Study on Manufacturing of Dissolving Pulp and Mechanical Pulp from Oil Palm Empty Fruit Bunch in Indonesia

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ACRONYMS AND ABBREVIATIONS

AA	Active alkaline
cP	Centi Poise
CPO	Crude palm oil
CSF	Canadian standard freeness
CTMP	Chemi-thermomechanical pulping
DP	Dissolving pulp
E	Alkaline extraction
Ep	Alkaline extraction with hydrogen peroxide
ECF	Elemental chlorine-free
EFB	Empty fruit bunch
IB	Internal bond strength
ISO	International Organization for Standardization
JIS	Japanese Industrial Standards
MDF	Medium density fiberboard
MLH	Mixed light hardwoods
MOE	Modulus of elasticity
MOR	Modulus of rupture
MUF	Melamine urea formaldehyde
O	Oxygen
P	Alkaline peroxide
PHL	The prehydrolysis liquor
PKO	Palm kernel oil
P_{sa}	Peroxymonosulfuric acid

Py-GC/MS	Pyrolysis Gas Chromatography/Mass Spectrometry
SNI Standard	The Indonesian National Standard
Soda-AQ	Soda-anthraquinone
TCF	Totally chlorine free
TMP	Thermomechanical pulping
Z	Ozone

Chapter 1. Introduction

1. Palm oil industry in Indonesia

Indonesia is a tropical country with high rainfall, which encompasses an area of about 1.9 million km2, extending over 5,100 km from west to east. These geographical conditions create great potential for producing agricultural products, with Indonesia currently being one of the main producer of palm oil in the world. In 2013, around 26 million tons of crude palm oil (CPO) was produced[1)]. The oil palm plantations area was estimated approximately 11 million hectares[2)]. This oil can be used by industries, mainly food, for the production of cooking oil, margarine, and ice cream, or by non-food industries for producing candles, soap, shampoo, detergents, lubricants, cosmetics, health related products.

Plantations of oil palm (Elaeis guineensis) in Indonesia are the most prevalent on the Sumatera, Kalimantan, and Sulawesi island resulting in an increase in the income of these communities and total value of Indonesian exports. **Table 1-1**, shows the growth in production of palm oil by province in Indonesia from 2007 to 2012, where overall product increased by more than 50%. Palm oil also represents a strategic industry in other neighboring countries such Malaysia and Thailand, in that it is expected to support national economic growth in the future. Despite these positive impacts, there are environmental concerns related to the processes used both in the palm fields and factories to separate CPO and palm kernel oil (PKO) from fruit, as these generates liquid, solid, and gaseous waste products.

Solid waste from production processes includes shell, fiber, frond, palm kernel cake, and empty fruit bunch (EFB) wastes. Most of the residues are unutilized. EFB is the main by-product of palm oil fruit industry process and is generally left to rot on the plantation site without further processing.

Table 1-1 The production of palm oil by province (thousand ton)[1)]

Province	Crude Palm Oil (CPO)						
	2007 *	**2008**	**2009**	**2010**	**2011**	**2012**	**2013***
Aceh	696.30	564.70	693.00	616.50	592.20	676.98	654.80
Sumatera Utara	3908.90	3 882,4	3 862,4	3899.60	4010.70	4147.65	3975.43
Sumatera Barat	979.20	961.50	896.30	985.90	927.40	960.43	930.12
Riau	4104.00	4 812,9	5 311,4	5496.00	5895.50	6499.82	6384.54
Kepulauan Riau	-	10.60	11.30	14.10	15.20	38.46	37.20
Jambi	1301.50	1 626,5	1 499,9	1644.10	1773.10	1760.35	1718.29
Sumatera Selatan	1751.30	1 891,4	2 313,5	2542.80	2417.70	2552.41	2492.90
Kepulauan Bangka Belitung	314.50	412.90	446.60	490.20	504.60	522.07	504.60
Bengkulu	369.40	560.30	736.00	796.00	827.10	825.76	802.02
Lampung	382.00	416.30	389.30	405.70	424.00	459.73	433.82
DKI Jakarta	-	-	-	-	-	-	-
Jawa Barat	15.60	12.60	20.70	16.40	22.10	24.86	22.72
Banten	37.80	25.00	25.10	26.00	21.80	29.86	27.67
Jawa Tengah	-	-	-	-	-	-	-
DI Yogyakarta	-	-	-	-	-	-	-
Jawa Timur	-	-	-	-	-	-	-
Bali	-	-	-	-	-	-	-
Nusa Tenggara Barat	-	-	-	-	-	-	-
Nusa Tenggara Timur	-	-	-	-	-	-	-
Kalimantan Barat	1057.20	1 124,4	1 331,7	1426.90	1443.90	1942.15	1830.86
Kalimantan Tengah	703.10	1 295,7	1 798,1	1724.70	2499.30	3055.05	2958.28
Kalimantan Selatan	433.10	891.10	1 041,4	1049.20	1045.20	1279.65	1233.08
Kalimantan Timur	297.10	338.40	456.40	700.00	905.50	1393.72	1298.14
Sulawesi Utara	-	-	-	-	-	-	-
Gorontalo	-	-	-	-	-	-	-
Sulawesi Tengah	112.10	126.60	144.30	145.80	185.10	237.53	234.08
Sulawesi Selatan	83.70	21.50	28.20	34.90	42.70	48.91	47.18
Sulawesi Barat	216.00	325.80	260.50	264.40	249.30	257.50	253.73
Sulawesi Tenggara	10.30	10.60	0.00	15.20	16.20	30.03	28.86
Maluku	-	-	-	-	-	-	-
Maluku Utara	-	-	-	-	-	-	-
Papua	46.30	49.50	66.70	136.40	103.10	97.70	93.48
Papua Barat	71.10	40.00	57.70	66.20	54.10	54.83	53.72
Indonesia	**16890.50**	**19 400,8**	**21390.50**	**22496.90**	**23975.70**	**26895.45**	**26015.52**

Catatan : *provisional figures

Actions must be taken to prevent and eliminate the negative impacts of these wastes. One such solution is to turn the waste into secondary product, which could also simultaneously increase the positive impacts of the palm oil fruit industry. The potential availability and economics of deploy agro-industry residues is highly interesting despite the limited practical implementation[3]. Rosli and Law[4] reported that the fiber of the oil palm stem, fronds, and fruit bunches is a good raw material for paper production.

Compared to woody materials, non-wood raw materials have a similar cellulose content, and have lower lignin and higher pentosan (hemicelluloses) and silica content[5]. Non-wood materials offer some advantages for utilization in the pulp and paper process (compared to woody materials) such as the relatively inexpensive nature of most agricultural by-products (residues) compared to woody materials. However, non-wood materials also suffer from certain disadvantages, such as greater bulk of the material, longer preparation required for the raw material, and a higher dust load.

Production of pulp for paper making from non-wood fibrous raw materials has continued to grow, driven by the increasing demand for paper and paper products and the insufficient wood supply[6]. Mossello et al.[7] projected that non-wood fiber, as an alternative fiber for cellulosic plant materials for papermaking, will increase as the world population increases.

The optimal conditions required to reach an acceptable standard has not yet to be determined and requires further study. Compared to kraft cooking, soda-anthraquinone (AQ) cooking, as a sulfur-free cooking method, is preferable and is more suitable for cooking non-woody materials from the perspective of environmental concerns. The cooking performance of soda-AQ is comparable to that of kraft cooking[8]. Research on the optimal conditions required for the production of dissolving pulp and paper from EFB through chemical and mechanical pulping is thus indispensable.

2. Objective

The purpose of this research is to improve the utilization of waste from the palm oil industry (especially EFB) for the manufacturing of derivative products to support sustainable development of the palm oil industry. The utilization of EFB as a derivative product will become an added value for the industry and at the same time reduce the extent of environmental pollution.

3. Literature Review

3.1 Empty Fruit Bunch

Oil palm (*Elaeis guineensis*) is a rapidly growing commodity in the plantation sector. Oil palm is the raw material of crude palm oil (CPO). CPO, as the foremost result of oil palm fruit, can be used in different branches of industry. Some food products using CPO as main ingredient for example cooking oil, margarine, and ice cream. Therefore, in the non-food industry, CPO is used for producing candles, soap, shampoo, detergents, lubricants, cosmetics, and pharmaceutical products.

Physically, empty fruit bunches from oil palm consist of various kinds of fibers composed of cellulose, hemicellulose, and lignin[9)]. However, EFB has certain demerits such as largeness, high moisture content, the thickness of the bunch, and the product is not easily dried due to the high remaining oil content[10)]. Therefore, for EFB handling, especially for the drying and transport processes, size reduction of the material is required.

Photo 1-1 shows the condition of the palm oil plantations in Indonesia, while **photo 1-2** shows the fresh fruit bunch (FFB) after harvesting and before separation and steaming in palm mills (**photo 1-3**). High-temperature steaming as shown in **photo 1-4**, a fruit bunch of about 50 cm in length is harvested from palm oil trees (usually about 10 m height), and is then separated into 3–5 cm long palm fruits and an empty fruit bunch (EFB). This process

invariably leaves residues such as the kernel. shell, fruit fibers, fronds, palm kernel cake, and EFB wastes, most of which are unutilized. Sample preparation in this study was started with the conversion of EFB to short fibers, as can be seen in **photo 1-5**. TMP refiner used in this study are shown in **photo 1-6**.

3.2 Mechanical Pulping

Mechanical pulping is the process of separating wood fiber using mechanical force. This process has a high yield, around 95%, but the resulting paper has lower strength properties, high color reversion, and low brightness.

Figure 1-1 shows a schematic of the process inside the refiner. There are two disks in the refiner; the first is a rotor and the second is called a stator. The gap between two disks is the refining process zone, where the materials are processed gradually before exiting through the outlet.

Many methods are used in mechanical pulping, the initial process is called groundwood process, where the sample is inserted and pressed lengthwise against a rugged rotating grinding stone. Another process, refiner mechanical pulp (RMP), utilizes woodchips that torn into fibers in between the large rotating disk known as refiner. Rusu et al.[11] reported that the main effect of refining is internal and external fibrillation, which increases the flexibility of the fiber, thus expanding the inter-fiber contacts.

Weaker paper of paperboard is one of the results of the damage to the fibers that are separated mechanically. However, this process results in a yield of paper per unit volume of wood which is still greater than that produced by chemical pulping because both lignin and cellulose fibers remain intact. The structure and dimensional nature of chemical and mechanical pulp fibers influence the resulting products, and there are also differences in the surface chemistry of pulp generated chemically and mechanically. Chemical pulp fibers may be more active for sheet formation and consolidation[12].

Photo 1-1 Palm oil plantation

Photo 1-2 Fresh fruit bunch (FFB) after harvesting.

Photo 1-3 Fresh fruit bunch (FFB) before separation and steaming in palm oil mills.

Palm Oil Tree (Productive stem height: 3-15 m)

Fruit Bunch (50-60 cm in length)

Empty Fruit Bunch (EFB)

EFBs at the plantation site (Moisture content 50-60%)

Palm Fruits (3-5 cm)

Fig.1-4 Part of Oil Palm Fruit Bunch

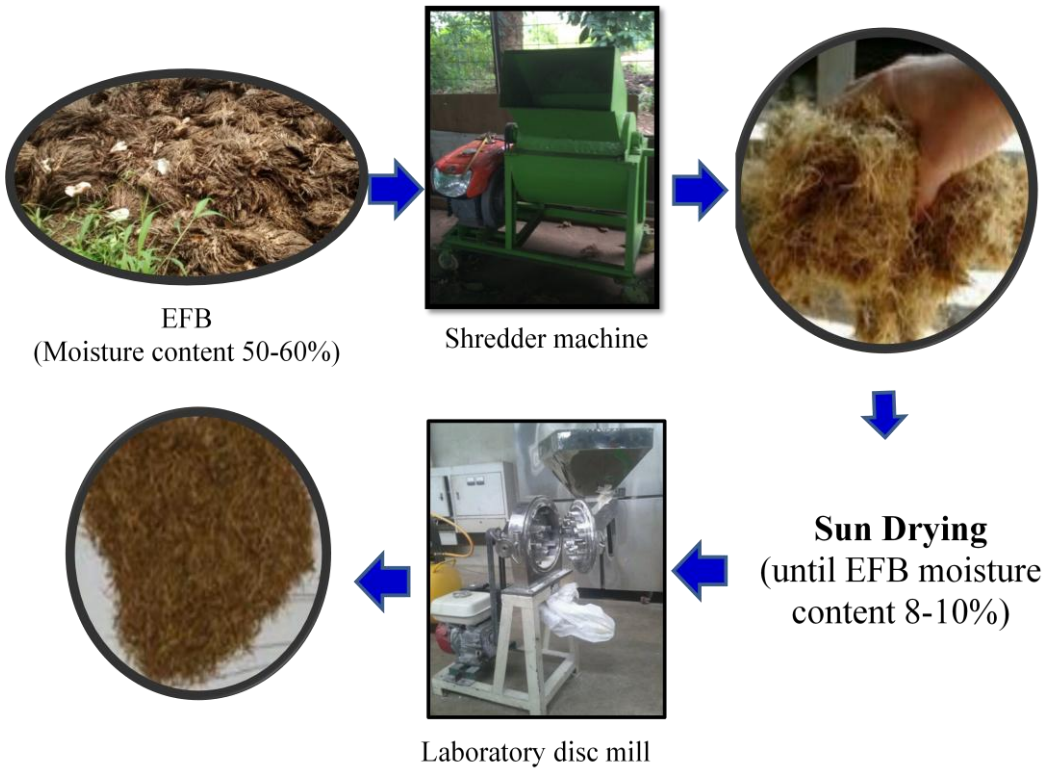

Fig. 1-5 Preparation of EFB fiber for laboratory experiment

Photo 1-6 TMP Refiner

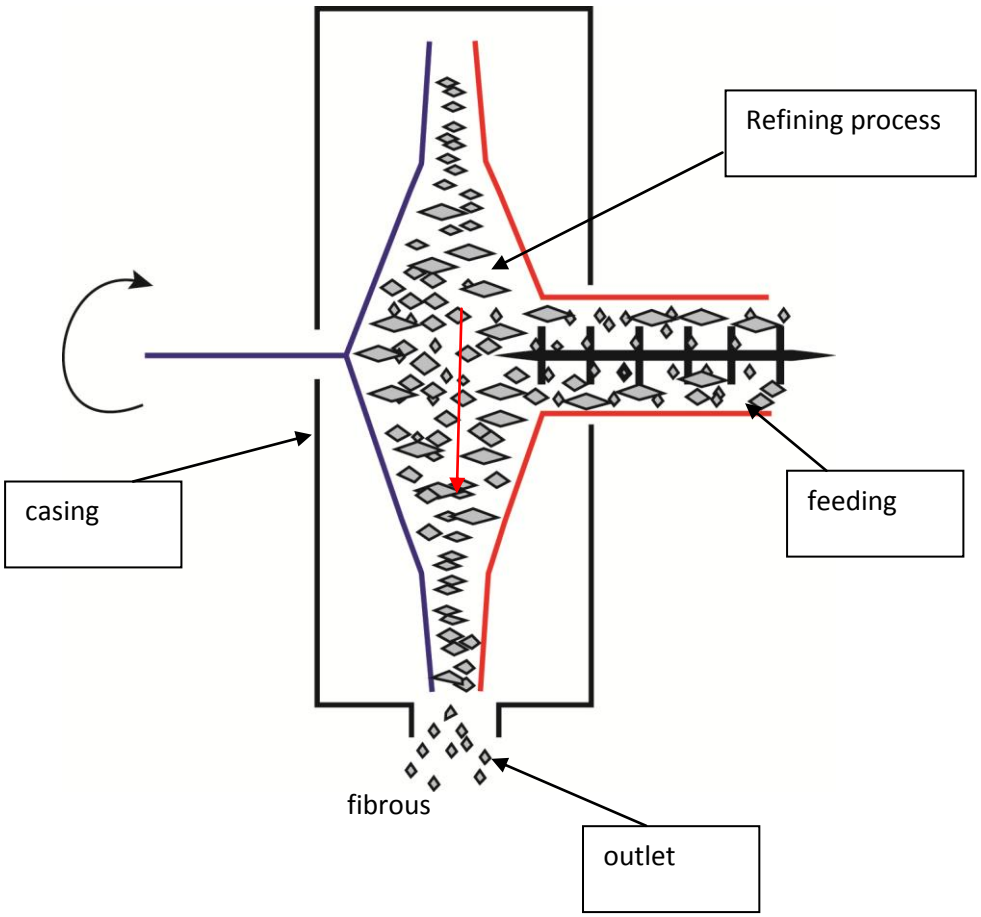

Fig.1-1 The schematic process in the refiner machine.

Thermomechanical pulping method or TMP is a modified version of the refiner mechanical pulping process. Heating wood chips with pressurized steam before they are refined results in softening of the lignin. In chemical thermomechanical pulping, a preliminary treatment was done by using mild sulfite to modify the lignin and achieve partial delignification before the thermomechanical process[13]. The pulp yield decreases slightly to between 85 and 90% with chemical thermomechanical pulping.

Generally, in order increase the papermaking potential of the available fiber post refining of fiber's beating (beating or additional beating) is required for semichemical and chemical pulp before paper making process. The severity of beating depends on the pulp and the paper to be manufactured from it. Beating (refining) has several general effects on the sheet quality, increasing the tensile strength up to a plateau followed by a decline with excessive beating. This result shows the increasing value of tensile strength followed by decrease with prolonged beating; the opacity and porosity of the paper also decrease after refining, and the sheet density increases after beating[14]. Beating is a mechanical treatment that is used in the stock preparation of fiber to improve the quality of the fiber and affects the fiber structure and properties through certain simultaneous changes[15].

3.3 Chemical thermomechanical pulping for paper production

To reduce the disadvantages of the mechanical pulping method, a combination method comprising chemical and mechanical pulping, termed chemical thermomechanical pulping, is used in many paper production processes.

Highly tear resistant paper is generated by "sulfonation" treatment compared with that obtained from the thermomechanical, refiner, or stone-ground pulps. Chemical thermomechanical pulping results in lower pulp yields ranging from 85 to 90%. Chemical

thermomechanical pulping is commonly used for manufacturing printing and writing papers, tissue, and packaging boards.

3.3.1 Chemical pre-treatments

The objective of the chemical pre-treatment processes in chemical thermomechanical pulping is to enhance softening of the wood chips before the refining process. Pretreatments such as physical, hydrothermal, chemical, and biological processes are recommended to augment the pulping and papermaking processes[16]. Thermomechanical pulp (TMP) and chemi-thermomechanical pulp (CTMP) produced from wood block materials that are subjected to axial pre-compression pretreatment may have increased bonding potential and fiber length retention compared with untreated wood chips, as reported by Frazier and Williams[17].

Zhai[18] also reported that NaOH could be used for chemical pretreatment of *Pinus massoniana* in other TMP pulp processes to enhance the fiber bonded area and increase the paper strength. The utilization of 3% NaOH in the refining pretreatment facilitated loosening of the silica bodies from the fibrous structure of the EFB fibers[19].

The advantages of pre-treatment before refining have been reported by Liu[20], who showed that pre-treatment with xylanase improved the tensile, burst and strength of aspen alkaline peroxide mechanical pulp. NaOH pre-treatment has also been utilized in the manufacture of pulp by the CMP pulping method[21].

3.3.2 Mechanical pulping process

In the case of high consistency refining, the pulp usually comes directly from the first stage refiners where the wood chips have just been broken down into mechanical pulp. After refining, the bulk density of the fibers increases and the fibers become shorter.

One potential drawback of the refining process is interruption of the fibers resulting in lower paper strength, whereas this process increases the bulk and improves printability. Refining influences the morphology and the surface chemical composition of pulp[22)], and thus may significantly impact the final paper properties.

The direct refining effect to the fibers may be reduced by shear and press forces. It has been proposed that the amount of impact exerted on the pulp during refining is a crucial factor affecting the refining phenomenon.

3.3.3 Physical properties of paper

Paper usage will be determined based on two main categories of physical properties, i.e., the basic properties and strength properties. The basic properties include the grammage, moisture, thickness, bulk, and density, while the strength properties include the tensile and bursting strength, tearing resistance, and bending resistance. Grammage is defined of paper or board, expressed as g/m^2. Paper and board moisture content will affects the dimensional stability, physical strength, paper runnability, calendering, embossing, and in particular, the printability.

The thickness, bulk, and paper and board density may be measured either for individual sheet or a pad of sheets. Bulking thickness term refers to the final thickness of a book produced with the paper concerned. The bulking thickness refers to the last thickness of a book produced with the paper concerned. Paper density is determined by measuring the weight and thickness (or caliper) of a sheet of paper. This parameter is well known as the area of density, which is defined as mass per volume unit of paper or board, calculated on the basis of the ratio between the materials' weight and thickness in kg/m^3 or g/cm^3.

3.3.4 Strength properties

The tensile strength is the maximum tensile force developed in a test specimen before rupture during a tensile test carried to rupture under prescribed conditions[23]. The bursting strength is the maximum pressure that a paper specimen can withstand without breaking when pressure is applied perpendicular to the plane of the test piece. Bursting strength unit was written in kilopascal, kPa. The burst strength is influenced by many factors in papermaking, such as the fiber type, degree of refining, presence of strength additives (e.g. starch), sheet formation, and moistmoisture content. The tear strength is a measure of the force perpendicular to the plane of the paper required to tear multiple plies through a specified distance using an Elmendorf-type tearing tester[24]. The tear strength is generally affected by the strength properties, for example tensile strength and elongation, as well as by the sum of long and well-bounded fibers. The definition of stiffness is the resistance of paperboard against bending that caused by applied force.

3.4 Chemical pulping

The pulping process can be distinguished into two processes, such as mechanical and chemical pulping. Pulping is the process involving wood or non-wood fibrous feedstocks, as the main raw material, converted into pulp; in this process, fibers are liberated from the initial material in a specific conditions. The can be done chemically or mechanically, or by combining the two treatments. Chemical pulping can be described as a method for wood delignification (or non-wood species) using chemical solution and through chemical degradation, while mechanical pulping utilizes mechanical or physical treatment. The pulp resulting from chemical pulping is often called chemical pulp, referring to the chemical pulping process.

Three major methods in chemical pulping currently known, viz. kraft, sulfite, and soda pulping. Although predominantly used in the papermaking industry, some pulps are processed into various cellulose derivatives and regenerated celluloses.

3.4.1 Kraft pulping

Kraft process or often called kraft pulping, is common utilized pulping method in the pulp and paper industry. The main reasons for the application of kraft pulping compared to other processes, are the excellent pulp strength properties, low demand of wood or wood quality, well-established recovery of cooking chemicals, energy, and by-products, and short cooking time.

In this process, wood chips are cooked with sufficient chemicals in an aqueous solution at adequate temperature and pressure in order to remove and dissolve away the lignin component and maintain most of the celluloses and hemicelluloses. Cooking systems are divided into two main systems: batch and continuous.

3.4.2 Sulfite pulping

Sulfite pulping is generally used only for special purposes given that the yield of sulfite pulp is much lower than that of kraft pulp. In the sulfite process, a solution of sulfurous acid (H_2SO_3) and calcium bisulfite $Ca(HSO_3)_2$ is used to cook wood chips in order to dissolve lignin. The resulting pulp is usually moderately strong, soft, and flexible. The active sulfur-containing species in the sulfite process are sulfur dioxide (SO_2), hydrogen sulfite ions (HSO_3^-), and sulfite ions (SO_3^{2-}) in proportions depending on the actual pH of the cooking liquor.

3.4.3 Soda pulping

About 5 to 10% of non-wood pulp comes from agricultural residues. This is produced by soda pulping. The most available agricultural crops among these are wheat straw and

bagasse. Pulping using the soda process is limited to easily pulp materials such as straw and some hardwoods but is not a primary process [25]. Reduction of carbohydrate degradation of the pulping process can be achieved by using anthraquinone as an additive. The soda-AQ process can also utilize the same mechanical recovery that is used for kraft pulping.

3.5 Production of dissolving pulp

Dissolving pulp (DP) is a pulp with superior pulp cellulose quality that has a very high content of cellulose with low hemicelluloses and lignin content. DP is a main material for viscose rayon, cellophane, chemical additives, and cellulose-based derivatives. Production process is a key factor of DP's purity (cellulose content) that later will determine its end use. For DP, high quality with a degree of purity exceeding 90% of a-cellulose, a relatively low content of hemicellulose, and an extremely low content (less than 0.05%) of lignin are generally required. In order to get a superior quality, removing hemicellulose from the wood fiber is crucial. Several methods have been developed for producing dissolving pulp.

Compared to kraft cooking, soda-anthraquinone (AQ) cooking, as a sulfur-free cooking method, is preferred and is more suitable for cooking non-woody materials based on environmental considerations. Dissolving pulp can be produced through several stages, namely, pre-hydrolysis (as a pre-treatment), cooking, and bleaching.

3.5.1 Pre-hydrolysis

Pre-hydrolysis is an important step in the manufacture of dissolving pulp, particularly for removing hemicellulose. Pre-hydrolysis is performed with water, acid, and alkali. H_2SO_4 is used for acid pre-hydrolysis while NaOH is used for alkali pre-hydrolysis. Water is used for aqueous pre-hydrolysis. The dissolution of hemicellulose occurs in hot water because of the change in uronic acid and *o*-acetyl cleavage during pre-hydrolysis to form various types

of organic acids, including acetic acid, which helps to break the chain hemicellulose into monomers and oligomers.

To obtain similar kappa numbers at similar cooking temperatures, the cooking of prehydrolyzed chips is significantly faster (40% shorter cooking times) than that of unhydrolyzed chips[26].

3.5.2 Cooking

Soda-AQ and kraft cooking provide similar cooking performance[27]. Anthraquinone (AQ) is used to enhance the degradation of lignin and also to reduce the degradation of cellulose in the soda pulping process; this process is termed soda-AQ pulping. Previous studies[28] indicated the potential applicability of dissolving pulp that has been previously hydrolyzed by soda-AQ. Pre-hydrolyzed soda-AQ dissolving pulp was produced from Populus deltoides with a high α-cellulose content, with acceptable final yield, brightness, and viscosity.

The addition of AQ in both kraft and soda pulping at lower cooking temperature (160oC) inhibited the decline in the viscosity without causing a reduction of kappa number[29]. A previous study also indicated that high-quality pulp could be produced from oil palm could be utilized for printing paper application via soda-AQ pulping and TCF bleaching[30].

3.5.3 Bleaching

The objective of the bleaching process is to enhance the brightness of cellulosic materials by application of a chemical process. A cellulosic pulp is generally subjected to bleaching processes as an aqueous suspensions of individual fibers separated from woody or non-woody raw materials, for example, bunch, straw, reed, jute, sugarcane bagasse, bamboo, etc. The absorbance of visible light by wood (or non-wood) fibers is mainly caused by lignin, one of the building materials of wood. The brightness may be increased either by removal or

decolorization of lignin. In the manufacture of chemical pulps, almost all lignin content is removed during cooking, and further lignin removal is achieved during pulp bleaching.

Bleaching chemicals vary in their effectiveness and efficiency, which in turn depend on the wood species, pulping process, and bleaching conditions. The approaches for pulp bleaching can be classified into three groups as follows: elemental chlorine, elemental chlorine-free (ECF), and totally chlorine free (TCF) bleaching. Elemental chlorine pulp bleaching was once widely used by most existing bleaching plants. To brighten the pulp, chlorine (Cl2) and hypochlorite are use in this process.

Currently, hypochlorite is not being used any longer while chlorine dioxide has been introduced to replace chlorine as a bleaching agent in elemental chlorine-free bleaching (ECF). Totally chlorine-free (TCF) bleaching utilizes agents such as oxygen, peracetic acid, ozone, and peroxide without using chlorine. TCF eliminates chlorine related pollutants in the effluent.

Efficient extraction is very important in the first stage of the bleaching process. For example, about 30–50% of lignin is removed before alkaline extraction. Sodium hydroxide is the most efficient agent for reducing the kappa number of pulp. Around 80 to 90% of the lignin is removed after alkaline extraction[31)].

3.5.4 Bleaching chemicals

The main goal of bleaching is to confer greater whiteness and brightness to the pulp through multiple stages or sequences[32)]. The extent of bleaching is determined by the brightness required. This is achieved by using one of a number of bleaching sequences described as CEH, CEDED, CEHDED, etc. C in the sequences stand for chlorine, while E, H, D are stand for sodium hydroxide, hypochlorite, chlorine dioxide, recpectively. Most bleaching techniques involve multi-sequence steps and may include up to six stages in all.

Chlorine

The principal active chemicals in an aqueous chlorine solution consist of elemental chlorine and hypochlorous acid. In bleaching, the relative amounts of these chemicals are mainly determined by the elemental chlorine's concentrations and hydrogen ions.

Chlorine dioxide

Chlorine dioxide is an oxidant that more expensive and stronger per unit of oxidizing power than chlorine. Chlorine dioxide is generally utilized within an acidic condition as well as in tandem with oxygen delignification. There is a limit to lignin removal degree that can be achieved with further oxidation, and another extraction step is often required to reactivate the pulp for the final oxidation stage.

Alkaline extraction

The purpose of the alkaline extraction serves as a lignin removal that has been made potentially soluble in the previous acidic oxidizing stage and activate the pulp again or further oxidation.

There is a limit to the degree of lignin removal that can be achieved with further oxidation, and another extraction step is often required to reactivate the pulp for the final oxidation stage.

Oxygen

Alkaline oxygen delignification (O) is ideal for the kraft process as it exploits oxidation of white liquor as an alkali source and the generated spent liquor is reused in the kraft digester counter wash. Oxygen delignification is complex and involves free radical mechanisms. The treatment removes a notable amount of residual lignin without serious cellulose degradation.

The level of EFB soda-AQ pulp that can be obtained by oxygen bleaching is limited to 34%[33]. Beyond this limit, degradation is accelerated without achieving satisfactory delignification.

Ozone

Ozone is a strong oxidant that readily reacts with lignocelluloses. The most critical stage of the TCF bleaching process for dissolving pulps is ozone treatment, which is important to eliminate residual lignin. Because ozone delignification is accompanied by serious cellulose damage, the amount of ozone utilized during bleaching process should be as low as possible[34].

Hydrogen peroxide

The alkaline hydrogen peroxide stage is commonly utilized in pulp industry to bleach lignin-rich pulp to a brightness level of 80-83% ISO brightness (International Organization for Standardization) without any substantial dissolution of lignin. The hydrogen peroxide used in chemical pulp bleaching has long been limited to the final stages to improve not only the pulp brightness, but also its brightness stability. Hydrogen peroxide posses several advantages including low cost and strong bleaching effect.

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Chapter 2. Preparation of Dissolving Pulp from Oil Palm Empty Fruit Bunch by Prehydrolysis Soda-AQ Cooking Method

1. Introduction

Indonesian oil palm trees are located in rural areas, spread mostly on Sumatera and Kalimantan islands. In recent years, Indonesia has successfully encouraged the expansion of the crop in more remote locations on the islands of, Sulawesi, and Papua. The Ministry of Agriculture of the Republic of Indonesia estimated the oil palm area of 22 provinces to be approximately 11 million ha[1,2)]. The location of the palm oil plantations in Indonesia is shown in **Fig. 2-1**[1,3)].

Palm oil mills separate CPO and palm kernel oil (PKO) as main products, leaving residues such as wastes of shells, fibers, fronds, palm kernel cake, and empty fruit bunch (EFB). EFB and fronds are expected to be further processed into paper and paperboard, while fibers and shells can be utilized as a boiler fuel to generate steam in palm oil mills. However, most of the residues are not utilized, and most of the EFBs are left rotting in the plantation site without further processing. The Food and Agriculture Organization of the United Nations estimated that the global production of CPO generated was 50 million tons per year, in 2012. Southeast Asia is the main contributor, with Indonesia and Malaysia accounting for 47% and 37%, respectively. In the production of one kg of palm oil, approximately 4 kg of dry biomass is produced, of which one-third is EFB. Thus, the potency of EFB materials was about 30 million tons per year in Indonesia.

In 2003, the first pulp and paper mill using oil palm-based materials was set up by the Forest Research Institute Malaysia and a pulp and paper manufacturer (Borneo Advanced Sdn. Bhd.) located in East Malaysia (Sabah). In the paper mill, EFB is converted into pulp using the caustic soda technology[4)]. However, as mentioned before, EFBs have not been

optimally utilized until now. In practice, EFBs have been usually burned in palm oil mill incinerators, causing environmental pollution problems in nearby localities. The moisture content of fresh EFB is usually over 60%; consequently, without drying, it is a poor fuel and presents considerable emission problems[5].

EFB is composed primarily of cellulose, hemicellulose, and lignin as a lignocellulosic material. Previous studies have reported that the amount of cellulose was estimated to be around 30–50%, while hemicellulose and lignin contents were around 15–35% and 20–30%, respectively[6].

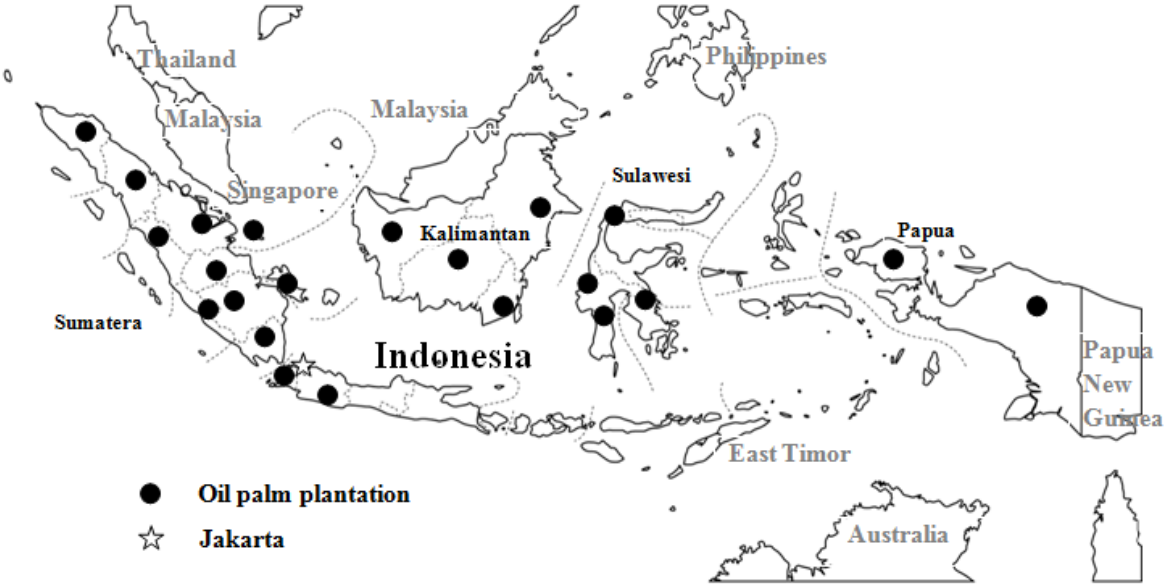

Fig. 2-1 Palm oil plantation in Indonesia[1,3)]

This study is aimed at determining the suitable conditions for preparing DP from oil palm EFB with the combination of prehydrolysis and soda-AQ cooking, followed by elementary chlorine-free (ECF) or TCF bleaching.

Dissolving pulp (DP) is used as a material for viscose rayon, cellophane, and other cellulose derivatives. It requires high-quality α-cellulose with a pure degree (higher than 90%), a relatively low content of hemicellulose, and an extremely low content (less than 0.05%) of lignin. Several methods have been developed for producing DP. Prehydrolysis is one of the most important steps to remove hemicellulose and assist the separation of lignin from raw materials. Compared to kraft cooking, soda-anthraquinone (AQ) cooking as a sulphur-free cooking method is preferable and more suitable to cook non-woody materials, considering the environmental concerns. Andrade and Colodette[7] have reported on production of DP from sugarcane bagasse by adopting these processes. Prehydrolysis followed by the soda cooking process has been proposed as a suitable preparation method for DP from EFB. In previous studies, the best conditions for prehydrolysis were found to be 170°C for 60 min, followed by soda cooking[8]; the optimum variables for soda cooking were statistically analyzed to be 161°C for 100 min with 26.1% of alkali level to attain a pulp with 31.2% of screened yield, 6.0 of kappa number, 96.9% of α-cellulose, 16.1 cP of viscosity, and 0.15% of ash content[9]. Furthemore, total chlorine-free (TCF) bleaching methods were also proposed using oxygen (O) bleaching for EFB soda pulp[10], ozone (Z) bleaching and alkaline peroxide (P) bleaching for the kraft pulp[11]. Although the O–Z–P bleaching for the prehydrolysis soda-AQ pulp was also proposed[12], the bleaching conditions and the DP quality have not been clarified. The DP quality depends both on the properties of the raw materials and the cooking and bleaching processes.

2. Experimental

2.1 Raw materials and analysis of chemical composition

EFB was obtained from a palm oil mill of PT. Perkebunan Nusantara VIII in Bogor, West Java, Indonesia. EFB was cut into 10 to 15 cm fiber fragments and dried to 8–10% of moisture content; the fibers of EFB were then further cut to a length between 0.2 and 1.0 cm with a laboratory disc mill for cooking. EFB materials were milled with a Wiley mill and sieved to retain particles of 40–80 mesh in size. They were kept at room temperature and air-dried. The milled materials were then soxhlet-extracted with acetone and *n*-hexane to determine the content of the extractives, according to the TAPPI Test Method[13]: T204 om-88. Meanwhile, air-dried pulp sheets were prepared to measure the pulp yield (as oven-dried) after cooking.

The acid-insoluble lignin (klason lignin) content of the raw materials was determined using the method of Yoshihara *et al.*[14] with modified procedures as follows[15]; the prepared sample (0.5 g, as oven-dried weight) was primarily hydrolyzed with 72% sulfuric acid for 2.5 h and further hydrolyzed with 4% sulfuric acid at 121°C for 1 h. The sample was then filtered to obtain the residue and filtrate with a glass filter (1 GP 16). The weight of the residue was measured as acid-insoluble lignin. The amount of acid-soluble lignin was determined according to UV spectrophotometry at the wavelength of 205 nm (TAPPI Test Method: UM-250). The amounts of glucose and xylose were determined from the 1000 times diluted filtrate by using ion chromatography according to the procedures described in a previous study[16]. The system using Dionex ICS 3000 ion chromatograph (Dionex, Sunnyvale, CA, USA) consisted of a single pulp model (SP-1), an electrochemical detector (ED), an IonPac AS 7 column (Φ 4 mm x 250 mm), an IonPac AS 7 guard column (Φ 4 mm x 50 mm), and an auto sampler (AS). The EFB materials were incinerated at 575°C for 4 h to determine the ash content according to the TAPPI Test Method: T211 om-93.

2.2 Pyrolysis gas chromatography/mass spectrometry (Py-GC/MS)

The EFB materials (150–200 μg, as oven-dried weight) were used for the Py-GC/MS analysis. The suitable conditions for the Py-GC/MS analysis of lignin were chosen according to a previous study as follows[17]; JHP-5 (Japan Analytical Industry Co., Ltd., Tokyo, Japan) was used as the pyrolyzer. The pyrolysis temperature, pyrolysis time, pyrolyzer temperature, and transfer tube temperature were set at 500°C, 4 s, 250°C, and 250°C, respectively. GCMS-QP 5050 A (Shimadzu Corporation, Kyoto, Japan) was used as the GC/MS system. An HP 1-MS (30 m × 0.25 mm; film thickness: 0.25 μm) column was used with *n*-eicosane solution as the internal standard. The concentration of *n*-eicosane in ethyl acetate was 1 μg/μL.

2.3 Prehydrolysis and cooking

EFB materials (35 g, as oven-dried weight) were placed in a 300 mL steel reactor and subjected to prehydrolysis with distilled water for 90 to 180 min at 150°C. After prehydrolysis, a part of the prehydrolysis liquor (PHL) was removed or non-removed. The liquor to solid ratio was 7 or 3 (mL/g) for the treatment of PHL removal or non-removal, respectively. After prehydrolysis, the solid residue was subjected to soda-AQ cooking for 180 min at 160°C with 19%, 20%, or 21% of active alkali (AA) dosages. AQ (SAQ: 1,4-dihydro-9,10-dihydroxyanthrcene sodium salt, provided by Kawasaki Kasei Chemicals Ltd.) dosage and liquor to solid ratio were 0.1% and 7, respectively.

2.4 Bleaching sequences and conditions

The following sequences and conditions are summarized in **Table 2-2**. For a D_0–E_p–D_1 ECF bleaching sequence, prehydrolysis soda-AQ pulp (10 g, as oven-dried weight) was treated in a bag made of polyvinylidene chloride sheets with ClO_2 at the pulp consistency (PC) of 10%. After washing, the pulp was treated in a polyethylene bag with NaOH and H_2O_2, washed again, and treated in the polyvinylidene chloride bag with ClO_2. For the O–P_{sa}–E_p–

P_{sa}–E TCF sequence, prehydrolysis soda-AQ pulp (10 g, as oven-dried weight) was oxygen (O) bleached at the PC of 30%. Peroxymonosulfuric acid (P_{sa}) was synthesized according to a previous procedure[18] by dropping 95% sulfuric acid (Wako Pure Chemical Industries, Ltd.) into 45% hydrogen peroxide aqueous solution (Mitsubishi Gas Chemical Company, Inc.) at 70°C. A target amount of P_{sa} and a small amount of NaOH aqueous solution for adjusting to pH 3.0 were added to the pulp suspension at the PC of 10% in a polyethylene bag.

2.5 Evaluation of pulp qualities

The contents of acid-insoluble lignin and acid-soluble lignin, and the carbohydrate composition were determined according to the methods mentioned previously. Kappa number, viscosity, brightness, and α-cellulose were determined using the TAPPI Test Methods: T236 om-13, T254 cm-10, T452 om-08, and T429 cm-10, respectively. The brightness (ISO) was measured using a Tokyo-Denshoku Digital Color Meter Model TC-1500 SX. The classification of the fiber length distribution was measured according to JIS P 8207 (Pulp test method for classification with screen) using 710, 355, 180 and 75 μm opening screens (24, 42, 80 and 200 mesh, respectively). The fiber length and fiber width of some pulps were also determined using a Lorentzen-Wettre fiber tester CODE 912.

3. Results and discussion

3.1 Chemical characteristics of EFB materials

As reported in **Table 2-1**, the contents of glucan (cellulose), xylan (hemicellulose), and total lignin (acid-insoluble and acid-soluble) were 35.7%, 20.1%, and 27.6% for EFB material, respectively. The total of the extractives in the *n*-hexane and acetone solvents was 7.1%. The Py-GC/MS analysis confirmed that EFB lignin comprised guaiacyl and syringyl types, and that EFB materials still contained palmitic acid and oleic acid as components, which are suggested to be originally derived from the palm oil of fresh fruit (**Fig. 2-2**).

Table1 2-1 Chemical composition of EFB materials and EFB pulp

	Pulp yield	Kappa number	Acid-insoluble lignin	Acid-soluble lignin	Glucan	Xylan	Other sugars	Extractive	Ash	Unknown
						%				
EFB	-	-	24.6	3.0	35.7	20.1	1.6	7.1	5.5	2.4
Soda AQ pulp[1)]	41.8	17.3	-	-	-	-	-	-	-	-
Prehydro-lysis Soda AQ pulp[1,2)]	31.1	9.6	0.9	0.2	22.4	4.1	-	-	0.4	3.1
Prehydro-lysis kraft acasia pulp[3)]	45.2	13.2	0.7	0.4	39.2	4.3	-	-	0.1	0.5

Note: 1): Cooking temperature: 160°C, cooking time 180 min, AA dosage: 20%

2): Prehydrolysis time: 180 min, prehydrolysis temperature: 150°C

3): *Acacia mearinsii*[18)], (%): based on material weight

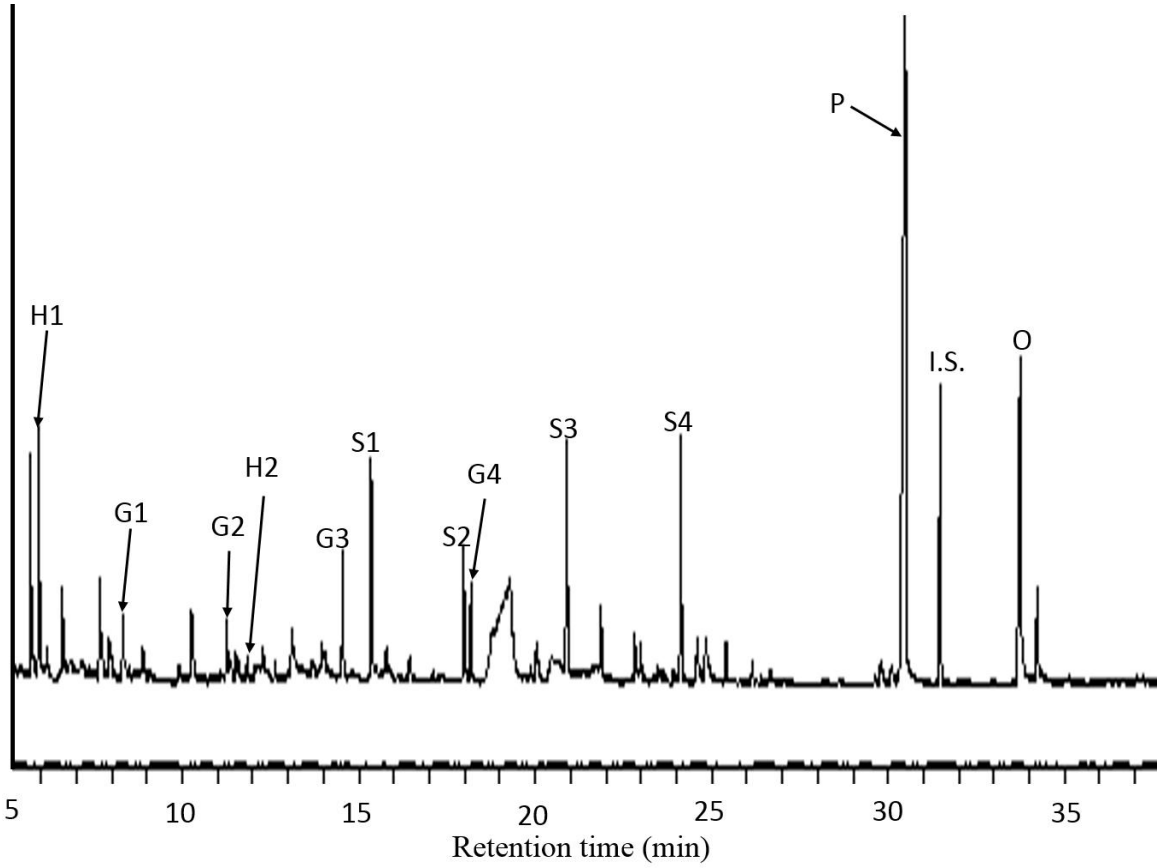

Fig. 2-2 Total ion chromatogram of pyrolysis products of raw material by Py-GC/MS

Legend:

Peaks	Products	Main MS ions, *m/z* (relative intensity %)	Retention time (min)
H1	phenol	94 (M^+, 100), 66 (38)	5.9
G1	guaiacol	124 (M^+,83), 109 (100)	8.3
G2	4-methylguaiacol	138 (M^+,100), 123 (93), 95 (42), 77 (22)	11.3
H2	4-vinylphenol	120 (M^+,100), 91 (55), 65 (23), 43 (25)	12.3
G3	4-vinylguaiacol	150 (M^+,100), 135 (83), 107 (34), 77 (36)	14.5
S1	syringol	154 (M^+,100), 139 (55), 111 (26), 93 (33)	15.3
S2	4-methylsyringol	168 (M^+,100), 153 (49), 125 (27), 107 (16)	18.0
G4	*trans*-isoeugenol	164 (M^+,100), 149 (35), 131 (24), 103 (22)	18.2
S3	4-vinylsyringol	180 (M^+,100), 165 (40), 137 (32), 122 (14)	20.9
S4	*trans*-4-prophenylsyringol	194 (M^+,100), 179 (18), 151 (13), 131 (18)	24.1
P	palmitic acid	256 (M^+), 239, 227, 213	30.4
I.S.	*n*-eicosane	282 (M^+), 269, 253, 238	31.5
O	oleic acid	282 (M^+), 264, 247, 235	33.7

We compared the chemical composition of EFB with those of other two lignocellulosic materials: sugarcane bagasse[19] (an agricultural residue biomass) and *Acacia mearnsii* wood[18] (a plantation hardwood for a resource of paper production). The comparison suggested that EFB is a high-potential resource, revealing a high content of total carbohydrate (glucan and xylan) of 55.8%, similar to those of sugarcane bagasse (62.0%) and *A. mearnsii* (58.6%) for paper and paperboard. The key advantage of EFB as a potential resource for DP production in Indonesia is provided by its considerably more abundant stock than bagasse, which has been estimated to reach only around 2.9 million tons annually[19]. Moreover, EFB is a waste biomass while *A. mearnsii* is an economically important resource in pulp and paper industry. However, EFB had a slightly higher lignin content than sugarcane bagasse (24.1%) and *A. mearnsii* (23.2%).

3.2 Application of soda-AQ cooking to EFB

The yields of DPs from woods (given by acid sulfite, multistage sulfite, and prehydrolysis kraft methods) are generally between 35% and 40%[20]. The prehydrolysis process is preferred for preparing DPs from hardwoods with an α-cellulose content higher than 90%.

In this study, effects of prehydrolysis treatments of EFB were thoroughly evaluated based on the delignification of the soda-AQ cooking process. As shown in Table 2-1, a kappa number and a pulp yield of EFB prehydrolysis soda-AQ pulp were 9.6 and 31.1, respectively, which were significantly lower than those soda-AQ pulp (17.3 and 41.8, respectively). Differences in kappa numbers and yields between the two pulps cannot be explained by the amounts of lignin and carbohydrates which were dissolved during the prehydrolysis. Previous studies[21, 22] suggested that prehydrolysis with water may diminish the linkages between lignin and carbohydrates, and that the dissolution of hemicelluloses would also create some

holes in the cell walls of the material, resulting in an efficient treatment in the subsequent process.

Table 2-1 presents the chemical composition of EFB and acacia pulp as a reference. The glucan content in the prehydrolysis soda-AQ pulp was 22.4%, showing a 13.3% decrement with respect to the EFB materials. The EFB materials had a relatively high ash content. The main elements in the EFB ash are reported to be potassium, calcium, and silica, with a significant amount of chlorine and sulfur[23]. The ash content decreased after cooking from 5.5% of the EFB material to 0.4% of the prehydrolysis soda-AQ pulp. Compared with the acacia, the contents of lignin, xylan, and ash of the EFB were high, and therefore the prehydrolysis conditions for EFB were more strengthened than those of acacia.

3.3 Effect of prehydrolysis time

As confirmed in **Fig. 2-3** and **Fig. 2-4**, the extension of prehydrolysis period assisted the decrease of the kappa number and pulp yield. Prehydrolysis for 180 min followed by soda-AQ cooking with 20% of AA dosage was selected as the optimum condition, as it resulted in a kappa number lower than 10. Thus, it was expected to give a slightly higher pulp yield than that obtained after 150 min of prehydrolysis and more than 21% of AA dosage in the cooking (**Fig. 2-4**). Further addition of AA was shown to have no significant effect on the kappa number decrement while resulting in a pulp yield decrement.

Interestingly, the prehydrolysis condition for 180 min followed by soda-AQ cooking with 20% of AA dosage resulted in the highest value of viscosity with a kappa number of 9.6 and pulp yield of 31.1% (**Fig. 2-5**). Further addition of AA was shown to result in a decrement of viscosity. The optimum kappa number with a suitable viscosity will provide an advantage in the following bleaching step.

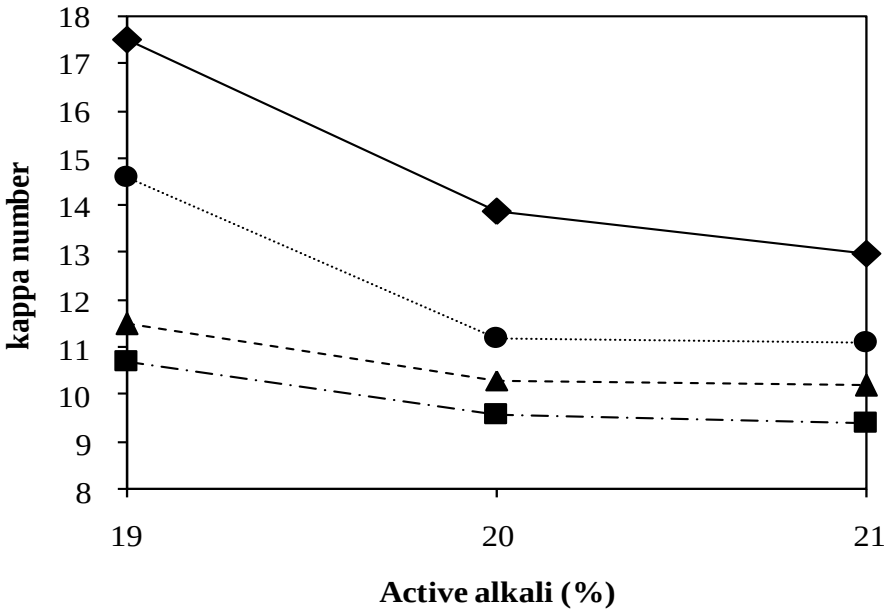

Fig. 2-3 Effect of prehydrolysis time on kappa number of soda-AQ pulp

Legend: ◆: 90 min; ●: 120 min; ▲: 150 min; ■: 180 min

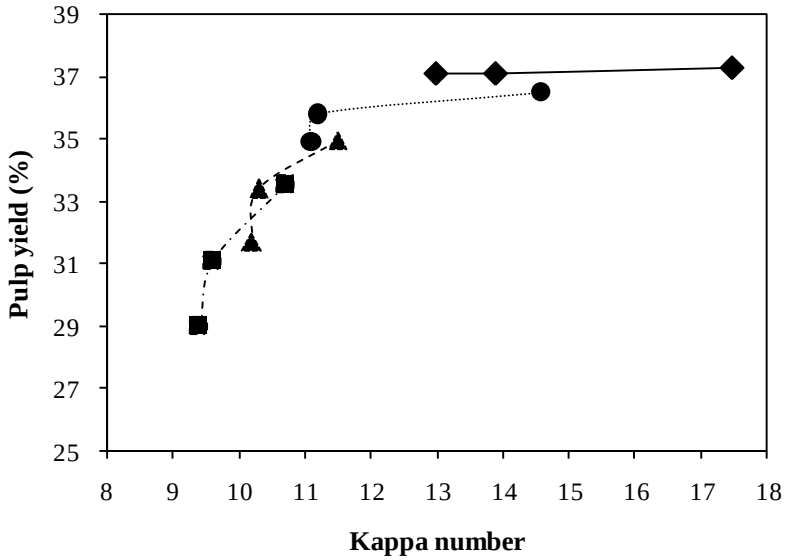

Fig. 2-4 Relationship between kappa number and pulp yield of prehydrolysis soda-AQ pulp

Legend: ◆: 90 min; ●: 120 min; ▲: 150 min; ■: 180 min

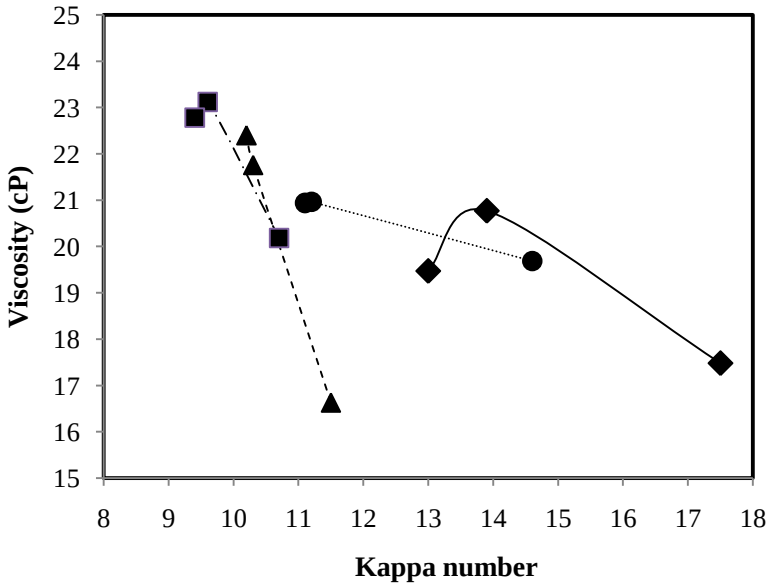

Fig. 2-5 Relationship between kappa number and viscosity of prehydrolysis soda-AQ

Legend: ◆: 90 min; ●: 120 min; ▲: 150 min; ■: 180 min

3.4 Effect of PHL removal

As shown in **Fig. 2-6**, the removal of PHL clearly affected the decrement of the kappa number. Moreover, **Figs. 2-6, 2-7, and 2-8** show that the PHL removal generated lower pulp yields, kappa numbers, and viscosities than the PHL non-removal at the same AA dosages. It was reported in a previous study[18] that the prehydrolysis treatment could remove a part of xylan and a small part of lignin from wood into the PHL. Therefore, the soda-AQ delignification might be affected by the presence of organic compounds such as a part of xylan and lignin fraction as well as acetic acid in the PHL. The presence of acetic acid also decreased the pH of cooking with the non-removal PHL, thus causing a more alkaline consumption during the soda-AQ delignification.

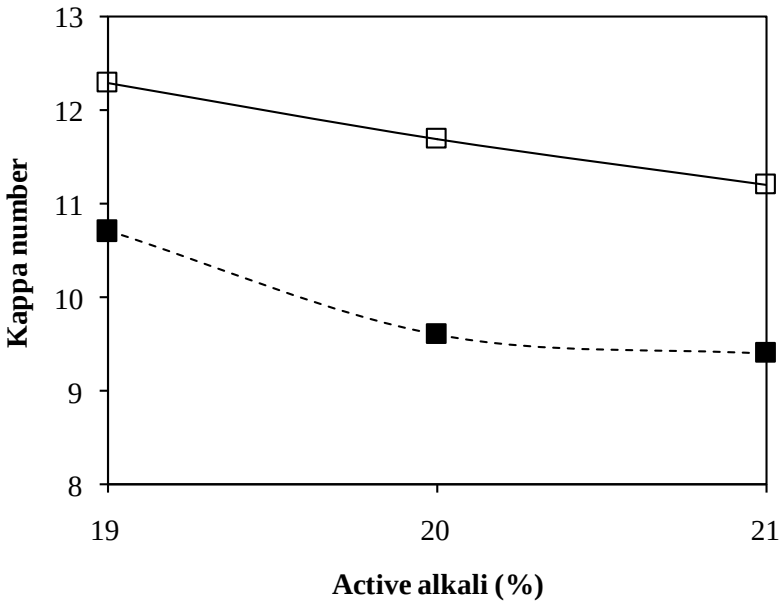

Fig. 2-6 Effect of PHL removal on kappa number of soda-AQ pulp

Legend: □ : non-removal of PHL; ■: removal of PHL

Generally, saving chemicals of active alkali resulted in higher pulp yield and kappa number (**Fig. 2-7**), and would results in pulp viscosity. However, pulps with the highest viscosities were obtained at 20% of AA dosage in both cases of the PHL removal and non-removal (**Fig. 2-8**). As mentioned before, this is a very interesting behavior. Further addition of AA to obtain a kappa number lower than 10 should result in a further decrement of pulp yield and viscosity in the case of the PHL non-removal. These results suggested that the PHL removal process was preferred.

Pulp with a kappa number of 9.6 and viscosity of 23.1 cP was obtained after 180 min of prehydrolysis and soda-AQ delignification with AA dosage of 20% (**Table 2-1**). In addition, the AA dosage of 20% was found to be optimal to maintain the viscosity level. This phenomenon is probably caused by a higher extent of hemicellulose degradation, which should increase

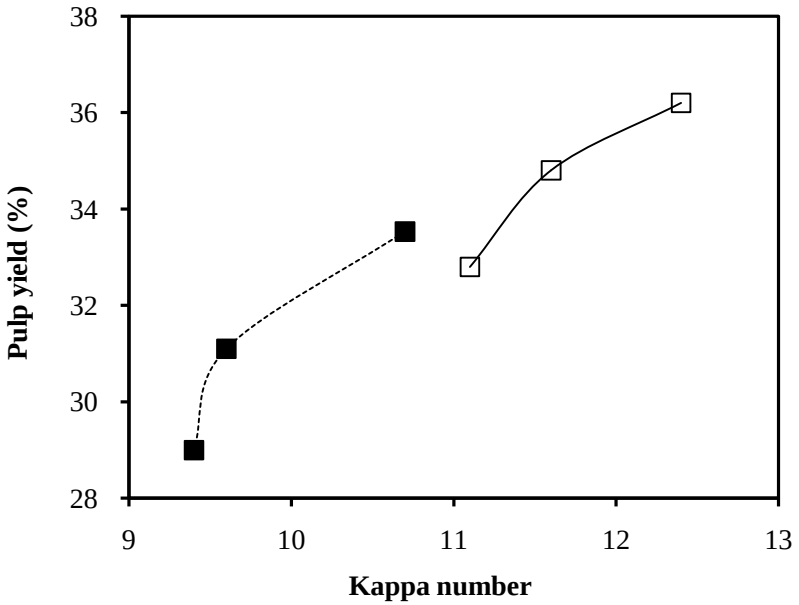

Fig. 2-7 Relationship between kappa number and pulp yield of soda-AQ pulp with PHL removal

Legend : □ : non-removal of PHL; ■: removal of PHL

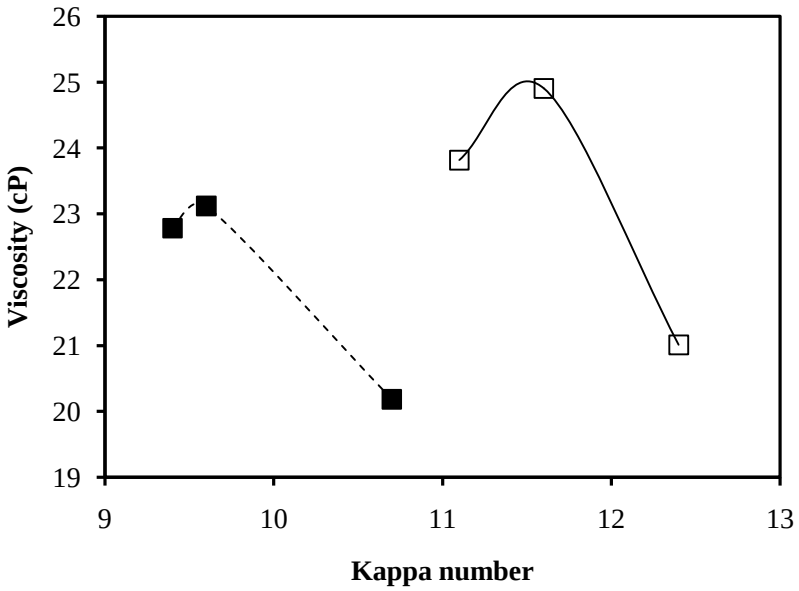

Fig. 2-8 Relationship between kappa number and viscosity of soda-AQ pulp with PHL removal

Legend : □ : non-removal of PHL; ■: removal of PHL

the cellulose content and the viscosity, than that of the treatment with AA dosage of 19%. However, it can be considered that for an AA dosage higher than 21%, cellulose degradation should start and thus cause the viscosity decrement.

3.5 Effect of bleaching sequence on pulp properties

Pulp with a kappa number of 9.6 and viscosity of 23.1 cP was treated with several bleaching sequences. As shown in **Table 2-2**, pulps with sufficient levels of viscosity and α-cellulose content were obtained by three sequences. Brightnesses were 89.2% and 90.7% ISO for the ECF sequences. Sharma *et al.*[24] recently reported on a pilot process of EFB soda-AQ for ECF bleaching. In this study, higher brightness pulps were obtained although dosage of ClO_2 was low. According to the Indonesian National Standard (SNI Standard)[25], where 6.2 cP viscosity, 94% α-cellulose, and 88% ISO brightness are required as minimum levels, these results of **Table 2-2** indicate a potential for the production of DP from EFB by using a combination of prehydolysis, soda-AQ cooking, and modified ECF bleaching with P_{sa} treatment.

For the TCF bleaching sequence, the oxygen delignification should be the first stage of the sequence, as it has the ability to delignify and increase the pulp brigthness without substantial reduction in pulp yield, and viscosity. Although the target brightness of 88% ISO could not be reached for this TCF bleaching sequence, the brightness level 81.6% ISO still showed a potential for TCF bleaching application to the EFB pulp.

Table 2-2 Viscosity, brightness and α-cellulose of bleached pulp

Bleaching sequence	Viscosity (cP)	Brightness (% ISO)	α-Cellulose (%)
D_0	13.5	58.9	-
D_0-E_p	10.4	79.8	-
D_0-E_p-D_1	10.3	89.2	98.2
P_{sa}-D_0-E_p-D_1	8.5	90.7	98.6
O-bleached pulp	13.0	66.2	-
P_{sa}-treated pulp	14.0	54.5	-
O-P_{sa}-treated pulp	10.3	68.3	-
O-P_{sa}-E_p-P_{sa}-E	9.6	81.6	95.9

Bleaching conditions

ECF bleaching sequence: D_0-E_p-D_1 and P_{sa}- D_0-E_p-D_1	
Chlorine dioxide (D_0)	ClO_2 dosage: kappa factor 0.30 (2.9%), 60 min, 70°C, pH 3.4, PC: 10%.
Peroxymonosulfuric acid (P_{sa})	P_{sa} dosage: 0.2 %, 60 min, 70ºC, pH 3.0, PC: 10%.
Extraction with peroxide (E_p)	NaOH dosage: 1%, H_2O_2 dosage: 1.4%, 60 min, 70ºC, PC: 10%.
Chlorine dioxide (D_1)	ClO_2 dosage: 0.5%, 60 min, 70ºC, PC: 10%.
TCF bleaching sequence: O-P_{sa}-E_p-P_{sa}-E	
Oxygen (O)	Oxygen pressure: 0.5 MPa, NaOH dosage: 1%, 60 min, 115ºC, PC: 30%.
Peroxymonosulfuric acid (P_{sa})	P_{sa} dosage: 0.2 %, 60 min, 70ºC, pH 3.0, PC: 10%.
Extraction with peroxide (E_p)	NaOH dosage: 1%, H_2O_2 dosage: 1.4%, 60 min, 70ºC, PC: 10%.
Extraction (E)	NaOH dosage: 1 %, 60 min, 70ºC, PC: 10%.

PC: pulp consistency

3.6 Characterization of fiber length distribution

As shown in **Fig. 2-9**, the largest fractions of the fiber length for EFB unbleached and bleached prehydrolysis soda-AQ pulps were those with a 180–355 μm opening screen. Fully bleaching did not change the fiber length distribution, according to this method. Meanwhile, the largest fractions for eucalyptus wood prehydrolysis kraft pulps were those a with 355–710 μm opening screen. This result shows that the EFB pulp has shorter fibers than the eucalyptus pulp.

4. Conclusions

The removal of PHL during the prehydrolysis process was preferred for an effective soda-AQ delignification of EFB, and a pulp with a lower kappa number and lower hemicellulose content could be produced. The prehydrolysis for 180 min followed by soda-AQ cooking with 20% of AA dosage was selected as the optimum condition, as it resulted in a kappa number lower than 10 and a higher viscosity. A brightness level higher than 88% ISO brightness was obtained by ECF sequences with sufficient levels of viscosity and α-cellulose. The production of DP from EFB with the combination of prehydolysis, soda-AQ cooking, and modified ECF bleaching with P_{sa} treatment was demonstrated. Although the target brightness was not obtained for the proposed TCF bleaching sequence, this sequence still showed a potential for application.

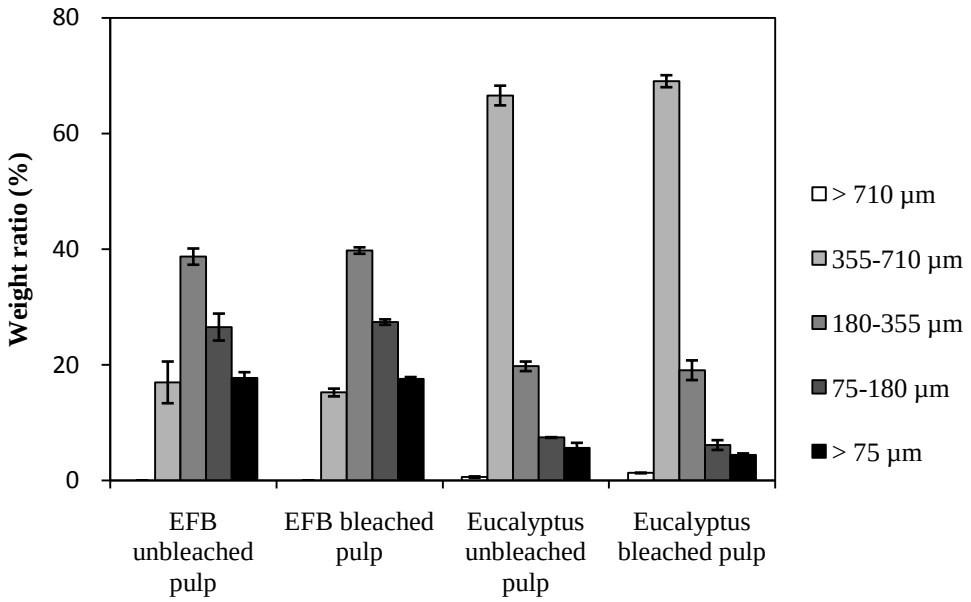

Fig. 2-9 Fiber length distribution of EFB and eucalyptus pulp

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Chapter 3. Properties of fibers prepared from oil palm empty fruit bunch for use as corrugating medium and fiberboard

1. Introduction

Though it is the main by-product of palm fruit and oil industries, EFB is generally left rotting at the plantation site without any further processing. On average, EFB represents around 22–24 % of the total weight of fresh fruit bunches processed in palm oil mills, and so clearly action must be taken to reduce the negative impact of this waste. One possible solution is to utilize this waste for other purposes, thereby increasing the positive impacts of the palm fruit and oil industries[1)]. EFB has considerable potential to be used in organic fertilizers or animal fodder, and for use as a raw material for brick and mat-making, or as a growth media for fungi and plants, and as a lignocellulosic raw material for paper making.

Compared to other woody materials, non-wood raw materials tend to have a similar cellulose content, but are lower in lignin and higher in hemicelluloses and silica[2)]. This gives non-wood materials like EFB some advantages when used in pulp and paper processes, which has led to a number of researchers studying this possibility. For instance, Tanaka *et al*.[3)] and Jimenez *et al*.[4)] have suggested that paper sheets can be produced from EFB, while Singh *et al*.[5)] have discussed the prospect of using EFB for the manufacture of pulp and paper. The production of pulp for paper making from non-wood fibrous materials has grown significantly with the increasing demand for paper and paper products, especially when wood supplies have proven insufficient. Some research into the use of EFB as a dissolving pulp has also been undertaken[6-8)], but the optimal conditions required to reach an acceptable standard of quality have yet been determined. Thus, further research is needed into the optimal conditions required to produce dissolving pulp and paper from EFB through chemical and mechanical pulping.

Mechanical pulping is typically used for softwood and offers the advantage of a great yield of around 85−95% of the dried weight of the input material. However, its disadvantages are that it requires a great deal energy, and the strength of mechanical pulps is generally weaker than that of chemical pulps. The development of refiner mechanical pulping uses heat and/or chemicals to help the softening of wood. These are typically referred to as thermo-mechanical pulping (TMP) and chemi-thermomechanical pulping (CTMP), respectively. The quality of both the pulp and paper is influenced by both the material quality and the refining conditions used[9]. In the TMP process the wood chips are steamed for several minutes under pressurized conditions, in contrast, the CTMP of softwood chips involves treating with a mild sulfite solution to modify the lignin and slightly delignify the wood prior to refining.

Some hardwoods require chemical pre-treatment to produce a suitable mechanical pulp, which has led to the development of alkaline peroxide mechanical pulping (BCTMP or APMP). This offers several advantages in terms of providing good pulp qualities and energy savings compared to conventional CTMP, making it a good pulping method for low-density hardwoods such as for aspen and eucalyptus[10]. This method is used along with TMP and CTMP to treat the raw materials used to create papers such as newsprint and printing paper. This study though looks at the possibility of using EFB as a raw material for paperboards such as corrugating medium.

It is well-known that EFB morphologically consists of various kinds of plant cells[11], but sourcing it as a byproduct introduces some undesired characteristics from a utilization point of view. For example, as EFB residue is quite bulky and contains a large amount of moisture, it is difficult to dry and often retains a small amount of oil[12]. This makes it necessary to reduce the size of the material to facilitate drying and transport. As a lignocellulosic material, EFB contains around 18–23% lignin, 35% α-cellulose, and 25% hemicellulose[13]. This has prompted several studies into the use of EFB waste as a wood

replacement material in the creation of particleboard[14], composites[15] or briquettes[16]. In addition, lignin and carbohydrates isolated from EFB have the potential to provide the resources for new biomaterials and bioethanol production.

The goal of this study is to improve the utilization of waste from the palm oil industry by identifying products that can be manufactured from EFB. To this end, the optimal pretreatment conditions needed to produce mechanical pulp were first identified, and were then applied to creating medium density fiberboard (MDF) from EFB. As the complex structure of organic materials in these EFB fiberboards can potentially provide a food source for fungi, the second objective was to evaluate the degradation resistance of EFB fiberboards. The final objective was to utilize EFB waste as a raw material for manufacturing MDF.

2. Experimental

2.1 Raw materials for paperboard and fiberboard

Two kinds of EFB material were obtained from the palm oil mills of PT. Perkebunan Nusantara VIII in Bogor, West Java, Indonesia. Unrefined EFB fibers were obtained from these materials through several stages of processing. The EFB byproducts were shredded into long fiber fragments (10–15 cm), sun dried until reaching 8–10% moisture, and then further cut into 0.2–1.0 cm or 0.5–4.0 cm lengths by a laboratory disk mill.

2.2 Production of chemically pretreated-mechanical pulp

The EFB fibers cut to a length 0.5–4.0 cm were used for making a mechanical pulp by means of a thermo-mechanical pulp (TMP) refiner (Kumagai Riki Kogyo Co., LTD., Nerima, Tokyo) with a disk clearance set to 0.10, 0.15 or 0.20 mm. The chemical pretreatment conditions used were as follows: liquor to EFB fiber ratio: 7 L/kg; NaOH addition level: 0, 1, or 2% based on the oven-dried weight of EFB fibers; pretreatment temperature and time: 121°C for 1, 2, or 3 h. After pretreatment, the yield loss and pH of the liquor were determined.

Refining of the pretreated EFB fibers was carried out under atmospheric conditions as no way could be found of feeding sufficient fibrous material into the process otherwise.

2.3 Evaluation of chemically pretreated mechanical pulp

Classification of the refined fibers (i.e., the fiber length distribution) was conducted according to Japanese Industrial Standards (JIS) 8207 (pulp test method for classification with screen) using 710, 355, 180 and 75 µm opening screens (24, 42, 80 and 200 mesh, respectively). After chemi-mechanical pulping, the refined pulp was further refined by a PFI mill at some beating revolution speed, and the physical properties of handsheets were determined along with the tensile and tear indices in accordance with ISO/FDIS 5270:2011(E), 1924:2008(E), and 1974:2011(E), respectively. The fiber length and fiber width of some pulps were also determined using a Lorentzen-Wettre fiber tester.

2.4 Preparation of EFB fiberboard

Fiberboard was made from unrefined EFB fibers (0.2–1.0 cm in length) by first blending dried EFB fibers with 12% melamine urea formaldehyde (MUF) resin and 1% wax (based on EFB oven-dried weight) in a rotating drum-type mixer fitted with a pneumatic spray gun. This mixture was manually distributed in a wood mold to form mats, which were then cold-pressed at 3 MPa for 30 seconds, followed by hot-pressing (180°C) at 5 MPa for 60 seconds. For comparison, MDF was also made from refined fibers of mixed light hardwoods (MLH) provided by a MDF mill. The dimensions of the EFB fiberboards and hardwood MDF was 300 x 300 x 12 mm (9 mm after polishing), with a target density of 700 kg/m^3. All samples were conditioned at 20ºC and 65% relative humidity for a week. The above experiment was conducted by Hokushin Co., Ltd., Kishiwada, Osaka.

2.5 Preparation of EFB pellet

Trials of pelletizing EFB fibers (0.2–1.0 cm in length) were conducted at the Hokkaido Research Organization, Forest Research Development, Forest Products Research Institute

(FPRI), Asahikawa, Hokkaido, and at the Dalton Co., Ltd. Tokyo Test Center, Itabashi, Tokyo.

2.6 Refining of EFB fibers and EFB pellets

The refining of EFB fibers (0.5–4.0 cm in length) was performed by FPRI, Asahikawa, Hokkaido, though machine conditions such as the pressure of the steam boiler source (2 MPa), temperature of hot water in the refiner (148ºC), and temperature of the dryer (210ºC) were defined in advance. To produce refined EFB fibers, the TMP refiner was set to a disk pressure of 0.7 MPa, 166°C refining temperature and 0.15 mm disk clearance. These conditions were very important because the steam pressure induces change in the chemical components of the fibers, and it is well-known that the physical and mechanical properties of the refined fibers are strongly influenced by the refining pressure as a result[17]. Refining of the EFB pellets at 0.4 MPa (140°C) was performed by Kumagai Riki Kogyo Co., LTD, Nerima, Tokyo using a disk clearance of 0.15 mm.

2.7 Decay testing

Decay testing was conducted on blocks of fiberboard made from unrefined EFB fibers and on MDF produced from refined MLH fibers according to JIS K 1571. The decay test results were considered acceptable if the weight loss of sugi (*Cryptomeria japonica*) wood specimens was greater than 30 % for *Fomitopsis palustris* (brown-rot fungus), and greater than 15 % for *Trametes versicolor* (white-rot fungus).

To provide a growth medium, sea sand in a glass jar was infused with nutrient solution and fungal inoculum (*Fomitopsis palustris* or *Trametes versicolor*). When mycelium fully covered this medium, three replicate specimens of EFB fiberboard or control material prepared by sterilizing with gaseous ethylene oxide were placed on top of the glass jar. A plastic mesh spacer was used between the specimens and mycelium for *F. palustris*. The test jars were then incubated at 27°C for 12 weeks, and the extent of fungal attack in this time

was expressed as the average mass loss (%) calculated from the oven-dried weights of five of the nine specimens before and after the decay procedure.

3. Results and discussion

3.1 Properties of chemically pretreated, mechanically pulped EFB

3.1.1 Effect of pretreatment on yield loss

Rosli *et al.*[18] used room temperature sodium hydroxide (NaOH) as a chemical pretreatment to produce serviettes from EFB, though in this study NaOH at 121°C was used for chemical pretreatment of EFB. Rosnah *et al.*[19] also demonstrated that the time of alkaline hydrogen peroxide treatment has a significant effect on the brightening and softening of EFB. With tropical bamboo, pretreatment by soaking in 2% NaOH for 6 h significantly is known to affect the properties of the mechanical pulp[20], but using alkaline pretreatment to improve the strength of EFB materials causes a decrease in yield. This is shown in **Table 3-1**, in which we see that an increase in NaOH dosage causes yield loss, as does increasing the pretreatment period except when the final liquor pH (6.20) is sufficient to re-precipitate previously dissolved hemicellulose. Fatehi *et al.*[21] have also observed a decrease in mechanical pulp yield with increasing alkali dosage, which suggests that this is caused by a reduction in hemicellulose and organic acid.

3.1.2 Effect of pretreatment on fractionation of refined fibers

There are two disks in the TMP refiner, one of which is bound to a moving rotor, while the other is a static stator. The space between these two disks is termed the refining zone, and is where materials are processed gradually from the feeder to the outlet. In this study, refining of pretreated EFB fibers was carried out under atmospheric condition, as it was found that fibrous materials like EFB could not be processed smoothly into the refining zone under pressurized conditions and so needed to be fed manually. As shown in **Fig. 3-1**, the biggest rate fraction of the five fiber fractions obtained by refining with a 0.15 or 0.20 mm disk

clearance was achieved with an opening size of more than 710 µm, while a 0.10 mm disk clearance required a 355–710 µm opening size. This indicates that the fibrous EFB materials could not be sufficiently refined into single fibers (cells), and that instead shives of fibers remained in the pulp.

Figure 3-2 shows that the ratio of small and fine fiber fractions with a 180–355 µm opening, 75–180 µm opening and <75 µm opening decreased with increasing NaOH dosage. This can be explained by a decrease in fiber cutting through an increase in the flexibility of the fibers. Similarly, **Figure 3-3** shows that the ratio of small and fine fiber fractions with a 180–355 µm opening and 75–180 µm opening decreased with increasing pretreatment time from 1 to 2 h, with the similarity in results between 2 and 3 h suggesting that 2 h of pretreatment is sufficient.

Table 3-1 Effect of chemical pretreatment on yield loss

NaOH dosage [a] (%)	Treatment time (h)	After pretreatment at 121ºC	
		Liquor pH	Yield (%)
0	1	6.76	94.9
0	2	6.46	93.6
0	3	6.20	94.0
1	1	6.77	93.0
1	2	6.77	92.6
1	3	6.74	92.2
2	1	7.76	92.0
2	2	6.96	91.2
2	3	6.90	87.3

[a] Liquor to EFB fiber ratio: 7 L/kg

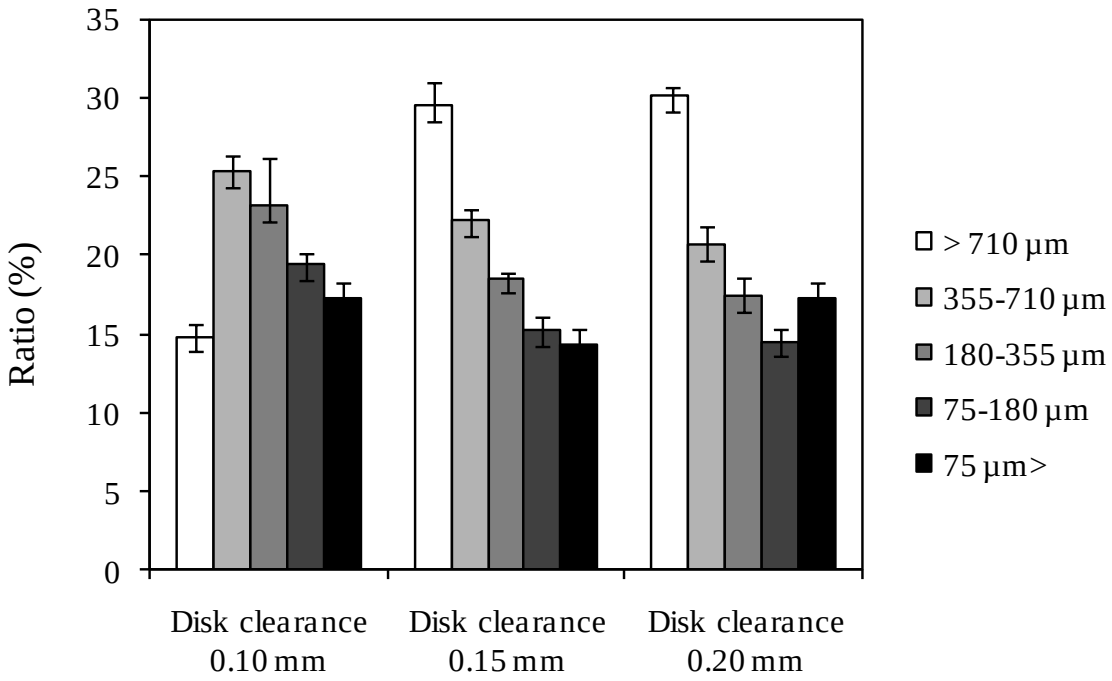

Fig. 3-1 Effect of disk clearance of refining on fiber fractionation
Note : Conditions: NaOH dosage: 0%; pretreatment time: 3 h.

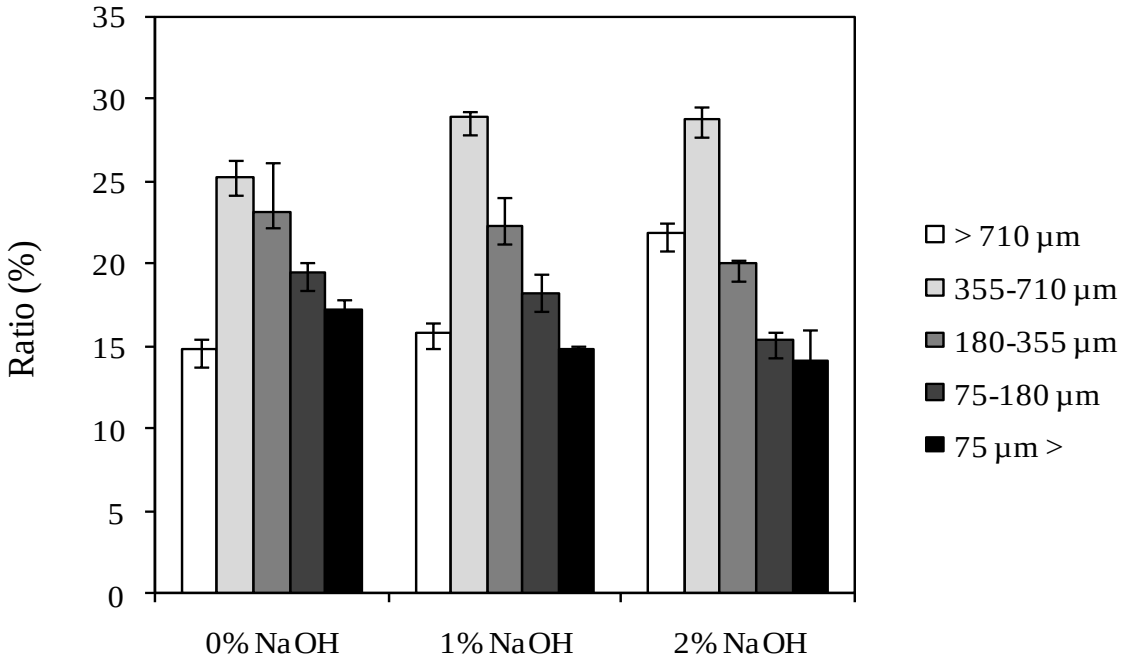

Fig. 3-2 Effect of NaOH dosage of pretreatmnet on fiber fractionation
Note : Conditions: Prereatment time: 3 h; disk clearance of refining: 0.10 mm.

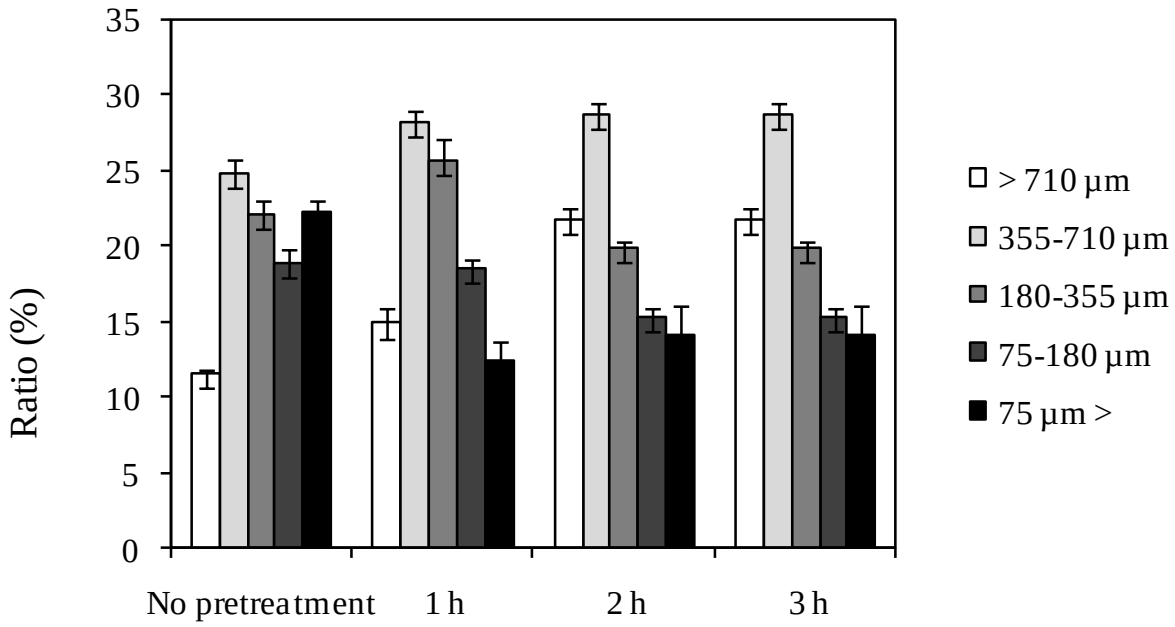

Fig. 3-3 Effect of pretreatment time on fiber fractionation
Note : Conditions: NaOH dosage: 2 %; disk clearance of refining: 0.10 mm.

3.1.3 Effect of pretreatment on the physical properties of a pulp

It is well-known that refining can induce many changes in the properties of a fiber, such as its internal fibrillation, external fibrillation and cutting[22]. One of the important consequences of refining is that it reduces fiber length, and hence has an adverse effect on the physical strength of paper. It is thought that internal fibrillation is caused by a compressive force from the disk absorbed to the material breaking the internal pore structure between cellulose microfibrils, which in turn makes the pore structure inside the cell wall expand and swell[23]. External fibrillation (or peeling) of the surface of the fiber cell wall causes the fibers to become more flexible.

Tensile strength is a function of fiber strength, fiber length and fiber-fiber bonding[24], whereas tear strength is a measure of the perpendicular force required to separate multiple plies of paper through a specified distance and is therefore also influenced by the orientation of the fibers in a paper sheet[25]. In **Table 3-2**, the tensile and tear indices of the pulp are shown alongside their respective pretreatment and refining conditions. It is evident from this that an increase in the NaOH dosage from 1 to 2% with a 2 h pretreatment time leads to an improvement in both the tensile and tear indices from 11.9 N·m/g and 5.67 mN·m^2/g to 19.7–20.4 N·m/g and 6.67–8.75 mN·m^2/g, respectively. These values are comparable with those of recycled pulp sheet from an old corrugated fiberboard box, which gave a tensile index of 19.8 N·m/g and a tear index of 8.69 mN·m^2/g. These indices were also improved by the extending the pretreatment period from 1 to 2 h, but decreased when the time was extended further to 3 h. The tensile strength increased with PFI beating from 7500 to 10000 rpm, but decreased at 12500 rpm. Based on this, EFB chemically pretreated mechanical pulp is considered suitable for use in the preparation of corrugating medium.

Table 3-2 Effect of pretreatment and refining conditions on pulp physical properties

Pretreatment		Refining		Pulp physical properties			
NaOH (%)	Time (h)	TMP disk clearance	PFI revolution	CSF (ml)	Density (kg/m^3)	Tensile ($N \cdot m/g$)	Tear ($mN \cdot m^2/g$)
0	0	0.10	10000	274	0.33	6.18	0.42
0	2	0.10	10000	278	0.33	7.96	4.88
1	2	0.10	7500	285	0.32	9.64	2.83
1	2	0.10	10000	242	0.33	11.9	5.67
1	2	0.10	12500	192	0.34	9.55	2.51
2	2	0.10	7500	250	0.35	19.7	8.75
2	2	0.10	10000	184	0.39	20.4	6.67
2	2	0.10	12500	154	0.39	18.6	4.01
2	3	0.10	10000	188	0.43	18.2	2.94
Recycled pulp[a]		-	-	542	0.42	19.8	8.69

[a] Prepared from old corrugated fiberboard box

3.2 Properties of fiberboard prepared from unrefined EFB fibers

3.2.1 Mechanical properties

The second step of this study was to determine the physical properties of fiberboard made using unrefined EFB fibers. As shown in **Table 3-3**, the MOR, MOE and IB of the fiberboard were 5.6 N/mm^2, 648 N/mm^2, and 0.2 N/mm^2, respectively, which are still below the minimum requirements mandated by the JIS. However, as these low values are largely due to the fibrous structure, they should improve when formed into a mat and combined with resin.

3.2.2 Decay resistance

Decay tests were conducted on blocks of fiberboard made from unrefined EFB fibers and MDF made from refined MLH fibers, with sugi (*C. japonica*) being used as a control specimen. In this test, the weight loss of the sugi control when exposed to *F. palustris* (brown-rot fungus) and *T. Versicolor* (white-rot fungus) was 31.6 and 27.4%, respectively, which is considered an acceptable result according to JIS K 1571 (**Fig. 3-4**). Kartal and Green III[26)] have presented similar results based on ASTM D2017, wherein the decay resistance is divided into four categories based on the total weight loss after 16 weeks exposed to brown-rot fungus or *T. Versicolor*: 0–10%: highly resistant; 11–24%: resistant; 25–44%: slightly resistant; >45%: non-resistant.

In this study, the weight loss of the sugi specimens was less than 50% after 12 weeks according to JIS K1571, while the weight loss of the EFB fiberboard caused by *F. palustris* and *T. versicolor* was 9.9 and 18.0%, respectively. The resistance of the EFB fiberboard to both brown-rot fungus and white-rot fungus was therefore better than that of sugi.

Table 3-3 Mechanical properties of fiberboard from unrefined EFB fibers and MDF from refined MLH fibers

	Material	Density (kg/m^3)	MOR[a] (N/mm^2)	MOE[a] (N/mm^2)	IB[a] (N/mm^2)
Fiberboard	EFB[b]	671	5.6	648	0.2
MDF	MLH[c]	728	36.4	3311	0.7

[a] MOR: modulus of rupture in static bending; MOE: modulus of elasticity in static bending; IB: internal bond strength;

[b] Unrefined fibers

[c] Refined mixed light hardwood fibers

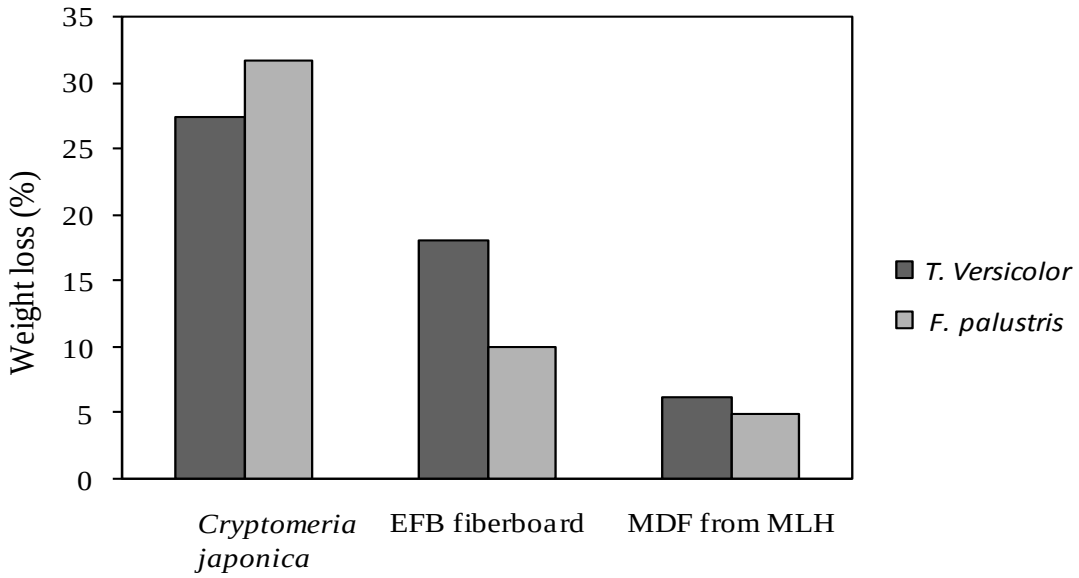

Fig. 3-4 Weight loss after decay test

3.3 Properties of EFB fibers refined under pressurized conditions

3.3.1 Pelletizing of EFB fibers

EFB materials cut to a size of 0.2–1.0 cm were pelletized at FPRI in Hokkaido prior to refining in order to facilitate feeding the fibers into the disk part of the refiner. Here, it was found that there were some necessary conditions for the raw materials to be formed into pellets: a moisture content of about 20%, a fiber length of no more than 1.0 cm. The diameter and length of the pellets formed were 0.5 cm and 1.5 cm, respectively (**Photo 3-1**).

3.3.2 Refining of EFB fibers

EFB pellets (0.5 cm diameter and 1.5 cm length) were also made at the Dalton Tokyo Test Center, and were subsequently refined to EFB fibers at 0.4 MPa (140°C) using a TMP refiner, and then immediately air-dried without dewatering. The refined fibers obtained were characterized according to JIS 8207 and compared with a pretreated-mechanical pulp (**Fig.3-5**). This found that TMP produces a smaller fraction of fine material (less than 75 μm) than mechanical pulping, with a subsequent increase in the large size fraction (355–710 μm). As both pulps were refined using a 0.15 mm disk clearance, both should also contain shives of fibers given that the ratio of the biggest size fraction (more than 710 μm) was still high. However, a decrease in the amount of fine fraction and an increased amount of the big size can be caused by a decrease in fiber cutting due to an increased flexibility of the fibers when refined at a higher temperature of 140ºC.

Refined EFB fibers were also made from EFB materials at 0.7 MPa (166°C) and were continuously dried by a hot-air dryer combined with the TMP refiner (**Photo 3-2**). The results of fiber fractionation using screens in accordance with JIS 8207 revealed the fiber length distribution characteristics of each pulp, but did not provide information as to the fiber length and fiber width of each pulp. These were therefore determined using a Lorentzen-Wettre fiber tester for a chemi-mechanical pulp and two kinds of thermo-mechanical pulps (166 and

EFB Fiber EFB Pellet (0.5 cm diameter and 1.5 cm length)

Photo 3-1 Pelletizing EFB fiber for thermo-mechnical refining

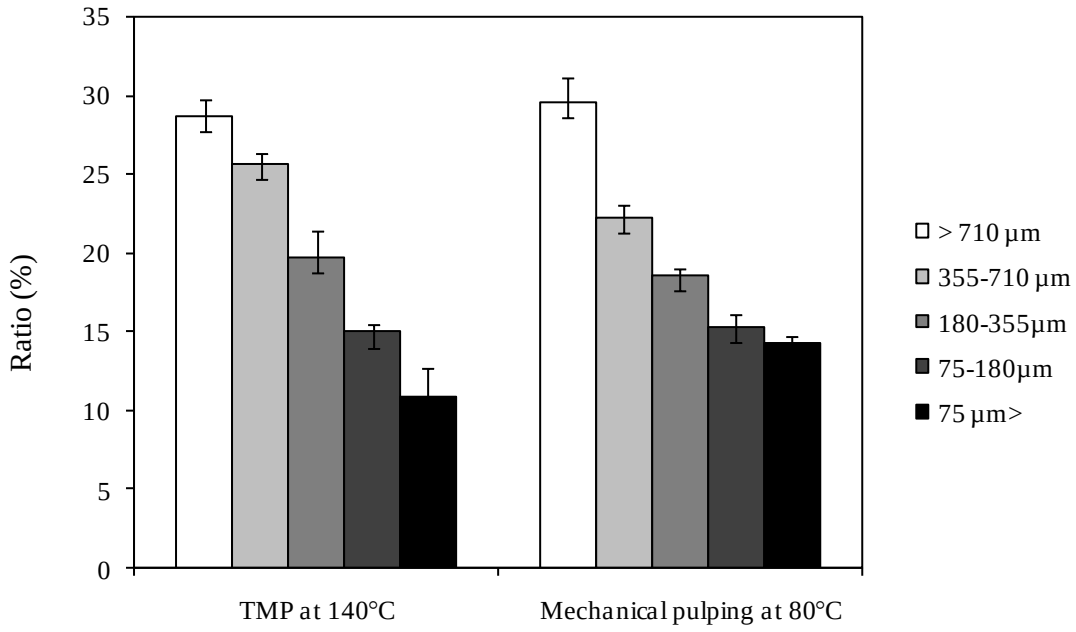

Fig.3-5 Effect of refining temperature on fiber fractionation

Note : TMP: Refining temperature 140°C; disk clearance of refining: 0.15 mm

Mechanical pulp: Pretreatment: 121°C for 3 h (NaOH dosage: 0%);

refining: at atmospheric pressure; disk clearance: 0.15 mm.

Photo 3-2 Unrefined EFB fiber (left) and TMP-refined and hot air-dried pulp (right)

140°C). As shown in **Table 3-4**, the chemi-mechanical pulp contained fibers that were longer and wider than those in the chemical pulp (an unbleached prehydrolysis soda-AQ pulp prepared as a dissolving pulp). Furthermore, a comparison of the two kinds of thermo-mechanical pulps shows that longer and wider fibers are obtained by increasing the refining temperature from 144 to 160°C. The fiber length and width of the dissolving pulp are considered to be small because of the low pulp yield, while the increase in temperature of TMP refining should increase fiber length by preventing the cutting of fibers.

Table 3-4 Fiber length and width by Lorentzen-Wettre fiber tester for refined EFB fibers.

	Mean length (μm)	Mean width (μm)
Chemi-mechanical pulp[a]		
Before screen fractionation	676	29.5
After screen fractionation		
More than 710 μm opening	ND[b]	ND
355-710 μm opening	1403	40.4
180-355 μm opening	750	29.9
75-180 μm opening	402	25.5
Pass of 75μm opening	371	23.2
Thermo-mechanical at 166ºC and hot air-dried pulp[c]	1023	41.2
Thermo-mechanical at 140ºC and warm air-dried pulp[c]	737	42.1
EFB chemical pulp[d]	556	18.0

[a] NaOH dosage: 2 %; pretreatment time: 3 h; disk clearance of refining: 0.10 mm

[b] Not determined

[c] Disk clearance of refining: 0.15 mm

[d] Reference 8); kappa number: 9.6; pulp yield: 31.1%

4. Conclusions

The pulp properties indicate that the optimum conditions for chemical pretreatment and mechanical pulping under atmospheric pressure are 121°C for 2 h with 2% NaOH, and a TMP refiner disk clearance of 0.10 mm. The tensile and tear indices obtained under these conditions were 19.7–20.4 N·m/g and 6.67–8.75 mN·m^2/g, respectively, which are similar to the values for a pulp prepared from waste corrugated fiberboard box and indicate that EFB fibers can feasibly be used in the preparation of corrugating medium.

The mechanical properties of fiberboard made from unrefined EFB fibers were still under those required by the JIS, but its resistance to attack by a brown-rot fungus (*Fomitopsis palustris*) and white-rot fungus (*Trametes versicolor*) was better than that of sugi (*Cryptomeria japonica*) specimens.

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Chapter 4. General Conclusion

In this study, EFB was converted into several derivative products such as dissolving pulp, paperboard, and particleboard.

In Chapter 2, we clarified the optimal conditions for preparing DP from EFB with combined prehydrolysis and soda-AQ cooking followed by elementary chlorine-free (ECF) or totally chlorine-free (TCF) bleaching. The selected optimum conditions for preparing DP involved the removal of PHL in the prehydrolysis for 180 min at 150°C followed by soda-AQ cooking with 20% AA at 160°C for 180 min. The pulp obtained has a kappa number of 9.6 and a viscosity of 23.1cP. DP with brightness : 90.7% ISO, viscosity : 8.5 cP, and α-cellulose : 98.6 % values exceeding the Indonesian standard (SNI) was obtained by the ECF bleaching with P_{sa}-D_0-E_p-D_1 sequences. The possibility of producing DP from EFB by the combination of prehydrolysis, soda-AQ cooking, and the modified ECF bleaching was demonstrated. Although the target brightness was not obtained with the proposed TCF bleaching sequence, the TCF bleaching sequence still exhibited potential for practical application.

In Chapter 3, the objective to improve the utilization of waste from the palm oil industry by identifying products that can be manufactured from EFB was addressed. Toward this end, the optimal pretreatment conditions required to produce mechanical pulp were first identified, and were then applied to creating medium density fiberboard (MDF) from EFB. As the complex structure of organic materials in these EFB fiberboards can potentially provide a food source for fungi, the second objective was to evaluate the degradation resistance of the EFB fiberboards. The final objective was to utilize EFB waste as a raw material for manufacturing MDF.

Analysis of the pulp properties indicated that the optimum conditions for chemical pretreatment and mechanical pulping under atmospheric pressure are: 121°C for 2 h with 2%

NaOH, and a TMP refiner disk clearance of 0.10 mm. The tensile and tear indices obtained under these conditions were 19.7–20.4 N·m/g and 6.67–8.75 mN·m^2/g, respectively, which are similar to the values for pulp prepared from waste corrugated fiberboard boxes, and indicate that EFB fibers can feasibly be used in the preparation of corrugating medium.

The mechanical properties of fiberboard made from unrefined EFB fibers were still below the values required by the Japanese Industrial Standards (JIS), but it should be noted that the degradation resistance of unrefined EFB fibers to be attacked by brown-rot fungus (*Fomitopsis palustris*) and white-rot fungus (*Trametes versicolor*) was better than that of sugi (*Cryptomeria japonica*) specimens.

Based on the above result, it can be concluded that the utilization of palm oil industry waste, especially EFB can be improved for the manufacturing of derivative products to support sustainable development of palm oil industry. The utilization of EFB as derivative products becomes an added value for the industry and at the same time decreases the pollution of the environment.