

EXPLICABILITY OF THE H-FACTOR TO ACCOUNT FOR THE DELIGNIFICATION EXTENT AND PROPERTIES OF PLANTATION FOREST WOOD PULP IN THE KRAFT COOKING PROCESS

(Penerapan Faktor-H untuk Menelaah Tingkat Delignifikasi dan Sifat Pulp Empat Jenis Kayu Hutan Tanaman Industri pada Proses Pengolahan Kimia Sulfat)

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ABSTRAK

Semakin terbatasnya sumber serat kayu di Indonesia dan anjuran mengurangi ketergantungannya dari hutan produksi alam untuk industri pulp dan kertas menyebabkan kekhawatiran serius. Satu usaha mengatasinya adalah pembangunan hutan tanaman industri (HTI) sebagai pemasok serat kayu. Perbedaan jenis kayu HTI bisa mempengaruhi sifat pengolahan dan mutu hasil pulp/kertas tersebut.

Percobaan pengolahan pulp sulfat/kraft secara individu terhadap empat jenis kayu HTI (sengon, gmelina, meranti kuning, dan kapur) dilakukan pada kondisi tetap pemasakan: alkali aktif 16 persen, sulfiditas 22,5 persen, dan perbandingan kayu dengan larutan pemasak 1:4. Sedangkan suhu maksimum pemasakan bervariasi (170°C dan 175°C), masing-masing dipertahankan dalam 4 taraf waktu (0, 30, 60, dan 90 menit). Tingkat delignifikasi selama pemasakan hingga selesai ditelaah dengan faktor H, dan juga kaitannya dengan sifat pengolahan pulp dan sifat fisik/kekuatan pulp

Tingkat delignifikasi tertinggi hingga terendah terjadi pada jenis kayu gmelina, sengon, meranti, hingga kapur. Tingkat delignifikasi lebih dipengaruhi oleh perbandingan banyaknya inti siringil dengan inti vanilin (S/V) dalam lignin ($R^2 = 0.5972$), dari pada oleh berat jenis kayu ($R^2 = 0.5212$). Tingkat tersebut berkorelasi negatif dengan rendemen pulp total dan persentase pulp reject, dan positif dengan rendemen pulp tersaring. Pulp dengan rendemen pulp tersaring tinggi dengan persentase reject rendah berindikasi tingkat degradasi fraksi karbohidrat rendah dan tidak undercooked, dan ternyata menghasilkan lembaran pulp/kertas dengan sifat kekuatan tinggi; dan sebaliknya. Sifat fisik/kekuatan lembaran pulp dipengaruhi secara positif oleh perbandingan S/V dan secara negatif oleh berat jenis kayu.

Kata kunci: Kayu hutan tanaman, faktor H, tingkat delignifikasi, berat jenis, dan perbandingan S/V

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ABSTRACT

The limited availability of wood-fiber sources in Indonesia and the proposed encouragement to lessen their reliance on natural production forest for pulp/paper industries have inflicted serious concerns. As one solution is the establishment of industrial plantation forest (IPF) to supply wood fibers. The diversity in the IPF wood species can affect fiber qualities and hence pulp as well as its processing properties.

Individual kraft pulping was conducted on four IPF wood species (i.e. sengon, gmelina, meranti kuning, and kapur) at fixed cooking conditions: 16 % active alkali, 22.5 % sulfidity, wood to liquor ratio at 1:4. Maximum cooking temperature varied from 170°C to 175°C, each held for four durations (i.e. 0, 30, 60, and 90 minutes). The delignification extent (lignin removal action) was assessed using the H-factor, and also evaluated its possible association with pulp-processing and pulp strength properties.

Highest delignification extent until the lowest occurred at consecutively sengon, gmelina, meranti, and kapur wood species. The delignification extent was more affected by the ratio of syringil to vanillin units (S/V) in the lignin ($R^2 = 0.5972$) than by wood density ($R^2 = 0.5212$). Such extent correlated negatively with total pulp yield and pulp reject, and positively with screened pulp yield. Pulps with high screened yield and low pulp reject were indicatively associated with low carbohydrate degradation and fewer undercooked chips, thereby affording their high strength properties; and vice versa. Pulp strength was affected by S/V ratio positively, and wood density negatively.

Keywords: industrial plantation wood, H-factor, delignification extent, density, dan S/V ratio

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I. INTRODUCTION

The ever limited availability of wood-fiber sources along with the proposed encouragement to lessen their reliance on natural production forest has brought about serious concerns. As of this occasion, the Indonesia's forest destruction has proceeded at staggering rate, i.e. 2.87 million hectares (ha) per year. Consequently, the Indonesian government intends to gradually terminate the use of natural forest wood by all wood industries beginning 2004. Previously, the government has also issued the policy on ending wood supply from natural forest for pulp/paper industries that will be imposed in 2009. Accordingly, this situation has enforced the related wood-based entrepreneurs, including pulp/paper endeavors, to seek other wood sources which should be dependable, secure their sustainability, and at the same time fulfill their continual supply, among others: through the establishment of industrial plantation forest (IPF) as also urged by the government. In this regard, the wood supply from natural forest can expectedly be replaced in stages by the IPF (Anonym, 2006; and Kustiawan, 2006).

Until 2005, the realization of IPF establishment achieved 2.5 million ha with production about 20 – 22 million m³ of wood per year. From that area, roughly 1.4 million ha has been allocated for IPF pulp. Further, the targeted IPF area (including the IPF for pulp) can have reached 9 million ha by the year 2014, ensuring the wood supply over 53 million m³ of wood per ha (Anonim, 2002 and 2006). Such enhancement on the establishment of pulp IPF is also commensurate with the fact that the trend of Indonesia's paper consumption steadily increased from 2,783,430 tons in 1998 to 5,510,000 tons in 2005 (Anonim, 2003; and Mansur, 2006). Besides accelerating the IPF establishment, wood-based industries should impose alteration on their technology and production system. Particularly for pulp/paper industries, their implemented technology should adjust

themselves to the characteristics of the processed IPF wood species, thereby producing pulp/paper with appropriate yield and satisfactory qualities/properties.

There are several wood species reserved for the IPF establishment, among others (Sutigno and Pasaribu, 1990): sengon, gmelina, meranti kuning, and kapur. Those four species belong to tropical hardwood group, which among them can exhibit their considerably varying physical and chemical properties (Hoadley, 1990; and Martawijaya, *et. al.* 2005); and this could affect the implemented technology or processing system into pulp and hence the qualities of the resulting pulp/paper products. For the processing condition of pulp from wood or other lingo-cellulosic fibrous matters using chemical means, the desired results are those with extensive delignification extent (i.e. lignin removal action) and lowest carbohydrate degradation thereby affording high screen pulp yield, low pulp reject, and high physical/strength properties of pulp/paper as well. In addition, the implementation of appropriate pulping conditions allows more efficient uses of chemicals, additives, and energy (Casey, 1980).

Related with those narrations, the kraft chemical pulping on each of those four IPF's wood species has been experimentally carried out employing fixed and variable cooking conditions. The fixed conditions were active alkali, sulfidity, and wood to cooking liquor ratio. Meanwhile, the variable conditions were maximum temperatures, and the duration required to achieve each of those two temperatures from room temperature as well as the duration retained at those temperatures. Those varying cooking temperatures and durations were manipulated into single variables the so-called H-factors using Vroom method (Sjostrom, 1982). Further, the calculated H-factor values/levels were used to asses the delignification extent during the pulping of each IPF's wood species. Also assessed was the possible relation between the delignification extent and the pulping properties with the further details forthcoming.

II. MATERIALS AND METHODS

A. Materials and Equipment

The materials were four IPF's hardwood species, i.e. sengon (*Paraserianthes falcataria*), gmelina (*Gmelina arborea*), meranti (*Meranti* spp.), and kapur (*Dryobalanops aromatica*). Wood samples of the first two wood species were procured from Jatinangor area in Sumedang (West Java), while the latter two species from Berau region (East Kalimantan). Main and supplement chemicals used for this experiment were sodium hydroxide (NaOH), sodium sulfide (Na₂S), ethanol, benzene, sulfuric acid, chloroform, nitrobenzene, starch indicator, chloric acid, potassium permanganate, chloroform, and filter paper. Meanwhile, the laboratory equipment was among others: gas chromatography apparatus, soxhlet extractor, slotted screen, digester, handsheet former machine, analytical balance, graduated cylinders, beaker glasses, and instruments for the testing on physical/strength properties of pulp sheets.

B. Methods

1. Analysis on wood samples

Samples were prepared from each of four IPF's wood species for basic density determination and particular chemical analysis (i.e. lignin content) in accordance with the TAPPI procedures (Anonim, 1972). Lignin content determination was adopted from Klason method (Browning, 1967; and Moore and Johnson, 1967). The resulting Klason lignin was subsequently subjected to nitrobenzene oxidation to convert sinapyl alcohol monomers and coniferyl alcohol monomers in the lignin into consecutively syringaldehyde and vanillin (Figure 1). Further, the ratio between the number of syringaldehyde (syringil-type) units and the number of vanillin-type units could be figured out through gas chromatography analysis on the results of nitrobenzene oxidation, adopting the procedures of McNair dan Bonelli (Anonim, 1992). Gas chromatography apparatus of Perkin Elmer

Model 3920 was used with the specifications: BPX5 05-type column, N₂ carrier gas, flow rate at 0.4 ml per minute, 200°C – 240°C column temperature, and 300°C injection temperature.

2. Kraft pulp preparation followed with the related evaluation

Each of the four IPF's wood species was manually chipped with the size measuring about 3 cm in length, 2.25 cm in width, and 2 – 3 mm in thickness. The wood chips were aerated for some particular duration to reach their air-dry conditions, and then their moisture content determined in accordance with the TAPPI procedures (Anonim, 1972). The air-dry chips of each IPF's wood species were cooked into pulp by the kraft process in an electrically heated rotary stainless digester of 20-liter batch capacity. The fixed conditions were active alkali (16 %), sulfidity (22.5 %), the ratio by weight between wood chips to cooking liquor (1 : 4), and the raising rate of cooking temperature (1.580 °C per minute). Meanwhile, the variable conditions were maximum temperature (170°C and 175°C), and the overall cooking durations required to achieve each of those two temperatures from room temperature plus the durations retained at those temperatures (i.e. 0, 30, 60, and 90 minutes). As such, those overall durations, and the maximum temperatures (170°C and 175°C) were further manipulated using the Vroom method (Sjostrom, 1982) for the calculation of H-factor levels, in accordance with the formula:

$$\text{H-factor} = \int_0^t \exp(43.2 - 16,113 / T) dt, \text{-----} \text{-----} \quad (\text{I})$$

Where:

t = total cooking duration beginning from room temperature, ramping temperature until end of the keeping temperature

T = absolute cooking temperatures (in °K) at particular cooking duration (t), including the

room temperature where the cook starts, raising temperature, and keeping (maximum) temperature

After blowing down the digester which took about 30 minutes, the resulting kraft pulp was thoroughly washed and screened using a 0.25 mm slotted packer screen. The yield of the kraft pulp passing through the screen was determined (as screened pulp yield). The yield was also determined on the pulp before screening (as unscreened or total pulp yield). The determination of both total and screened yield was in accordance with the TAPPI procedures (Anonim, 1972). The pulp fraction retained on the screen could be determined by subtracting the total pulp yield with the screened yield (as pulp reject).

Subsequently, to evaluate how far or until what degree (stage) the kraft cooking on wood chips into pulp had achieved afterwards, evaluation was conducted on kappa number of the screened pulp and on residual (final) lignin content in unscreened pulp, consecutively according to the TAPPI procedures (Anonim, 1972). Results of such residual lignin determination taking into account the unscreened (total) pulp yield, were further used to calculate the residual/final lignin content as if in the original wood chips, following the kraft cooking. Afterwards, the delignification extent during the cooking was figured out merely as the ratio between the value of implemented H-factor and the corresponding residual/final lignin content in the wood chips.

With regard to the screened pulps, the ones considered satisfactory at one reasonable H-factor level, through the assessment on the screened yield, pulp reject, and delignification extent, were made directly (without beating and fibrillation action) into pulp sheets with the targeted 60 gram per m² basis weight, using handsheet former machine. Following the conditioning in the room with particular temperature and relative humidity, the resulting

pulp sheets were examined of their real basis weight, and strength properties (tear factor and breaking length) in accordance with the TAPPI procedures (Anonim, 1983).

C. Data Analysis

To evaluate the implementation of the H-factors at various levels (Table 1) on the kraft pulp-processing properties (e.g. screened pulp yield, kappa number, residual lignin content) from each of the four IPF's wood species, completely randomized design was adopted with factorial pattern and split-plot hierarchy. As the factor in the main plot was the IPF's wood species, while the H-factor allocated in the secondary plot. On the other hand, the data analysis on the physical/strength properties of pulp sheets (real basis weight, tear factor, and breaking length) whereby they are examined only at one particular H-factor, a completely randomized design was used with single factor (simple CRD). The factor was merely the IPF's wood species. Further, each level of the combined factors between the IPF species and H-factor either in the split-plot design or in simple CRD was replicated three times throughout the experimented kraft cooking (pulping). Also evaluated were the possible relations of kraft delignification extent (lignin removal action) with pulp processing properties and with physical/strength properties of pulp sheet, using correlation assessment (coefficients of correlation and determination).

III. RESULTS AND DISCUSSION

A. Wood Properties

The examined properties of four IPF's wood species covered basic density, lignin content, and the ratio between syringil units and vanilin units in the lignin (Table 1).

Table 1. Basic density, lignin content, and ratio of syringaldehyde to vanillin units in the lignin
 Tabel 1. Kerapatan dasar, kadar lignin, dan perbandingan banyaknya inti siringil dengan inti vanilin dalam lignin

No	Wood species (Jenis kayu)	Basic density (Kerapatan dasar) gram/cm ³	Lignin content (Kadar lignin) %	S/V ^{*)}
1	Sengon (<i>Paraserianthes falcataria</i> (L) Nielsen)	0.45	26.72	2.03
2	Gmelina (<i>Gmelina arborea</i> Roxb)	0.48	25.50	2.02
3	Meranti kuning (<i>Shorea</i> spp.)	0.57	24.89	1.87
4	Kapur (<i>Dryobalanops</i> spp.)	0.62	26.40	1.30

^{*)} Ratio in the number of syringilaldehyde to vanillin units (*Perbandingan antara banyaknya inti siringil dengan inti vanilin dalam lignin*); for the structural formula of syringaldehyde (syringil-type) and vanillin-type units, please refer to Figure 1 (*rincian rumus bangun siringil dan vanilin disajikan pada Gambar 1*).



Figure 1. Structural formulae of syringaldehyde (S) and vanillin-type (V) ^{*)}
 Gambar 1. Rumus bangun persenyawaan siringil (S) dan vanilin (V) ^{*)}

Remarks (*Keterangan*): Syringaldehyde (syringil-type) and vanillin-type units are derived from their precursors phenyl propane monomers that exist in the structural lignin polymer, i.e. consecutively coniferyl alcohol and sinapyl alcohol, through nitrobenzene oxidation (refer to Figure 2) / *Siringil dan vanilin merupakan turunan dari monomer utama fenil propan pada polimer lignin, yaitu koniferil alcohol dan sinapil alcohol, melalui oksidasi nitrobenzene (lihat Gambar 2)*

^{*)} Source (*Sumber*): Sjoström (1982)

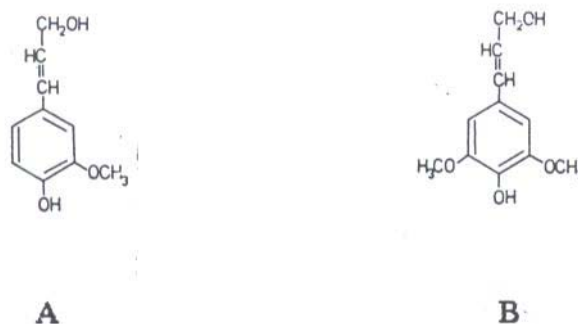


Figure 2. Precursor monomers that make up lignin polymer (phenyl propane monomers), i.e. among others: coniferyl alcohol or vanillin-type unit (A) and sinapyl alcohol or syringil-type unit (B) ^{*)}

Gambar 2. Monomer utama yang membentuk polimer lignin (monomer fenil-propane), yaitu diantaranya: koniferil alcohol atau tipe vanilin (A) dan sinapil alcohol atau tipe siringil (B) ^{*)}

*) Source (Sumber): Sjostrom (1982)

B. Calculation of the H-factor

The brief results of calculating the H-factor as a means of expressing cooking temperature and time (duration) as a single variable during the kraft pulping are presented in Table 2. The greater value of H-factor level implies the more severe (intense) kraft cooking condition, and vice versa.

Table 2. Brief details of calculating the H-factor in the kraft cooking (pupling) process
Tabel 2. Rincian hasil penghitungan faktor H dalam proses pemasakan sulfat

Temp °C	$t_{(Tr \rightarrow Tm)}$ minutes (menit)	t_{TM} minutes (menit)	t_{Tot} minutes (menit)	H-factor (Faktor H) *)
170	90.00	0.00	90.00	117.88
170	90.00	30.00	120.00	579.34
170	90.00	60.00	150.00	1040.81
170	90.00	90.00	180.00	1502.25
175	93.15	0.00	93.15	173.87
175	93.15	30.00	123.15	866.56
175	93.15	60.00	153.15	1559.25
175	93.15	90.00	183.15	2182.67

Remarks (Keterangan):

Temp = maximum cooking temperature (*suhu maksimum pemasakan*)

$t_{(Tr \rightarrow Tm)}$ = the duration that took from the room temperature raising to maximum cooking temperature (*waktu yang dibutuhkan dari suhu kamar hingga meningkat menjadi suhu maksimum pemasakan*)

t_{TM} = the duration at maximum cooking temperature (*waktu pada suhu maksimum pemasakan*)

t_{Tot} = sum of: $t_{(Tr \rightarrow Tm)}$ + t_{TM} (*jumlah dari: $t_{(Tr \rightarrow Tm)}$ + t_{TM}*)

*) Calculated using Vroom formula (Sjostrom, 1982), refer to equation I / *Dihitung menggunakan rumus Vroom (Sjostrom, 1982), lihat persamaan I*

C. Kraft Pulping Properties

Analysis of variance revealed that the changes in H factor significantly affected the overall kraft pulping properties (Table 3). However, the H-factor effect varied with wood species (Table 4). Nevertheless, there were similar trends for particular properties in that the increase in H-factor (i.e. more intense cooking condition) brought about the decrease in total (unscreened) pulp yield, pulp reject, kappa number in the screened pulp, residual/final lignin content in unscreened pulp, and final lignin content in unscreened pulp based on the

original wood (its oven dry weight); and concurrently the increase in the extent of delignification (lignin removal action). These trends were confirmed through the assessment on the score values resulting from the manipulation of honestly significant difference (HSD) test (Table 4). The phenomena that occurred are attributable to the selective reaction of kraft cooking liquor with the wood lignin rendering it more soluble in the liquor and concurrently leaving behind less lignin in the resulting pulp. About the decrease in pulp-reject percentage, this could also be due to more dissolution of lignin with the more intense cooking condition, thereby inflicting more complete separation of fiber bundles and defiberation of uncooked wood particles (Casey, 1980; and Siagian, *et. al.*, 2004). Further, the more vigorous lignin dissolution and fragmentation as such at higher H-factor was explicitly associated with the increasing extent of delignification (Table 4). As the relevance, it was shown that the extent of delignification was negatively correlated with the total pulp yield (Figure 3) and with the pulp reject (Figure 4)

With respect to the screened pulp yield, for sengon and gmelina wood species, it tended to increase with the H-factor (Table 4). For sengon species, the pulp yield steadily unchanged at the H-factor greater than 1502.25. Meanwhile, for gmelina species, the maximum yield achieved at 1502.25 H-factor, beyond which it decreased. For meranti kuning and kapur wood species, however, the increase in H-factor brought about the decrease in their screened pulp yield. The increase in screened yield of sengon and gmelina pulps was closely and positively associated with the more delignification extent that further inflicted more separation of fiber bundles and shives (Figure 5). On the other hand, the decrease in the yield of gmelina pulp at the H-factor beyond 1502.25, and in the yield of meranti kuning pulp and kapur pulp at the overall H-factor (117.88 – 2182.67) indicated that the more intense cooking condition particularly (i.e. increasing H-factor) for meranti kuning and kapur wood species not only brought about more dissolution of lignin, but also

induced more degradation on carbohydrate fractions (i.e. cellulose and hemicellulose) in those wood species.

Through the assessment on H-factor particularly at 1502.25 or greater, it turned out that the residual lignin content in the unscreened pulp (based on the dry weight of original wood) and the delignification extent following the kraft cooking for sengon and gmelina wood species were consecutively smaller and less severe than those for meranti kuning and kapur species, as confirmed through the HSD test (Table 4). This indicated that lignin removal, as progressively associated with the delignification extent, in the latter two species was more difficult than in the former species. Such phenomena that occurred were apparently more attributable to the ratio between syringaldehyde (syringil-type) units and vanillin-type units (S/V) in the lignin rather than to the initial lignin content and density of the related wood species (Table 1). This is because, when linked to the initial lignin contents, although the variation in the contents seemed obvious across the different wood species there occurred no definite trend with the delignification extent. Still related, the delignification extent correlated more strongly with ratio of S/V in lignin ($R^2 = 0.5972$; R^2 partial = 0.4812) than with its corresponding wood density ($R^2 = 0.5212$ and R^2 partial = 0.4126) as illustrated in Figures 6 and 7, respectively. This was explicable that although varying wood density can affect its permeability to the pulping liquor, this was not a problematic case for the kraft cooking/process, since it has been proved applicable on a wide range of tropical wood species with various characteristics (Siagian, *et. al.* 2004). In addition, the strongly alkaline liquor during the kraft cooking diffuses at nearly equal rate in longitudinal, radial, and tangential directions of the cooked wood chips (Browning, 1967). In all, this clarified that the delignification extent (lignin removal action) as observed in this experiment seemed less affected by wood density and its initial lignin content, but could be more apparently blamed on the S/V ratio.

As evidence for such, it revealed that wood species with higher S/V ratio in their lignin, i.e. sengon and gmelina, tended to sustain greater delignification extent or more intense lignin removal than those with lower S/V ratio, i.e. meranti kuning and kapur species (Tables 1 and 4, and Figure 6). The explicable indication are that lignin with greater S/V ratio allows more possibility occurring for reaction mechanisms I (Figure 8), and less possibility for reaction mechanisms II (Figure 9).

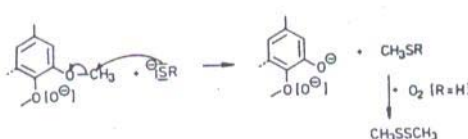


Figure 8. Reaction mechanisms I in which the syringil-type monomer units in the lignin entities during the kraft process are partially demethylated forming more soluble lignin fragments, since this structure contains phenolate ions (Sjostrom, 1982)
Gambar 8. Mekanisme reaksi I dimana monomer tipe siringil dalam lignin selama proses pemasakan sulfat mengalami demetilasi sehingga membentuk senyawa yang mudah larut, karena terdapatnya ion fenolat dalam struktur ini (Sjostrom, 1982)

In the reaction mechanisms I, sinapyl alcohol monomers (as syringil-type unit) in the lignin entities during the kraft cooking are partially demethylated by the action of hydrosulfide (HS^-) ions at C-5 and C-3 positions forming more soluble fragments in the cooking liquor (Sjostrom, 1982). This mechanisms reaction in the lignin entities will be more intense at the higher S/V ratio or with the fewer number of coniferyl alcohol monomers (vanillin-type units) which only have C-3 positions, thereby intensifying the delignification extent (lignin removal) as for the case of sengon and gmelina wood species; and vice versa (meranti kuning and kapur woods).

On the other hand, lignin entities with lower S/V ratio or greater number of vanillin-type monomer units, as for case with meranti kuning and kapur wood species (Table 1), will inflict more possibility of reaction mechanism II (Figure 9), in which condensation process occurs at the unoccupied C-5 position of this monomer type, forming larger molecular-

weight fragments, thereby becoming less soluble and hence inhibiting lignin removal (i.e. less intense delignification). However, this reaction mechanism and its negative consequences as such will occur less frequently in the lignin fragments with higher S/V ratio or greater number of syringil-type monomer units, as for the case with sengon and gmelina species (Table 1).

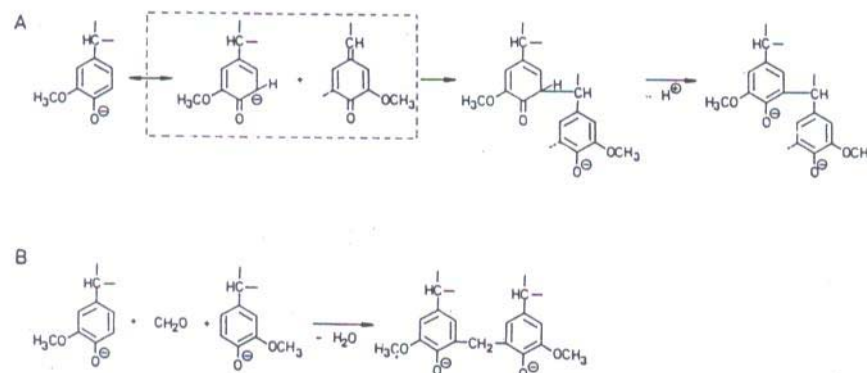


Figure 9. Condensation reactions (A and B) that can occur at the unoccupied C-5 positions of the vanillin-type monomers in the lignin fragments during the kraft process forming into less soluble compounds (Casey, 1980).

Gambar 9. Reaksi kondensasi (A dan B) yang terjadi pada posisi kosong C-5 monomer tipe vanilin dalam lignin selama proses pemasakan sulfat, sehingga terbentuk senyawa yang lebih sukar larut

Still related, it is necessary to draw thorough attention that native lignin (i.e. natural lignin in wood or other lingo-cellulosic fibrous matters), which has not sustained changes due to chemical action or other chemically altering treatments) can contain several types of linkages between its phenyl propane monomer units, among others biphenyl linkages or C5 - C5 bond type (Figure 10). The number of these C5 - C5 bonds will be fewer in the native lignin with higher S/V ratio or contains greater portions of syringil-type monomers particularly for the case of sengon and gmelina woods; and vice versa (i.e. meranti kuning and kapur woods). The C5 - C5 bonds are difficult to break apart by chemical action particularly during the kraft process rendering the lignin less soluble (Casey, 1980), and therefore their existence in great number as indicated in meranti kuning and kapur woods can also retard the lignin removal or decrease the delignification extent.

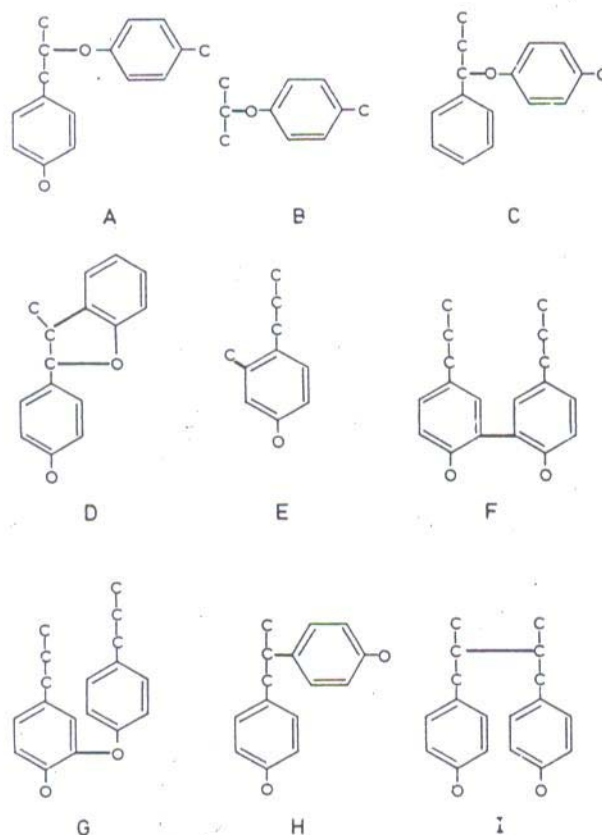


Figure 10. The most common linkages between phenyl propane monomers in the native lignin (Casey, 1980).

Gambar 10. Ikatan yang umum terdapat antara monomer fenil-propan dalam lignin yang belum mengalami perlakuan yang mengakibatkan perubahan kimia (Casey, 1980)

Legend (Keterangan): A = Arylglycerol- β -aryl ether; B = Glycerolaldehyde-2-aryl ether; C = Noncyclic benzyl-aryl ether; D = Phenylcoumaran; E = Structures condensed in 2- or 6-position / Struktur yang terkondensasi pada posisi 2 atau 6; F = Biphenyl (5-5 bond) / (Ikatan 5-5); G = Diaryl ether; H = 1,2-Diarylpropane; and I = β - β -linked structure

To infer, the extent of reaction mechanisms I (Figure 8) and II (Figure 9) that occurred in the kraft cooking of a particular wood species itself is affected by the S/V ratio in its lignin. In addition, the proportion C5-C5 bonds of the overall linkage types between phenyl-propane units in the lignin (Figure 10) can also be associated with the S/V ratio. All those phenomena (Figures 8, 9, and 10) can hence affect the delignification extent or lignin removal action. As previously described, the lower the S/V ratio in the lignin, i.e. for the

case with meranti kuning and kapur wood species, then the more difficult or more retarded the lignin removal during the chemical pulping (e.g. kraft process); and vice versa (i.e. sengon and gmelina species). Such retardation during the kraft pulping of particular wood species, notably under severe or harsh cooking condition (e.g. greater H-factor) will inflict the more fragmentation/solubilization of the lignin in the cooking liquor, but concurrently also render the carbohydrate fraction (i.e. cellulose and hemicellulosa) more liable to depolymerization or degradation. As a result, not only was the total pulp yield and percentage of pulp reject decreased, but also the screened pulp yield declined as experimentally occurred in the kraft pulping of meranti kuning and kapur wood species (i.e. (Table 4 and Figures 5). Particularly interesting for their pulp reject, it tended to decline with the H-factor raising up to 1502 and afterwards increase. Those phenomena could be blamed again on the possible condensation reaction between lignin fragments (Figure 10) thereby inflicting lignin agglomeration and hence causing difficulty in the separation of the lignin-bonded fiber bundles

D. Physical and Strength Properties of the Kraft Pulp Sheet

The testing on physical and strength properties of the kraft pulp sheets (i.e. real basis weight, tear factor, and breaking length) from four IPF's wood species was carried out at only one particular H-factor (i.e. 1502.25). Such decision was taken by observing that two of the four wood species (sengon and gmelina) with the kraft cooking at 1502.25 H-factor could achieve the most satisfactory results with respect to the high screen pulp yields and practically low pulp rejects, as confirmed through the statistical HSD test (Table 4). It turned out that the higher the screened pulp yield and the lower the pulp reject, then the greater the real basis weight and physical/strength properties of the corresponding pulp sheet. It revealed also, through the analysis of variance (Table 5) and the subsequent HSD test (Table 6), that the highest real basis weight and highest strength properties (i.e. tear

factor and breaking length) of the pulp sheet were afforded by sengon wood species, followed in decreasing order by the species of gmelina, meranti kuning, and kapur. Such trend was interestingly associated as well with the decrease in the S/V ratio in the lignin of each of those four wood species (Table 1). Again, this could strengthen the suspect of possible severe degradation on the carbohydrate fractions in the wood fibers during the kraft pulping that further accounted for the lower physical/strength properties of the resulting pulp particularly from meranti kuning and kapur wood species; and vice versa (for sengon and gmelina pulp).

This degradation could produce the pulp with more fines and other minute fractions (particularly from meranti kuning and kapur species) thereby inflicting considerable loss during pulp-sheet forming and hence lowering the real basis weight far below the targeted 60 gram per m² basis weight (particularly as well for meranti kuning and kapur pulp sheets), and also physically weaken the entity of the fibers themselves consequently decreasing the pulp strength. Further, it is also noteworthy that such decrease in strength properties seems also related but negatively with basic density of the experimented wood species (Tables 1 and 6). This phenomena is traceable to the generally accepted theory that wood species with low density are related with thinner fiber wall that can afford more conformability and bonding ability of the fibers, thereby increasing the pulp strength properties, and vice versa. Therefore, it could be inferred that in this regard both the S/V ratio and wood density could explicably affect the pulp strength properties.

E. Highlights of the Implemented H-factor Associated with the Delignification Extent

The implementation of the H-factor (173.35 – 2182.67) in the kraft pulping on four IPF's wood species wood seemed responsible or accounting for the delignification extent (lignin removal action). Such extent or action, however, varied with the wood species at a given H-factor. Delignification or lignin removal in the pulping of meranti kuning and

kapur wood species was apparently more difficult than that of sengon and gmelina species, thereby inflicting negative effects on the pulping and pulp properties of the former species. Those phenomena were explicably attributed also to higher S/V ratio in the lignin of the latter wood species, and conversely to lower S/V ratio of the former species.

Results of this H-factor-related experiment suggested their less applicability for the kraft pulping process of meranti kuning and gmelina wood species at varied H-factor and other fixed cooking conditions (i.e. alkali active concentration, sulfidity, and wood to cooking liquor ratio) than that of sengon and gmelina species. This is because for the latter wood species, the kraft process could achieve optimum H-factor (1502.28) with convenient or positive properties of the processing results. Conversely, for the former species, the process suspiciously inflicted marked degradation on wood carbohydrate fractions and induced more condensation of lignin fragments, thereby exerting negative result properties.

For the kraft pulping (cooking) on meranti kuning and kapur wood species, the unsatisfactory results can expectedly be improved by altering cooking conditions, among others adjusting the sulfidity. This is because the sulfidity is associated with the formation of hydrosulfide (HS^-) ions in the cooking liquor, which further can more intensively inhibit or cope with the possible lignin-condensation mechanisms particularly during the kraft pulping of those wood species (Figure 9), on which the low S/V ratio in their lignin could be blamed (Table 1).

IV. CONCLUSIONS AND SUGGESTIONS

Explicability of the varying H-factors in the individual kraft cooking/pulping on four industrial plantation forest's wood species (i.e. sengon, gmelina, meranti kuning, and kapur) as suspiciously accounted for their delignification extent or lignin removal action, also under other particular fixed cooking conditions (i.e. 16 % active alkali, 22.5 % sulfidity, and

wood to cooking liquor ratio at 1 : 4), had been experimented and came up with significant highlights (e.g. quite specific for the particular pulped wood species):

The H-factor as implemented was basically a mean to express varying cooking temperature (in this experiment reaching the maximum: 170°C and 175°C) and overall cooking durations (the time required to achieve each of those two maximum temperatures plus the times allocated at each temperature, i.e. 0, 30, 60, and 90 minutes) as single variable (so-called the H-factor itself). As such, several values/levels of H-factors were acquired using Vroom method (i.e. 117.88, 173.87, 579.34, 866.561, 1040.83, 1502.25, 1559.25, and 2182.67), which implied the higher the factors then the more severe or intense the kraft cooking condition.

It turned out that the H-factors significantly accounted for the lignin-removal action. The action was more intensified or severe with the greater H-factor, as shown by the decreases in total pulp yield, pulp reject, kappa number, and residual lignin content, and the increase in the delignification extent; and vice versa.

The lignin-removal action (delignification extent), however, varied with the wood species at a given H-factor. Delignification or lignin removal in the pulping of meranti kuning and kapur wood species was apparently more difficult than that of sengon and gmelina species, thereby inflicting negative effects on the pulping and pulp properties of the former species. For the pulping properties, the kraft cooking on sengon and gmelina wood species at greater H-factor brought about the increase in the screened pulp yield up to particular extent, and the decrease in the pulp reject. However, for the case of meranti kuning and kapur species, greater H-factor induced the decrease in screened pulp yield and to some extent pulp reject. About pulp properties (i.e. real basis weight, tear factor, and breaking length), the highest one was achieved by sengon pulp, followed in decreasing order by gmelina, meranti kuning, and kapur pulp

Those phenomena were explicably associated with the higher ratio of syringil-type to vanillin-type units (S/V) in the lignin of the sengon and gmelina wood species, and conversely to lower S/V ratio in the lignin of meranti kuning and kapur species. The role of S/V ratio at a given H-factor seemed more pronounced on the lignin removal action (delignification) than that of wood density and its initial lignin content. As evidence, the S/V ratio correlated strongly and positively with the delignification extent ($R^2 = 0.5972^*$; and R^2 partial = 0.4812^*). Concomitant with such, positive correlation occurred between the delignification extent (lignin-removal action) and the screened pulp yield ($R = +0.758^*$). Meanwhile, delignification extent correlated negatively with the total pulp yield ($R = -0.923^{**}$) and with the pulp reject ($R = -0.776^*$). On the other hand, about the physical and strength properties pulp (real basis weight, tear factor, and breaking length), they seemed obviously affected by both the S/V ratio (positively) and wood density (negatively).

Results of this H-factor-related experiment implied their less applicability for the kraft pulping process of meranti kuning and gmelina wood species at varied H-factor and other fixed kraft cooking conditions (i.e. alkali active concentration, sulfidity, and wood to cooking liquor ratio) than that of sengon and gmelina species. In order to be more applicable for the former two species, their unsatisfactory results can expectedly be improved by altering the kraft-cooking conditions, among others by adjusting the sulfidity in addition to the H-factors.

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LEMBAR ABSTRAK:**Penerapan Faktor-H untuk Menelaah Tingkat Delignifikasi dan Sifat Pulp Empat Jenis Kayu Hutan Tanaman Industri pada Proses Pengolahan Kimia Sulfat**

Oleh

Han Roliadi ¹⁾ & Noor Rahmawati ²⁾**ABSTRAK**

Percobaan pengolahan pulp sulfat/kraft secara individu dilakukan terhadap empat jenis kayu hutan tanaman industri (sengon, gmelina, meranti kuning, dan kapur).

Tingkat delignifikasi tertinggi hingga terendah terjadi berturut-turut pada jenis kayu gmelina, sengon, meranti, hingga kapur. Tingkat delignifikasi lebih dipengaruhi oleh perbandingan banyaknya inti siringil dengan inti vanilin (S/V) dalam lignin, dari pada oleh berat jenis kayu. Tingkat tersebut berkorelasi negatif dengan rendemen pulp total dan persentase pulp *reject*, dan positif dengan rendemen pulp tersaring. Pulp dengan rendemen pulp tersaring tinggi dengan persentase *reject* rendah berindikasi tingkat degradasi fraksi karbohidrat rendah dan tidak *undercooked*, dan ternyata menghasilkan lembaran pulp/kertas dengan sifat kekuatan tinggi; dan sebaliknya. Sifat kekuatan pulp dipengaruhi secara positif oleh perbandingan S/V dan secara negatif oleh berat jenis kayu.

Kata kunci: Kayu hutan tanaman, faktor H, tingkat delignifikasi, berat jenis, dan perbandingan S/V

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ABSTRACT SHEET:***Explicability of the H-Factor to Account for the Delignification Extent and Properties of Plantation Forest Wood Pulp in the Kraft Cooking Process***

By

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ABSTRAK**ABSTRACT**

Individual kraft pulping was conducted on four industrial plantation forest's wood species (i.e. sengon, gmelina, meranti kuning, and kapur).

The highest delignification extent until the lowest occurred at consecutively sengon, gmelina, meranti, and kapur wood species. The delignification extent (lignin-removal action) was more affected by the ratio of syringil to vanillin units (S/V) in the lignin than by wood density. Such extent correlated negatively with total pulp yield and pulp reject, and positively with screened pulp yield. Pulps with high screened yield and low pulp reject were indicatively associated with low carbohydrate degradation and fewer undercooked chips, thereby affording high strength properties; and vice versa. Pulp strength was affected by S/V ratio positively, and wood density negatively.

Keywords: Industrial plantation wood, H-factor, delignification extent, density, dan S/V ratio

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