_1—O—O•$), leading to a higher radical content in the fully bleached pulps.

The temperature during the milling has also greatly influenced the formation of the mechano-radicals, whereby a distinct singlet signal was detected from the sample milled at higher temperatures compared to ambient temperature. This variation in the reactivity of the mechano-radical's reaction may be related to the poor diffusion of oxygen in cellulose fiber at certain low temperatures. Research by Stefanovic *et al.* (2014) compared cryo-milling with room temperature milling of lignocellulosic materials, whereby the findings revealed that amorphous regions are preferentially disrupted at room temperature.

The presence of lignin in the pulp promotes fibrillation owing to the radical scavenging potential (Dizhbite et al. 2004). Having that said, in the absence of lignin (or if present in traces), more cellulose radicals could engage in the recombination reactions that occur between cellulose radicals, resulting in crosslinking that counteracts the fibrillation.

Conclusion

Mechanical treatment of pulp creates mechano-radicals, and this illuminates the previous findings that both unbleached and completely bleached pulps have dissimilar fibrillation efficiency. Not only lignin but also hemicelluloses are extracted by bleaching which eventually affects both charge density and fiber structure, which likely to demote the pulp fibrillation. It should be noted that the highly reactive radicals formed quickly become different radical species or react with each other in radical recombination reactions.

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HIGHLIGHTS

WOOD TO SOLVENT: BIOCONVERSION OF OIL PALM MESOCARP FIBRE TO ACETOIN

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Introduction

Biomass can be classified as low-cost nutrient source available in a large amount from the specific industry such as corn steep liquid from the corn wet-milling industry. Its potential as sustainable raw materials for fermentable sugar (Ren and Sun, 2010). In the native form of the biomass materials, the efficient degradation of lignocellulosic biomass through the enzymatic process is influenced by the presence of firm structure of lignin and hemicellulose linked to cellulose polymer (Ren and Sun, 2010). The presence of lignin and hemicellulose components are the major barrier to compensate for the total usage of the cellulose. It will limit the accessibility of enzymatic hydrolysis and prevent the degradation of the lignocellulosic materials for sugar production (Ren and Sun, 2010). Hemicellulose is one of the elements of the cell walls of higher plants. It is a building component with different level of proteins and phenolics, and most complex components in the cell walls especially in woods and annual plants (cereals and grasses). The hemicellulose composition in different types of biomass varies from one another such as sugarcane rind (22%); sunflower husk (23%); rice straw (25%), oat straw (27%); wheat straw (32%) and corn cobs (37%) (Ren and Sun, 2010).

The palm oil industry is one of the major industries in the Southeast Asian region with Malaysia as one of the biggest palm oil exporters in the world (Mazaheri et al., 2010). In the palm oil processing mill, aggregate residuals of oil palm biomass were produced including liquid (palm oil mill effluent) and solid waste (biomass). About 182 million dry tons of oil palm biomass has been produced since 2011, and that is expected to increase to about 230 million tons by 2020 (Lee and Ofori-Boateng 2013). Oil palm biomass consists of oil palm empty fruit bunch (OPEFB), oil palm mesocarp fibre (OPMF), and palm kernel shell (PKS). About 20% of the biomass, mainly OPEFB, is utilized for nutrient recycling and is retained at the plantation. Another 40% (OPMF and PKS) is burnt for steam generation, but the remaining 40% is not effectively utilized. OPMF covers about 15% of the residuals produced in the mill (Lee and Ofori-Boateng 2013). In total, about 40% of OPMF is burnt for power generation, unfortunately, the remaining OPMF is not effectively utilized (Lee and Ofori-Boateng 2013). In addition, the power generated from the burning process is more than sufficient for the mill itself.

The production of acetoin from lignocellulosic feedstock through fermentation has attracted great attention due to the availability of a variety of substrates, including lignocellulosic biomass, effluent and oil palm fronds from the palm oil industry (Shamsudin et al., 2012) and eucalyptus wood chips from the paper industry (Lee et al., 2010), just to name two. The utilization of the feedstock as a substrate is also important to industry playmakers due to the low cost of raw materials and operation . Acetoin is a volatile and bioactive molecule that naturally exists in some fruits, plants and fermented products. It is classified as a bio-based platform chemical and widely used in the food and cosmetic industry as a flavour enhancer and fragrance agent (Bae et al., 2016) respectively. Acetoin has reported as potential building-block for producing hydrocarbon fuels (Zhu et al., 2016) and it is one of the priority chemical declared by U.S. Department of Energy on the development and its utilization (Sun et al., 2012). Current commercial acetoin has been produced through chemical synthesis using petroleum-based substrate (Dai et al., 2015). However, this process is imposed on environmental impact. Moreover, due to the depletion of fossil fuels and fluctuation of the price of crude oil, alternative renewable resources are tremendously required to fulfil market demands. Acetoin produced from natural sources becomes consumers' preference especially in the food and cosmetic industry. Unfortunately, natural-based acetoin is more expensive than the current product (Xiao & Lu, 2014). Thus, an appropriate substrate is important to synthesis acetoin in satisfactory quantity and quality.

Pretreatment of OPMF and its application

OPMF is a fibrous material and classified as lignocellulosic materials that mainly composed of polymeric components of cellulose, hemicellulose and lignin (Zakaria et al., 2014). The cellulose plays major support to the mechanical strength and exists as a framework of the cell wall in most of the plant cell (Bhatt and Shilpa, 2015). The dissolution of the β-(1,4)-glycosidic will form a monosaccharide of hexose sugars mainly glucose. Pretreatment was introduced to interrupt the robust structure of lignocellulosic components and disrupt native recalcitrant of biomass makes the carbohydrate-based components assessable to enzymes reaction (Hendriks and Zeeman 2009). The appropriate pretreatment is important to relieve the glycosidic bonds and expose complex carbohydrate structure to facilitate the biochemical conversion of lignocellulose components into the sugar platform product. Breakdown of lignin seal, along with high sugars recovery could be achieved by the hydrothermal pretreatment (Zakaria et al., 2015). The intensity of hydrothermal pretreatment contributed by variation of reaction temperature and retention time that commonly measures as pretreatment severity Log Ro (Zakaria et al., 2015). Besides, pretreatment also responsible to reduce the degree of cellulose crystallinity, increase porosity volume as well as surface area (Kumagai et al., 2015).

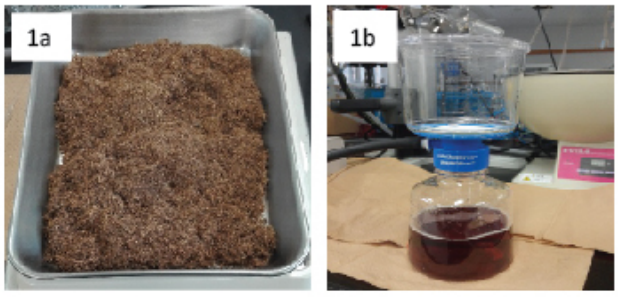

Figure 1: Raw oil palm mesocarp fibre (1a) was undergone pretreatment to produce OPMF hydrolysate (1b).

Monomeric sugars and by-products from OPMF

Hydrothermal treatment was performed using hot compressed water (HCW). It is known as a chemical-free approach since only water was used during the pretreatment process. There are a few reports that have been discussed on the pretreatment severities and its efficiency (Hideno et al., 2009). However, considering the specification of the apparatus, operational parameters and pretreatment conditions would give the different outcome of pretreated products. As a result, the chemical composition changes in pretreated OPMF (Table 1). Under a higher degree of pretreatment severities, a high percentage of cellulose composition was obtained which is in parallel to the dissolution of hemicellulose. Higher pretreatment severity resulted in disarray and destruction of hemicellulose and lignin that present as a protective covering to the complex lignocellulose carbohydrate. However, under high-temperature exposure, the condensation of lignin leads to the accumulation of pseudo–lignin (acid-insoluble polymer). This occasion would increase the amount of klason lignin (acid-insoluble lignin) that deposited in the solid part, especially higher pretreatment severity. The increment of klason lignin was due to the relocation and deposition of lignin components in the lignocellulosic fibres during hydrothermal treatment. This phenomenon has been observed and discussed elsewhere(Xiao et al., 2013). Besides, the fractionation and solubilization of lignocellulose component via hydrothermal intensity led in the decrement of solid recovery and resulted in the accumulation of by-product in the pretreated liquid (Fig 1b).

Table 1 Chemical compositions and components determined from raw, pretreated solid, pretreated liquid and hydrolysate of oil palm mesocarp fibre using HPLC.

Severity factor (log RO)	0	3.66	3.94	4.25
Condition	(Raw)	(180ºC, 20 min)	(190ºC, 20 min)	(200ºC, 20 min)
Cellulose (w/w %)	21.9 ± 1.0	29.2 ± 0.7	30.6 ± 0.9	31.8 ± 0.9
Hemicellulose (w/w %)	22.1 ± 1.0	12.0 ± 0.1	4.9 ± 0.6	3.5 ± 0.1
Klason lignin (w/w %)	36.0 ± 3.5	36.4 ± 3.7	33.4 ± 1.5	34.4 ± 1.8
Solid yield (%)	100	69.3	67.2	61.2
Final pH	–	4.4–4.5	4.1–4.3	3.8–4.0

Acetoin production in bioreactor system from OPMF hydrolysate

Concerning on the transition of fossil fuels as raw materials into renewable sources in the 21st century, finding an alternative sustainable substrate would be a great challenge for the industry to remain relevant in term of cost and profit margin (Sheldon, 2011). The huge amount of oil palm biomass available seems relevant to be used as one of the renewable feedstocks for commodity chemicals such as acetoin. The sustainability and the quality of oil palm biomass are proven since each step of palm oil processing were monitored intimately even the fresh fruit bunches have to undergo strict quality checked (Lee and Ofori-Boateng,

2013). Provided, the palm oil milling does not engage with any chemicals substance thus, the OPMF consider clean and safe to be utilized even for the manufacturer of edible products (Lee and Ofori-Boateng, 2013).

For efficient biomass utilization and industrial application, OPMF hydrolysate was utilized as the main carbon source for acetoin production. Under the optimized conditions, the strain produced 382 mg g^{-1} glucose of acetoin after 48h fermentation from 34.5 g L^{-1} glucose and 2.84 g L^{-1} xylose maximally. The theoretical yield was calculated to be 71% considering the carbon sources are contributed by glucose and xylose, simultaneously. The yield is merely lower than the previous study however, the productivity was 2-fold higher. Productivity is almost equal to reagent-grade glucose and xylose. The production rate of acetoin production from OPMF hydrolysate seems lower in comparison with reagent-grade sugar. This is consistent with the growth of the E. coli which maximum growth at stationary phase reached 7 of OD600 in hydrolysate while 10.5 observed from reagent-grade sugar, respectively.

Concluding remarks

The integration of metabolic engineered strain, efficient pretreatment process along with optimized culture condition deliver a significant enhancement in the development of acetoin production from OPMF. The hydrolysate supplemented with a nitrogen source is the highest acetoin production. Under optimized condition, the increment of production rate more than 10-fold to the control without yeast extract at the maximum theoretical yield of 97% of glucose and xylose as carbon sources. The application of low-cost material as nitrogen source could be an option to reduce the overall production cost as shown by corn steep liquid.

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PYROLIGNEOUS ACID AS WOOD PRESERVATIVE

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Introduction

In the world economy, wood plays an important role as a renewable resource. However, wood is subject to deterioration by mold, insects like termites, and wood degrading fungi. To avoid deterioration of wood during storage, manufacturing, transportation or when in service, it needs to be protected. Wood preservation can increase the durability and resistance to organisms that cause deterioration with different chemicals and processes.

Currently there are several commercial wood preservatives which are in the form of oil-borne (creosote and pentachlorophenol) and water-borne (alkaline copper quaternary compounds, copper azole, ammoniacal copper zinc arsenate, copper citrate, and copper). These are typically toxic, not environmental friendly and also not renewable. Due to the public concern, people are looking into more environmental friendly alternative wood preservative. One of the solutions is by introducing wood preservatives derived from plants, which is pyroligneous acid that produced from pyrolysis process of lignocellulosic biomass.

Pyroligneous acid extraction process

Pyroligneous acid is the product produced from high temperature carbonization of lignocellulosic biomass. However, the content of chemical compounds in pyroligneous acid depends on the biomass chemical composition mainly the three major components, i.e. cellulose, hemicellulose and lignin. The interaction of the three major components during pyrolysis show different reactivity depending on the decomposition temperature of each component (Lee et al., 2010; Diem et al., 2005). Interactions between the components and mineral matters that naturally present in lignocellulosic biomass catalyze numerous reactions during pyrolysis and affect the final content of pyroligneous acid (Lee et al., 2011; Bonelli et al., 2001; Liu et al., 2008).

Pyrolysis is the devolatilization of volatile matter in inert medium to produce pyrolytic liquids, solid char and gaseous fuel (Kirubakaran et. al., 2007). The yield and composition of the derived product from pyrolysis reaction are influenced by the heating rates and temperature. Primary wood pyrolysis involves the simultaneous decomposition of cellulose, hemicellulose and lignin in wood into charcoal and volatiles products. The secondary pyrolysis reaction involves the decomposition of products from primary reactions which is initiated when these hot volatiles products come into contact with unpyrolysed sawdust (Loo, 2008).

Physico-chemistry of pyroligneous acid

The chemical composition of pyroligneous acid includes acetic acid, methanol, acetone, wood oils and tars. Pyroligneous acid appears to be a reddish-brown liquid which resembles to the pleasing hue of black tea, beer or wine. The colour may also vary depending on the type of wood used and pyrolysis condition. The pH value of pyroligneous acid is low, ranging from 2 - 3, due to its high amounts (8 - 10 wt %) of volatile acids, mainly acetic acid and formic acid. The strong and pungent odor of pyroligneous acid is mainly due to glacial acetic acid. Its aroma, however, may differ depending on the nature of the raw material. Pyroligneous acid which is produced from the same species of wood and the same condition may create distinctive flavour and composition. In general, the flavour compounds such as phenol derivatives, furan and pyran derivatives, aldehydes and ketones, esters and nitrogenated derivatives are important contributors to the smoke aroma.

Pyroligneous acid consists of two phases, an aqueous phase and a non-aqueous phase. The former is composed of a wide variety of organo-oxygen compounds of low molecular weight such as acetic acid, acetone and methanol. The latter, also known as wood tar or bio-oil contains insoluble organics (mainly aromatics) of high molecular weight such as phenols and derivatives (Loo, 2008). A high extraction temperature will induce a higher acidic value and also a higher specific

gravity. If device other than an earthen kiln is used, for example combustion furnace or iron kiln, the specific gravity will also vary significantly (Lee et al., 2011; Ninomiya et al., 2004). Chemically, pyroligneous acid consists of water, water-soluble organic compounds and water-insoluble organic compounds. It was reported by Demirbas (2000) that pyroligneous acid contains about 50% methanol, 18.5% water, 18% acetone, 7% esters, 6% aldehydes, 0.5% ethyl and small amounts of furfural. Pyroligneous acids may also contain various phenolic compounds, fermentable sugars, furan compounds, organic acids, etc. The degradation of lignin and wood sugars is generally forming phenolic compounds while organic acid are believed to be derived from acetyl groups in the hemicelluloses (Tiilikkala et al., 2010).

Pyroligneous acid against biological degradation

Pyroligneous acid can be used as a fungicide for wood preservative. As reported by Mohan et al. (2007), pyroligneous acid can be used as preservatives for timber in wet condition while Bruce and Highley (1991) proved the effectiveness of pyroligneous acid in controlling the wood decay, *Basidiomycetes*. It could also inhibit the growth of pathogenic fungus, *Alternaria mali*, which is known as the agent of Alternaria blotch of apple plants (Jung, 2007). From the research published by Nakai et al. (2007), it is found that the resistance of wood against brown-rot fungi can be strengthened with pyroligneous acid from Sugi and Acacia sp. wood. Pyroligneous acids from coconut shell and bamboo have been applied as coagulator and antifungal agents in the production of natural rubber sheets (Zhai et al, 2015). Hwang et al. (2005) stated that phenols and guaiacols in the pyroligneous acid had a strong antimicrobial effect against tested microorganisms, but methanol and acetic acid exhibited little or no antimicrobial activity. Methoxyphenol was observed as a major phenolic compound in rubberwood and oil palm trunk pyroligneous acids (Lee et al, 2010). Salim et al. (2014) showed that pyroligneous acid contains guaiacol, cresol, 4-ethyl-2-methoxy-phenol, and 2,6-dimethoxy-phenol, has good antimicrobial activity. Velmurugan et al. 2009 presented that the pyrolysis of bamboo and broad-leaved trees produce acids (mainly acetic acid) and phenols, which have great antifungal properties that is found useful in wood treatment. Salim et al. (2014) also stated that 4-ethyl-2-methoxyphenol, 2, 6-dimethoxyphenol, guaiacol and cresol, in pyroligneous acids showed good antifungal activity. Lee et al. (2010), Yatagai et al. (2002) and Sameshima et al. (2002) run a correlation test of termiticidal activity with acetic acid and phenolic contents in pyroligneous acid produced from charcoal production. The resistance of wood towards termite attack was increased by treating the wood with pyroligneous acid.

Kartal et. al (2004) showed that concentration of phenolic compound increased with the increasing the pyrolysis temperature. Phenols derived from the dehydration of OH-groups in the alkyl chain of the lignin phenylpropane basic unit followed by the cleavage of interaromatic bonds (Nakai et. al, 2007). Wei et al. (2008) focused on the antimicrobial activities of pyroligneous acids from walnut shell collected at three temperature ranges (90 to 150, 150 to 310, and 310 to 550 ºC) using different fungi or bacteria. The antimicrobial activities of pyroligneous acids at 150 to 310 and 310 to 550 ºC were higher than those of pyroligneous acid at 90 to 150 ºC. It is suggested that these strong biological activities are correlated with the high contents of organic acids and phenolic compounds in pyroligneous acid (Zhai et al, 2015). Hwang et al., (2005) reported that the synergistic effects among the compounds in the pyroligneous acid which exhibited antimicrobial properties.

Pyroligneous acids have been applied for various purposes such as in agriculture and veterinary practice (Tiilikkala et al., 2010). In addition, pyroligneous acid has been used as a sterilizing agent, deodorizer, and fertilizer, as in studies by Souza et al. (2012) and Loo et al. (2007). Locally produced pyroligneous acid has been used as a pesticide in countries where the price of other chemicals has been too high for small scale farmers. Pyroligneous acids are utilized as a wood preservative against fungal degradation, as a fungicide against some plant pathogens, as a repellent and insecticide, herbicide and plant growth enhancer and as feed-stuff for pigs which will improve nutrient digestibility and reduce harmful intestinal coliforms (Tiilikkala et al., 2010).

Pyroligneous acid application on wood

The introduction of chemical substances into wood is usually done in order to improve its characteristic and impart new properties. There are few methods to apply pyroligneous acid into wood. Two most common methods are done by non-pressure process or pressure process. The process of dipping is done by immersing the wood in a bath of preservative for a few seconds or few minutes. A similar penetration can also be achieved by spraying and brushing. The

advantage of this dipping process is that it can minimize damage by hand labor. However, for small lots of timber, this process is often not adequate because the process requires more equipment and larger amount of preservative. The most widely used and permanent method for wood preservation is done by pressure process. Pressure process is done by using closed cylinders and applying pressure or vacuum onto the wood that needs to be treated. Pressure process poses more advantage compared to non-pressure process. One of the advantages is pressure process can achieve higher absorption of preservative by the wood. The penetration of preservative is more uniform in this process. Furthermore, the retention and penetration can be controlled by varying the condition or parameters of treatment. Normally this process is adapted to large-scale production. This treatment method is found very useful to protect ties, poles and structural timbers. However, the disadvantage for this pressure process is the high initial capital

Conclusion

Pyroligneous acid being regarded as one of the plant extractions having potential to be used as wood preservatives due to its specific chemical compositions. Temperature plays an important factor in the percentage recovery of pyroligneous acids during the production process. The content of chemical compounds in pyroligneous acid depends on the type of lignocellulosic biomass. Pyroligneous acid has played an integral part in the growth of new-generation preservatives against biological organisms like fungi and pest. The current trend is to diminish or eliminate the use of two common biocides namely arsenic (As) and chromium (Cr) for several wood protection applications because of the potential environmental issues. Pyroligneous acid has been proven to be able to serve as preservatives against biological organisms. However, research on the efficiency of pyroligneous acid application on wood in extending the service life of wood products is still very limited especially on the retention levels of pyroligneous acid in wood using different application techniques.

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HIGHLIGHTS

BIO-BASED COATING FOR FOOD PAPER PACKAGING APPLICATION

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Introduction

Food packaging is used to protect food from environmental contamination and other influences (such as odors, shocks, dust, temperature, light, microorganism and humidity) and it is key to ensure the quality and safety of food while also extending shelf-life and minimizing food losses and wastage (Ribeiro-santos et al., 2017; Narayanan et al., 2017). Robertson, 2014 attributes to four functions of packaging: containment, protection, convenience and communication. Containment, one of the primary and obvious objectives of the package is to contain the product. This is essential for the efficient transportation, storage and distribution of the product. Protection and preservation is the most important among the functions of packaging. Convenience has long been and continues to be among the chief selling attributes of foods and packaging contributes considerably to the convenience factor, in many ways. Communication: the quantity of information printed on a food package has been increased constantly in order to serve the purpose of product and brand identification (Berk, 2009). In the 20th century, many advancements in packaging technology appeared, including intelligent or smart packaging (IOSP) and active packaging (AP). These innovations further improved food quality, food safety and shelf-life (Brody et al., 2008).

Important of packaging paper

Paper is an excellent material for various applications such as packaging, printing/writing, microfluidic, and household products due to its biodegradability, lightweight, good mechanical properties, and recyclability. Paper-based packaging appears well positioned for a continued upward trend in a global market driven by sustainability agenda. Paper with its environmental benefits is desirable for packaging material. However, the hydrophilicity and porosity of the substrate may impose challenges to control and fully understand the adsorption and transport of water, gases, and oils, in particular for packaging purposes. The control over wetting and barrier properties (i.e., the transport of moisture and gases) is highly relevant in order to retain the shape and mechanical properties of the package by limiting the fiber swelling and quality loss of packed goods. These functionalities can be achieved by internal sizing or further treatment of paper with external barrier coatings. Biopolymers as barrier coatings for paper applications have the potential to replace current synthetic coating materials. They are based on renewable resources and offer numerous favorable environmental advantages such as biodegradability, better recyclability, non-toxicity and biocompatibility, compared to the conventional synthetic polymers (Rastogi & Samyn, 2015; Tang et al., 2012).

Figure 1: Packaging's Functions

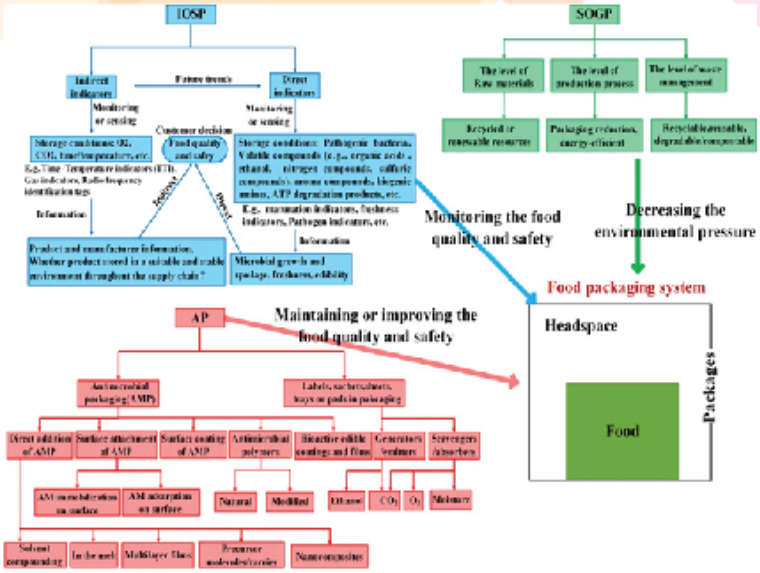

Figure 2: Classification of different types of food packaging system (Han et al., 2018)

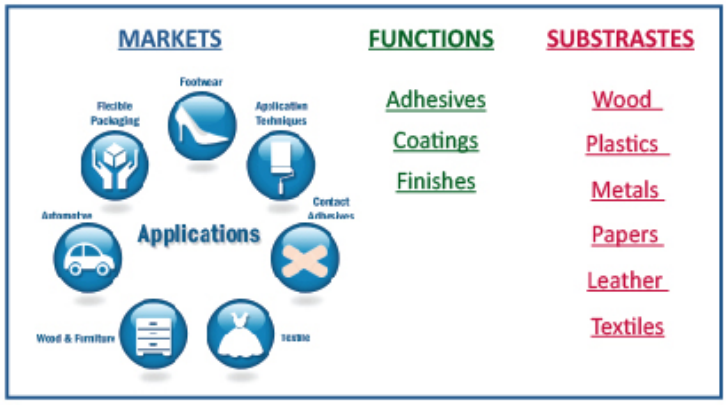

Figure 3: Industrial application of waterborne polyurethane

Waterborne polyurethane (WPU)

Waterborne polyurethane (WPU), a typical resin used in coatings, has become a research hot topic of waterborne coatings because of its advantages, such as non-polluting, safe and reliable, excellent properties, good compatibility and easy modification. WPU, utilizing water instead of organic solvent as a dispersion medium, will not cause environmental pollution because of free pollution or no solvent odor and complies with the requirements of environmental protection regulations (Meng et al., 2020). As only water evaporates during the process, these systems are not harmful to the environment. WPU are important in many industrial applications such as coatings, adhesives, ink binder, glass fibers, paper sizing, synthetic leathers, biomaterials, membranes and films for packaging and water proof textile (Honarkar, 2018).

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CURRENT ISSUE

COVID-19: THE NEW NORMAL IN LABORATORIES AT INSTITUTE OF TROPICAL FORESTRY AND FOREST PRODUCTS, UNIVERSITI PUTRA MALAYSIA

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The Novel Coronavirus found in 2019 also known as the COVID-19 has been declared as a pandemic throughout the world. The virus pretty much affects the whole world in many ways mostly due to the implementation of movement restriction order (MRO). Malaysia has no exception and has announced MRO on 17 March 2020. All sectors including industrial, academic, sports and recreational were put into a halt. Everyone was not ready and has no idea how the MRO could affect their daily lives and working routine. Daily movement routine is restricted, and you can't come to works, except for the essential services listed by the government. The tagline such as #stayathome was used to remind all citizens how serious the situations are. The safety measure such as closing the border, mobilization of full enforcement capacity which involve police, military, civil defense, local authorities etc., routine media update on current pandemic situation in the world and Malaysia and also routine advertisement info on information for self-care to protect COVID-19 infections have making the communities become aware and educate with essential information on COVID-19

At one point during this crisis, the government has declared MRO with slight moderation. Universities could welcome back their staff and postgraduate students to re-establish research works and the laboratories being put into operational again. Stringent standard operating procedure (SOP) could be used as staff and students need to prepare themselves with the new normal. There are several factors of the new normal in laboratories and two (2) of the most important is related to human and environment. The Health Ministry of Malaysia has come out with guidelines such as Avoid 3C and Practice 3W. The 3C were for all to avoid Crowded Place, Confined Space and Close Conversation and then Practice 3W which are Wash your hand frequently, Wear mask if you have symptoms and Warn for close contacts, sneezing and coughing ethics, regular disinfections, stay at home and receive treatment if you have symptoms such as shortness of breath, fever, dry cough or fatigue.Universiti Putra Malaysia (UPM) and all research laboratories were closed during MRO and now is set to be re-open again. Before we start to receive students, we at Institute of Tropical Forestry and Forest Products (INTROP) has developed a plan that includes all the factors that could initiate the spreads of the virus at the lab. During the planning phase, the lab management begin with imagining how the new normal would looks like at the end of the implementations and started to work backward by adding all the necessary factors. The factors include movement control, accessibility between lab, personal protection equipment (PPE), social distancing and cleanliness. On top of that, lab management has been actively collecting info from the Health Ministry and UPM Management so that we did not miss important details and keeping tab with all the requirement outlined. The planning also includes the establishment of staff schedule due to the requirement of only 30% staff could be presence at a time. This could see a large amount of staff will have to work from home (WFH) thus a proper guidelines and monitoring should be established. After the planning phase, the first important thing is to make sure the laboratory is safe and ready for working condition upon returning. Sanitizing the whole building or at the exact location that received most contact with human is required. This could avoid any unknown virus contact that might presence before this and most important is to make sure everybody is coming to the lab with free mind and feel safe. Secondly, we must list all consumables and personal protection equipment (PPE) that is compulsory to be used during the lab activities. This is to make sure the supply for PPE is ready the moment student and customer start to engage with lab activities and sample submission. Besides that, wearing a mask is compulsory for all and for those who are not wearing could be prohibited from

Figure 1: Wear a mask whenever you are in a crowded places inside the building

Figure 2: All movement inside the building must be registered in a logbook

entering the building. Figure 1 shows a staff is wearing a mask during his work in the laboratory. Next, we shall log all movement from all attendees inside the building during working hours. This is because the nature of the laboratories is to conduct research works and services from inside and outside customers. A lot of movement inside and outside of the laboratory will be going on and if there is a positive infection inside the building, the person under investigation (PUI) will be trace down his/her movement inside the building base on the movement log. All close contact person will be listed and reported, and the location visited by PUI must be sanitized. This could be extended towards shutting off the lab operational to prevent further outbreak inside the building. Figure 2 shows a person registering their particulars first before being allowed to enter the building. Finally, the lab consistently commits to educate on social distancing and self-cleanliness. All that is entering the building have to record their body temperature and maintain their distance at least 1 meter from each other as shown in Figure 4. Several locations that has been identified to be congested with people has been mark with necessary floor label to maintain their distance and the location for queue. Figure 5 shows a floor label to locate a safe distance between persons in the lab. All chairs or table inside the meeting room or other room that occupy mass amount of person were also marked as shown in Figure 5. Figure 6 shows the used of hand sanitizer that was placed at all entrance spot and to be used before and after entering the laboratories. Besides that, we encourage all to wash their hand using soap and water frequently too. By all these precaution steps and cooperation of all parties involve, we hope we will win our war with COVID-19 soonest possible even though the vaccine yet to be available. #togetherwefightCOVID-19 #Wemustwin!!.

Figure 3: Each person is compulsory to record their body temperature before entering INTROP

Figure 4: Floor label to maintainsocial distancing

Figure 5: Label on the table to maintain social distancing

Figure 6: The use of hand sanitizerbefore and after entering the building is compulsory

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