EFFECTS OF KRAFT PULPING VARIABLES ON PULP AND PAPER PROPERTIES OF *ACACIA MANGIUM* KRAFT PULP

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The influence of the pulping variables (active alkali charge, sulfidity, temperature and pulping time) on the pulp yield, Kappa number and strength properties of *Acacia mangium* kraft pulp was examined. The dissolution of wood components was seen as particularly sensitive to the variations produced in active alkali charge and cooking temperature. To optimize the process, one may use either high doses of chemicals (active alkali and sulfidity), as well as low temperature and short cooking time, or *vice versa*. When beaten to a freeness of 500 mL and a 50% yield, the Kraft pulp from *A. mangium* evidenced excellent physical properties.

Keywords: kraft pulping, *Acacia mangium*, Kappa number, pulp yield, physical properties, Central Composite Design (CCD), Pareto chart

INTRODUCTION

Acacia mangium Willd., a hardwood species growing in Australia and Southeast Asia,[1,2] is one of the major fast growing species used in forest plantation schemes, in both monsoon Asia and the Pacific. Due to its fast growth and good adaptability on degraded soils, it plays an important role in sustainable forestry practices.[3-5] Besides, as due to its nitrogen fixation ability,[5-7] it is widely used for land rehabilitation.[8] In fact, it is a multiple-purpose tree, its good physical characteristics making it suitable for various wood products, including pulp and paper.[4,9-11]

Pulping of *Acacia* species, particularly plantation-grown species, has gained significant attention in recent years. Research works on the chemical pulping of *Acacia* evidence promising potential uses of this species for producing high quality paper products.[12-19] This species gives high-yield chemical fibres, suitable for obtaining fine paper with a high light-scattering coefficient.[10,19] Meanwhile, other research works report that the *Acacia* species also have a great potential in high-yield pulping.[20-22] High-yield pulps from the fast-growing *A. mangium* can be an important source of papermaking raw material for products requiring high bulk and moderate optical and mechanical properties.

Albeit the knowledge gained on the papermaking properties of *A. mangium* reported by other researchers, a survey of the published literature provides only scarce information on the influence of the pulping variables on the pulp and paper characteristics. Such information is of special importance for optimizing the pulping conditions for yielding the best possible pulp quality with minimum chemical usage, which is actually the objective of the present investigation.

EXPERIMENTAL

Raw materials

Two 14-year old *Acacia mangium* trees were harvested from Byram Forest Reserves, Penang, Malaysia, one with the diameter of 22.5 and the other one – of 24.0 cm, at breast height. The logs were debarked and sawn up to 2” x 2” x 6’ (5.1 cm x 5.1 cm x 182.9 cm) timber sizes, then chipped and screened to remove the oversized particles; the average accepted chip size was of around 23, 22 and 6 mm in length, width and thickness, respectively.

Pulping and pulp characterization

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. Experiments were conducted in accordance with a design matrix based on the Central Composite Design (CCD) technique. The basic CCD for k variables consists of a 2k factorial design with each factor at two levels (-1, +1) superimposed on a star design or 2k axial points and several repetitions at the design centre points. Four pulping variables, which are most likely to affect the pulp and paper properties produced from kraft pulping, were identified and investigated using the CCD. These variables were: (1) % active alkali, A (expressed as Na_2O), (2) % sulfidity, S (expressed as Na_2O), (3) pulping temperature, T, (^oC), and (4) pulping time, t (min). The experimental design matrix with both the coded and real variables is shown in Table 1, where the former is calculated by Eqs. (1)-(4):

$$A_{code} = (AA - 18)/2.5 \quad (1)$$

$$S_{code} = (S - 25)/5 \quad (2)$$

$$T_{code} = (T - 170)/5 \quad (3)$$

$$t_{code} = (t - 120)/15 \quad (4)$$

Table 1
Experimental design for kraft pulping of *A. mangium*

Exp. No.	Code	Pulping Factors			
		Active alkali, (% Na_2O)	Sulfidity, (% Na_2O)	Temp., (°C)	Time, (min)
1	-1,-1,-1,-1	15.5	20	165	105
2	1,-1,-1,-1	20.5	20	165	105
3	-1, 1,-1,-1	15.5	30	165	105
4	1, 1,-1,-1	20.5	30	165	105
5	-1,-1, 1,-1	15.5	20	175	105
6	1,-1, 1,-1	20.5	20	175	105
7	-1, 1, 1,-1	15.5	30	175	105
8	1, 1, 1,-1	20.5	30	175	105
9	-1,-1,-1, 1	15.5	20	165	135
10	1,-1,-1, 1	20.5	20	165	135
11	-1, 1,-1, 1	15.5	30	165	135
12	1, 1,-1, 1	20.5	30	165	135
13	-1,-1, 1, 1	15.5	20	175	135
14	1,-1, 1, 1	20.5	20	175	135
15	-1, 1, 1, 1	15.5	30	175	135
16	1, 1, 1, 1	20.5	30	175	135
17	-2, 0, 0, 0	13.0	25	170	120
18	2, 0, 0, 0	23.0	25	170	120
19	0,-2, 0, 0	18.0	15	170	120
20	0, 2, 0, 0	18.0	35	170	120
21	0, 0,-2, 0	18.0	25	160	120
22	0, 0, 2, 0	18.0	25	180	120
23	0 ,0, 0,-2	18.0	25	170	90
24	0 ,0, 0, 2	18.0	25	170	150
25	0, 0, 0, 0	18.0	25	170	120
26	0, 0, 0, 0	18.0	25	170	120

The response values obtained allow the calculation of the mathematical estimation models for each response, by a SATGRAPHICS PLUS software.

For each cooking, 200 g (oven-dried mass) wood chips were used. The ratios of liquor to material and time to maximum temperature were maintained at a constant value of 6:1 and 90 min, respectively throughout the whole experiment. After cooking, the treated chips were mechanically disintegrated in a three-bladed mixer for 1 min at 2% consistency, and screened on a flat-plate screen with 0.15 mm slits (a 6-cut slot screen). Total pulp yield was determined on an oven-dry mass basis. The screened pulps were characterized without being further refined. Kappa number of the screened pulps was determined by the Tappi method T 236. Handsheets of 60 g/m^2 were prepared and conditioned at 23 °C and 50% RH for at least 24 h before testing, according to the appropriate Tappi standard methods.

RESULTS AND DISCUSSION

The experimental results of *Acacia mangium* kraft pulping are presented in

Table 2. For the sake of brevity, the screened yield and burst index were left aside.

The impact of each independent experimental variable on the Kappa number, total pulp yield, tensile index and tear index were analyzed with a STATGRAPHICS *PLUS* software. The relationships between the treatment factors (A: active alkali; B: sulfidity; C: temperature; D: time) and the dependent responses of the properties (total yield, kappa number, tensile and tear indices) were expressed by three types of graphic illustrations, namely a standardized Pareto chart, a main effects plot and an interaction plot.

The standardized Pareto chart (*e.g.*, Fig. 1) shows each of the estimated effects, in a decreasing order of magnitude. The length of each horizontal bar is proportional to the standardized effect, which is the estimated effect divided by its standard error, being equivalent to computing a t-statistic for each effect.

Table 2
Experimental results of kraft pulping of *Acacia mangium* (unbeaten pulps)

No.	Code	Kappa No.	Total Yield, %	Screened Yield, %	Tensile Index, N·m/g	Burst Index, kPa·m^2/g	Tear Index, mN·m^2/g
1	-1,-1,-1,-1	38.9	48.8	41.5	37.46	1.39	4.06
2	1,-1,-1,-1	19.7	45.2	43.2	39.12	1.50	4.33
3	-1, 1,-1,-1	36.6	49.7	39.2	35.79	1.32	4.01
4	1, 1,-1,-1	19.0	46.0	43.2	41.67	1.69	4.60
5	-1,-1, 1,-1	25.6	46.1	41.7	43.79	1.69	4.62
6	1,-1, 1,-1	13.0	42.5	40.2	35.84	1.29	4.26
7	-1, 1, 1,-1	24.8	47.0	40.9	40.79	1.58	4.56
8	1, 1, 1,-1	13.6	43.3	41.6	37.06	1.43	4.53
9	-1,-1,-1, 1	32.9	48.5	41.5	37.93	1.46	4.23
10	1,-1,-1, 1	15.0	44.8	42.5	44.09	1.74	4.62
11	-1, 1,-1, 1	33.6	49.3	40.2	35.60	1.23	4.19
12	1, 1,-1, 1	17.4	46.7	43.5	45.96	1.77	4.91
13	-1,-1, 1, 1	21.7	45.8	41.3	41.70	1.73	4.51
14	1,-1, 1, 1	10.5	42.1	39.0	38.24	1.49	4.28
15	-1, 1, 1, 1	23.9	46.6	41.4	38.02	1.46	4.47
16	1, 1, 1, 1	14.2	43.0	41.4	38.78	1.48	4.57
17	-2, 0, 0, 0	44.6	50.0	37.6	30.76	1.15	3.62
18	2, 0, 0, 0	15.7	42.2	39.3	33.18	1.28	3.98
19	0,-2, 0, 0	17.8	45.1	40.9	36.63	1.22	3.95
20	0, 2, 0, 0	19.3	46.7	41.0	35.51	1.14	4.19
21	0, 0,-2, 0	30.0	48.6	44.6	44.03	1.82	4.78
22	0, 0, 2, 0	13.6	43.2	42.7	43.19	1.84	4.99
23	0 ,0, 0,-2	22.3	46.2	42.6	45.22	1.78	4.83
24	0 ,0, 0, 2	16.8	45.5	42.4	47.42	1.90	5.04
25	0, 0, 0, 0	20.1	45.9	42.7	47.06	1.76	4.59
26	0, 0, 0, 0	20.2	46.0	42.7	47.06	1.76	4.59

The vertical line on the plot establishes the statistically significant effects, while the bars extended beyond the line correspond to the statistically significant effects at a 95% confidence level. For the sake of simplicity, only the effects significant at a 95% level were illustrated.

The main effects plot illustrates the trend of the influence of each independent treatment variable on a particular property, while the interaction plot indicates the trend of influence of two combined treatment factors on a property.

Total yield

Figure 1 shows that active alkali (A) and sulfidity (B) are the most important factors affecting the total yield, while the effect of temperature (C) and time (D) are relatively moderate. Total yield may be expressed by the following equation:

Total yield, % = 104.466 – 0.71 × active alkali + 0.093 × sulfidity – 0.276 × temperature – 0.009 × time

The R-squared statistics of the ANOVA indicates that the fitted model explains 99.3% of the variability in total yield by the four variables.

As shown in Figure 2, total yield drops sharply as the active alkali (A) or the treatment temperature (C) increases; both factors accelerate rapidly the dissolution of wood components. The loss in yield is rather mild with respect to the reaction time (D). Interestingly, the total yield increases to some extent when sulfidity increases. This slight increase, of ca. 1.8%, at a sulfidity increase from 20 to 30% (Table 2), is probably due to the condensation reactions of lignin developed towards the end of kraft pulping, a process during which the alkali concentration decreases.[23] These reactions lead to the formation of alkaline stable linkages resulting in an increase of the molecular size lignin fragments, which will induce redeposition on the fibres. The amount is related to the reaction conditions, *i.e.* sulfidity, alkalinity and dissolved lignin fragments.

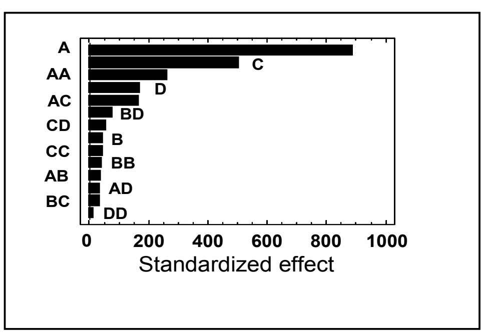

Figure 1: Standardized Pareto chart for total yield (A: active alkali; B: sulfidity; C: temperature; D: time)

Figure 3 plots the interactions of active alkali (A) with sulfidity (B), temperature (C) and time (D), sulfidity (B) with temperature (C) and pulping time (D), and temperature (C) with time (D). The minus (-) and plus (+) signs indicate, respectively, the situations in which the minimum and maximum values of the second factor are used. Any interaction of active alkali with other factors has drastically decreased the pulp yield. The interaction between temperature and time also sharply reduced the yield. However, the interaction of sulfidity with either temperature or reaction time improved the pulp yield, as due to the redeposition of the dissolved lignin on the fibres, as noted earlier.

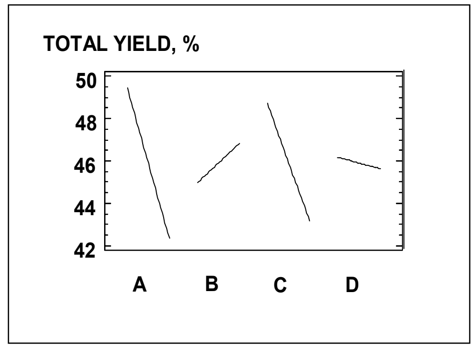

Figure 2: Main effects plot for total yield (A: active alkali; B: sulfidity; C: temperature; D: time)

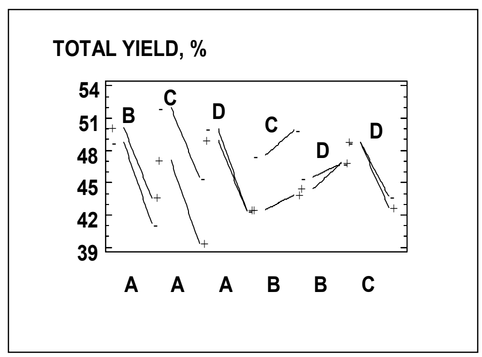

Figure 3: Interaction plot for total yield (A: active alkali; B: sulfidity; C: temperature; D: time)

Kappa number

Active alkali (A) is by far the most significant factor influencing both pulp yield (Fig. 1) and Kappa number (Fig. 4). As seen in Figure 4, all main experimental factors, as well as their interactions, have a significant influence on the Kappa number. A multiple linear regression model describing the relationship between Kappa number and the four independent variables, explaining 87.6% of the Kappa number variability is shown below:

Kappa number = 223.205 – 2.89 × active alkali + 0.073 × sulfidity – 0.821 × temperature – 0.092 × time.

Furthermore, the ANOVA evidences a statistically significant relationship between variables, at a 99% confidence level.

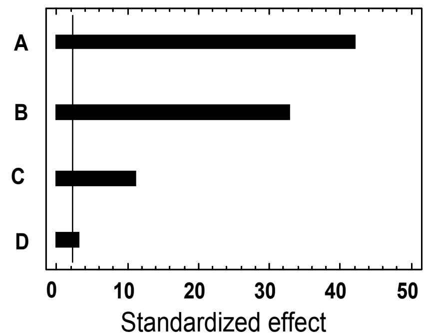

Figure 4: Standardized Pareto chart for Kappa number (A: active alkali; B: sulfidity; C: temperature; D: time)

The remarkable negative effect of active alkali (A) and temperature (C), as well as a much more moderate effect of sulfidity (B) and pulping time (D) on kappa number are illustrated in Figure 5. The influence of sulfidity (B) is rather interesting, since one would expect that, with lignin redeposition, as already discussed with regard to the total yield, the Kappa number would decrease, which was not observed.

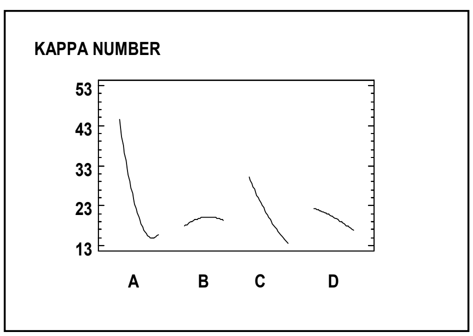

Figure 5: Main effects plot for Kappa number (A: active alkali; B: sulfidity; C: temperature; D: time)

A plausible explanation could be that the amount of lignin redeposited is rather small to have a considerable effect on the Kappa number. Alternatively, the lignin which redeposited on the fibres has different structures (fragmented forms[23]), as compared to the ones remaining in the fibre, for having a significant impact on the Kappa number. Of course, this situation may be applied only to the conditions under investigation, since it is inevitable that the Kappa number will be markedly influenced at some shorter period of cooking time or at lower temperatures. As observed in Figure 6, all interactions of the active alkali with other factors cause substantial drops in the Kappa number.

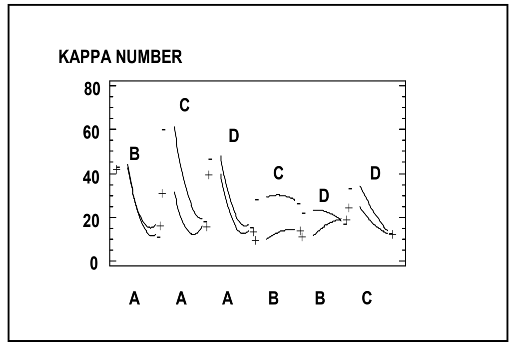

Figure 6: Interaction plot for Kappa number (A: active alkali; B: sulfidity; C: temperature; D: time)

Tensile index

The handsheet tensile index is significantly affected by the four main factors and also by their interactions (Fig. 7). The charge of active alkali (A) plays the most important role in developing tensile strength, as evidenced in the following equation:

Tensile index (N·m/g) = 39.94 + 0.241 × active alkali – 0.054 × sulfidity – 0.042 × temperature + 0.036 × time

However, this fitted model explains only 3.6% of the variability in tensile index. The ANOVA evidences no statistically significant relationship between the variables at a 90% or higher confidence level.

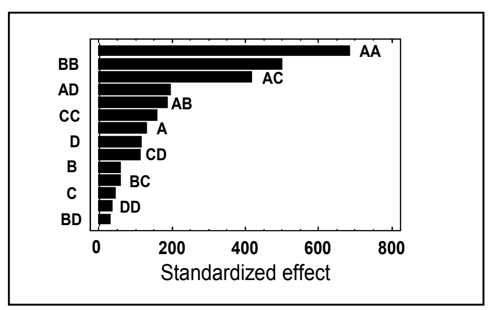

Figure 7: Standardized Pareto chart for tensile index (A: active alkali; B: sulfidity; C: temperature; D: time)

The main effects plot (Fig. 8) reveals an optimal value of both active alkali and sulfidity charge, for obtaining a peak value of the tensile index, which is also true for the cooking temperature. These optimum values seem to be located at the central point of the experimental design. As for the reaction time, albeit it has a more moderate positive influence on the tensile index, its length can gradually improve tensile strength, as due to the decreasing values of the pulp yield and Kappa number (Figs. 3 and 6, respectively). These observations are further confirmed by the interaction plot shown in Figure 9.

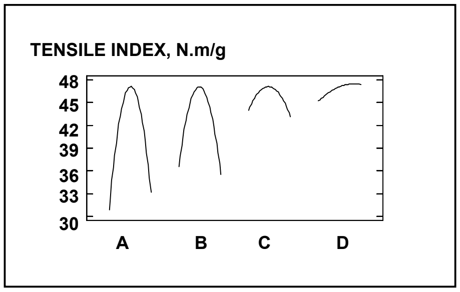

Figure 8: Main effects plot for tensile index (A: active alkali; B: sulfidity; C: temperature; D: time)

Tear index

All four experimental factors and their interactions affect considerably the tear index. However, it is the level of active alkali that has the most significant influence (Fig. 10), as compared to all the other previously mentioned properties. As suggested in the following equation, the contributions of sulfidity, temperature and time are comparatively small:

Tear index, $mN{\cdot}m^2/g$ = 1.103 + 0.036 × active alkali + 0.011 × sulfidity + 0.011 × temperature + 0.003 × time

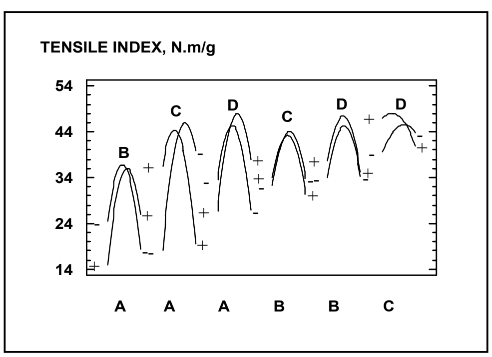

Figure 9: Interaction plot for tensile index (A: active alkali; B: sulfidity; C: temperature; D: time)

The R-squared statistics indicates that the model, although fitted, explains only 14.6% of the tear index variability. The ANOVA suggests no statistically significant relationship between the variables, at 90% or higher confidence levels.

The main effects plot (Figs. 11 and 12) indicates that the maximum value of the tear index can be attained when using the charges of active alkali and sulfidity situated at the central point of the experimental design, or at a high temperature and prolonged cooking time.

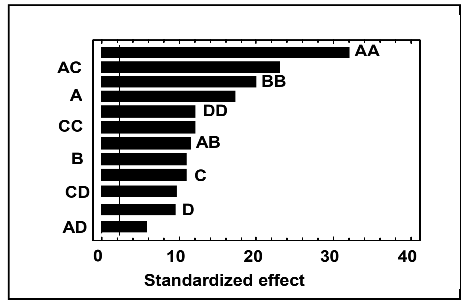

Figure 10: Standardized Pareto Chart for tear index (A: active alkali; B: sulfidity; C: temperature; D: time)

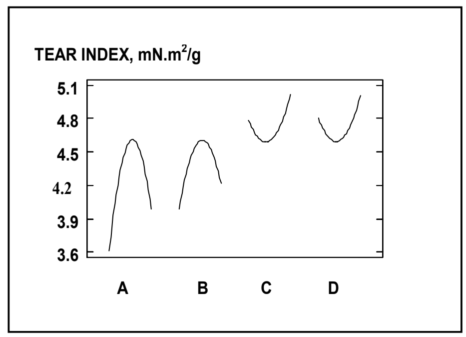

Figure 11: Main effects plot for tear index (A: active alkali; B: sulfidity; C: temperature; D: time)

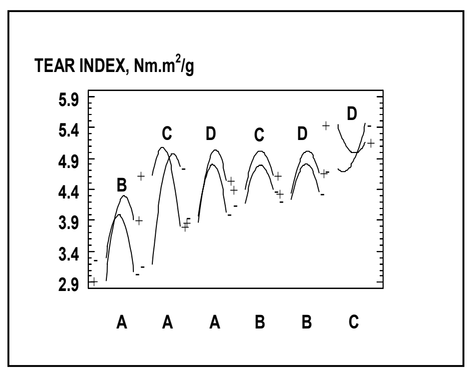

Figure 12: Interaction plot for tear index (A: active alkali; B: sulfidity; C: temperatue; D: time)

CONCLUSIONS

The pulping responses reveal that the level of active alkali and the cooking temperature have the most significant impacts on the dissolution of wood components. That is why, the total pulp yield and Kappa number are particularly sensitive to the variations of these two pulping factors.

As a result, the strength properties of the resultant pulps are also significantly influenced by these two factors.

With a targeted total pulp yield of 50%, experiments 3, 11 and 17 (Table 1) are of particular interest, as they permit to produce high quality pulp from *A. mangium*, by using either less active alkali (13%) and sulfidity (25%) at a higher temperature (170 °C) and longer cooking time (120 min), or more active alkali (15.5%) and sulfidity (30%) at a lower temperature (165 °C) and shorter pulping time (105 min). This would permit the pulp producer to establish the conditions assuring the most economic benefits. In fact, when beaten to a freeness of 500 mL, the pulps obtained under these three conditions give excellent tensile strength (81-86 N·m/g), burst index (4.5-4.9 kPa·m^2/g), tearing resistance (8.1-8.5 mN·m^2/g) and folding endurance (100-130) values.

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