# EFFECT OF LIGNIN ON ACACIA MANGIUM KRAFT PULP REFINING BEHAVIOUR

## W. D. WAN ROSLI, I. MAZLAN and K. N. LAW\*

School of Industrial Technology, University Sains Malaysia, 11800 Penang, Malaysia

\*Centre Intégré en Pâtes et Papiers, Université du Québec à Trois-Rivières, P.O. Box 500, Trois-Rivières, Québec, Canada G9A 5H7

Received September 27, 2010

The beating responses of 26 kraft pulps from *Acacia mangium* were investigated for the effect of their chemical composition on paper characteristics. It was observed that the different lignin content (Kappa number) of the pulps over a yield range of 40-50% was a determining factor, defining their beating behaviour at PFI refining. The pulps with kappa number below 25 behaved differently from those above 25, which could be used when selecting the optimum pulping conditions to obtain pulp with good yield and good papermaking properties.

Keywords: Acacia mangium, kraft pulp, kappa number, PFI refining, pulp properties

# **INTRODUCTION**

In a recent study,<sup>1</sup> the effects of pulping variables on the pulp and paper properties of kraft pulp produced from Acacia mangium were evaluated. The characteristics of this species<sup>2-6</sup> and its importance as a raw material for pulp and paper production<sup>7-14</sup> have been discussed. The objective of this study, a continuation of the authors' research on the quality of A. mangium chemical pulps, was to examine the beating responses of A. mangium kraft pulps over the 40-50% yield range. The pulps under study were prepared employing a wide range of pulping parameters,<sup>1</sup> permitting to examine the influence of Kappa number on the beating response of pulps. Such information is of special importance for pulp producers interested in optimizing their pulping processes and product quality.

Beating of pulp is a mechanical treatment producing physico-chemical changes in fibres through external and internal fibrillation of the cell wall.<sup>15</sup> The external fibrillation process breaks down the outer cell wall layer, which restricts the swelling of the cell wall material and fibrillates the S<sub>2</sub> layer of the secondary wall, resulting in an increased specific surface area for inter-fibre bonding through hydrogen bonds.<sup>16</sup> In addition to external fibrillation, beating also delaminates the cell wall lamellae (internal fibrillation), increasing the fibre flexibility, swellability and water uptake capacity and, consequently, sheet density.<sup>17-24</sup> These changes enhance the physical properties of paper, such as dry-sheet tensile strength, wet-sheet strength and strength of the rewetted sheet.<sup>16,25,26</sup> Other effects of beating include reduction in fibre length and coarseness, and generation of fines.<sup>27</sup>

The consequence of refining or beating is affected by the equipment used. It was reported that PFI mill beating produced different responses, compared to refining in an Escher-Wyss refiner; pulps refined at low intensities gave higher sheet tensile strength.<sup>28</sup> McKenzie<sup>29</sup> reported that the behaviour of a PFI mill equipped with stainless steel beating elements differs in some respects from that of a similar mill with bronze tackle. The beating responses are also affected by the treatment conditions, such as consistency, clearance between rotors and differential speed.<sup>30-33</sup>

## EXPERIMENTAL Materials

Two 14-year old *Acacia mangium* trees were harvested from Byram Forest Reserves, Penang, Malaysia. One had a diameter of 22.5 at breast height, and the other one – of 24.0 cm. The logs (1.5 m in length) were debarked and sawn into rods of approximately 5 cm x 5 cm x 1.5 m. They were then chipped and screened to remove the oversized particles; the average accepted chip size was ca. 23 mm, 22 mm and 6 mm in length, width and thickness, respectively.<sup>1</sup>

# **Pulping methods**

All pulping trials were carried out in a 4 L stationary stainless steel digester (NAC Autoclave Co. Ltd., Japan) fitted with a computer-controlled thermocouple. The experiments were conducted in accordance with a design matrix based on the Central Composite Design (CCD) technique. The experimental design matrix with both coded and real variables is shown in Table 1. The experimental parameters included active alkalinity, sulfidity, temperature and reaction time. For other details, see Wan Rosli  $et al.^{1}$ 

# Beating

Pulp beating was conducted at 10% consistency in a PFI mill, according to the Tappi method T248 wd-97 (TAPPI Standards Atlanta: TAPPI Press, 2002). The number of revolutions selected was 2000 (2K), 4000 (4K) and 6000 (6K). The unbeaten samples were designated as OK. Latency of the beaten samples was removed by shear-disintegration in hot water (≈95 °C), in a laboratory disintegrator. before standard handsheets of 60 g/m<sup>2</sup> were formed. The handsheets were conditioned at 23 °C and 50% RH for at least 24 h before testing, according to appropriate Tappi standard methods (TAPPI Standards, Atlanta, TAPPI Press, 2002).

Table 1	
Experimental design for Kraft pulping of A.	mangium

Exp.	Code	Active alkali,	Sulfidity,	Temperature,	Time,	Total yield,	Kappa
No.		$(\%Na_2O)$	(%Na <sub>2</sub> O)	°C	min	%	No.
1	-1,-1,-1,-1	15.5	20	165	105	48.8	38.9
2	1,-1,-1,-1	20.5	20	165	105	45.2	19.7
3	-1, 1,-1,-1	15.5	30	165	105	49.7	36.6
4	1, 1,-1,-1	20.5	30	165	105	46	19
5	-1,-1, 1,-1	15.5	20	175	105	46.1	25.6
6	1,-1, 1,-1	20.5	20	175	105	42.5	13
7	-1, 1, 1,-1	15.5	30	175	105	47	24.8
8	1, 1, 1,-1	20.5	30	175	105	43.3	13.6
9	-1,-1,-1, 1	15.5	20	165	135	48.5	32.9
10	1,-1,-1, 1	20.5	20	165	135	44.8	15
11	-1, 1,-1, 1	15.5	30	165	135	49.3	33.6
12	1, 1,-1, 1	20.5	30	165	135	46.7	17.4
13	-1,-1, 1, 1	15.5	20	175	135	45.8	21.7
14	1, -1, 1, 1	20.5	20	175	135	42.1	10.5
15	-1, 1, 1, 1	15.5	30	175	135	46.6	23.9
16	1, 1, 1, 1	20.5	30	175	135	43	14.2
17	-2, 0, 0, 0	13.0	25	170	120	50	44.6
18	2, 0, 0, 0	23.0	25	170	120	42.2	15.7
19	0,-2, 0, 0	18.0	15	170	120	45.1	17.8
20	0, 2, 0, 0	18.0	35	170	120	46.7	19.3
21	0, 0,-2, 0	18.0	25	160	120	48.6	30
22	0, 0, 2, 0	18.0	25	180	120	43.2	13.6
23	0,0,0,-2	18.0	25	170	90	46.2	22.3
24	0,0,0,2	18.0	25	170	150	45.5	16.8
25	0, 0, 0, 0	18.0	25	170	120	45.9	20.1
26	0, 0, 0, 0	18.0	25	170	120	46	20.2

# **RESULTS AND DISCUSSION**

The purpose of beating or refining pulp fibres is to improve the conformability of individual fibres in sheet consolidation, increasing the bonding surface between fibres in the fibrous network, and thus enhancing sheet strength. The effects can be brought about through mechanical treatment (*e.g.*, beating), the main purpose of which is to flexibilize the fibre wall structure by external and internal (delamination) fibrillation. The former creates fines and fibrils, thus augmenting the specific surface area, which is beneficial for inter-fibre bonding, while the latter improves fibre flexibility and water holding capacity, both being also beneficial for sheet consolidation. However, beating has undesirable effects, such as fibre shortening and creation of axial microcompression in the fibre wall. Fibre cutting or reduction in fibre length could negatively influence the paper strength values, such as tensile index and tearing resistance. Axial microcompression of the fibre wall would weaken intrinsic fibre strength, thus reducing paper strength. Understandably, the conditions used in producing pulp fibres could have significant impacts on all these phenomena, that is the beating behaviour of pulps.

## **Chemical characteristics**

As shown in Figure 1, the total pulp yield and Kappa number of the various pulps studied were highly correlated ( $\mathbb{R}^2 = 0.885$ ), which means that a higher yield corresponds to a higher lignin content. The chemical nature of pulp has a significant effect on the physical properties of fibres and on their response to beating. Although pulp yield is a good indicator of the chemical composition of fibres, the determination of the Kappa number (degree of delignification or residual lignin content) of pulp is a more precise assessment.

# Effect of Kappa number on pulp properties

The effect of Kappa number on pulp properties is illustrated in Figures 2 to 10. A total number of 26 pulps, studied over the 40-50% pulp yield range, were beaten in a PFI mill at 2000 (2K), 4000 (4K) and 6000 (6K) revolutions.







Figure 3: Sheet density vs. Kappa number

#### Freeness

As seen in Figure 2, the different pulps with a Kappa number of approximately 25 or lower, corresponding to a pulp yield in the range of 42-47%, reached a freeness of 500 to 400 mL at different levels of beating, compared to the initial freeness (OK) - of around 600 mL. The decrease of freeness for these pulps was proportional to the OK freeness, which was almost the same for all Kappa numbers. However, pulps with Kappa numbers above 25 exhibited a different pattern in their response to beating at the same levels of revolutions from initial freeness values above 600 to 400-550 CSF, following the same pattern of the OK freeness. Thus, the first set of pulps beats faster and to a lower freeness degree compared to those with a higher lignin content - rendering the fibres coarser and more resistant to mechanical action, and hinders their fibrillation.

## Sheet density

Generally, the pulps with low Kappa number (about  $\leq 25$ ) exhibited higher sheet density, compared to those with a Kappa number above 25 (Fig. 3). This suggested that, for the same energy consumption, pulps with a higher Kappa number behaved differently in response to beating than those with low Kappa numbers, indicating the important role of the pulp lignin content on the paper properties.



Figure 2: Freeness vs. Kappa number



Figure 4: Tensile index vs. Kappa number



Figure 5: Burst index vs. Kappa number

# Tensile and burst indices

Similar to sheet density, responses to beating were observed for tensile and burst indices (Figs. 4 and 5). Once again, a Kappa number  $\leq 25$  seemed to be a threshold delineating two sets of pulp properties. The tensile and burst indices tended to reach lower values under the same beating conditions, as the Kappa number increased above 25, compared to the pulps with lower Kappa numbers, which were in the same value range, despite the variation in Kappa numbers.

# Tear index

The effect of Kappa number on the tear index (Fig. 6) development under beating was much less pronounced compared to the other properties discussed above (Figs. 2-5). This difference is due to the fact that the tear resistance of a handsheet is mainly dependent on fibre length and thickness rather than on fibre bonding strength, as in the case of tensile and burst resistances. As seen in Figure 6, moderate beating at 2000-6000 revolutions gave a maximum of 9 mN  $m^2/g$  tear index for both sets of beaten pulps, with only slight differences, indicating the need of longer beating for the development of this property, which, after reaching a maximum, usually starts to decrease.



Figure 7: Tensile index vs. freeness



Figure 6: Tear index vs. Kappa number

## **Tensile-freeness relation**

As shown in Figures 7 and 8, the tensile indexes of the studied pulps were highly associated with both freeness ( $R^2 = 0.943$ ) and sheet density ( $R^2 = 0.964$ ). Freeness is a measurement of the drainage resistance of a fibrous mat. The more flexible fibres conform better in a mat formation, compared to the less flexible ones, thus forming more compact sheets or higher density ones, with higher drainage resistance or lower freeness. Moreover, the flexible fibres provide a larger inter-fibre contact surface, and consequently, improve tensile strength.

# The tensile-tear relation

The tensile tear relationship is supposed to indicate to what extent inter-fibre bonding strength affects tear resistance at a given beating level, as it is responsible for the resistance of the fibres against being pulled out of the fibrous sheet structure. As illustrated in Figure 9, a strong correlation  $(\mathbf{R}^2 = 0.816)$  is exhibited between tensile and tear resistances, indicating that, at moderate beating, fibres were been significantly shortened, since the tear index mainly depends on the fibre characteristics, especially for highly beaten (low freeness values) pulps.



Figure 8: Tensile index vs. sheet density



Figure 9: Tensile index vs. tear index

However, the pulps with a freeness above 300 mL used in the present study did not seem to be seriously damaged by cutting and micro-compression or by the frictional forces manifested among adjacent fibres.

## Tensile-light scattering relation

The unbleached kraft pulps studied had a high light-absorption coefficient (12-20  $m^2/kg$ ), high opacity (>99%) and low ISO brightness - of 20-30%. Usually, the lightscattering coefficient is significantly affected by beating and is of great importance for printing grade paper. As seen earlier (Fig. 3), the increase in sheet density with beating (low freeness) reduces the volume of the voids within the sheet structure and, as a result, the light-scattering power should decrease concomitantly with the increase in tensile strength. This reverse relationship between tensile strength and light-scattering coefficient (Fig. 10) is relatively moderate  $(\mathbf{R}^2 = 0.526)$ , due to the wide spreading of light-scattering data.

# **CONCLUSIONS**

PFI beating of Acacia manguim kraft pulps over a 40-50% yield range indicates the existence of a threshold in Kappa number, which delineates two sets of pulp properties. The properties of pulps with a Kappa number of <25 (approximate value) are comparable, despite the difference in Kappa number amongst the pulps. On the other hand, at higher Kappa number, the properties of pulps are dependent on the Kappa number, for a given degree of PFI beating. Pulps having yields up to 47% need moderate beating to give good paper properties.

ACKNOWLEDGEMENT: The financial support of Sains Malaysia University, by the IRPA EA grant 305/PTEKIND/612602 and Research University Grant No.



Figure 10: Tensile index vs. light scattering coefficient

1001/PTEKIND/8140151, gratefully is acknowledged.

# REFERENCES

<sup>1</sup> W. D. Wan Rosli, I. Mazlan and K. N. Law, Cellulose Chem. Technol., 43, 9 (2009).

<sup>2</sup> E. Forss, K. van Gadow and J. Saborowski, J. Trop. For. Sci., 8, 449 (1996).

<sup>3</sup> J. F. Francis, in "Tropical Tree Seed Manual", USDA Forest Service, 2002, p. 256.

Anon. 2008: http//www.mangiumindustries.com.my/Acaciamangium.

K. N. Law and J. L. Valade, Procs. International Conference on Acacia Species -Wood Properties and Utilization, Penang, Malaysia, March 16-18, 1998, pp. 20-31.

J. P. Bouillet, J. P. Laclau, J. L. M. Goncalves, M. Z. Moreira, P. C. O. Trivelin, C. Jourdan, E. V. Silva, M. C. Piccolo, S. M. Tsai and A. Galiana, Forest Ecol. Manag., 255, 3918 (2008).

A. F. Logan and V. Balodis, Malay. Forester, 45, 217 (1982).

<sup>8</sup> T. B. Peh, K. C. Khoo and T. W. Lee, *Malay*. Forester, 45, 404 (1982).

N. Yamada, K. C. Khoo and N. M. Yusoff, J. Trop. For. Sci., 4, 206 (1992).

<sup>10</sup> G. X. Xue, J. W. Zheng, Y. Matsumoto and G. Meshitsuka, Japan Tappi J., 58, 94 (2001).

<sup>11</sup> T. Miyanishi and K. Watanabe, Procs. TAPPI Fall Technical Conference Trade Fair, Atlanta, USA, 2004, pp. 110-121.

<sup>12</sup> W. Keigo and M. Takanori, Japan Tappi J., 58, 1097 (2004).

<sup>13</sup> C. P. Nato, A. J. D. Silvestre, D. V. Evtuguin, C. S. R. Freire, P. C. R. Pinto and A. S. Aantiago, Nordic Pulp Pap. Res. J., 19, 513 (2004).

R. O. Malinen, S. Pisuttipiched, H. Kolehmainen and F. N. Kusuma, Appita J., 59, 190 (2006).

<sup>15</sup> H. W. Emerton, in "Fundamentals of the beating process", (monograph), The British Paper and Board Industry Research Association, 1957, p. 79. <sup>16</sup> E. Claudio-da-Silva, *Tappi J.*, **65**, 99 (1982).

<sup>17</sup> M. D. Fahey, *Tappi J.*, **53**, 2050 (1970).

<sup>18</sup> R. P. Kibblewhite, Papper och Trä, 8, 519 (1975).

<sup>19</sup> J.-E. Levlin, Procs. International Symposium on Fundamental Concept of Refining, Institute of Paper Chemistry, USA, 1980, p. 131.

<sup>20</sup> H. W. Giertz, Procs. International Symposium on Fundamental Concept of Refining, Institute of Paper Chemistry, USA, 1980, p. 87.

O. Lidbrandt and U.-B. Mohlin, Procs. International Symposium on Fundamental Concept of Refining, Institute of Paper Chemistry, USA, 1980, p. 61.

<sup>22</sup> H. Corte and S. Agg, Procs. International Symposium on Fundamental Concept of Refining, Institute of Paper Chemistry, USA, 1980, p. 149.

<sup>23</sup> B. M. Allender and J. F. Waterhouse, Procs. International Conference on Advances in Refining Technologies, Birmingham, UK, 1986, Vol. II, Session III, Paper No. 8, 21 pp.

<sup>24</sup> T. Doo and R. J. Kerekes, J. Pulp Pap. Sci., 15, J36 (1989).

<sup>25</sup> R. S. Seth, *Tappi J.*, **79**, 170 (1996).

- <sup>26</sup> T. Kang and H. Paulapuro, Pulp Pap.-Can.,
- **107**, 51 (2006). <sup>27</sup> W. J. Batchelor, K.-A. Kure and D. Ouellet, Nordic Pulp Pap. Res. J., **14**, 285 (1999). <sup>28</sup> R. S. Seth, Prep. 84<sup>th</sup> Ann. Mtg. Tech. Sect.,
- Paptac, A143, 1998.
- <sup>29</sup> Å. W. McKenzie, *Appita J.*, **33**, 437 (1980).
- <sup>30</sup> A. J. Watson and F. H. Phillips, Appita J., 18, 84 (1964).

<sup>31</sup> A. J. Watson and F. H. Phillips, Appita J., 20, 47 (1966).

<sup>32</sup> F. H. Phillips, R. B. Bain and A. J. Watson, Appita J., 23, 342 (1970).

K. N. Law, J. L. Valade and E. Gonzales, Appita J., 52, 269 (1999).