Effect of the age of tree on pulping and paper properties of *Dipterocarpus crinitus* (Keruing mempelas)

(Research note)

By

Kinji SHIMADA[(1)], KHOO Kean Choon[(2)]
Mohd. Nor bin Mohd. YUSOFF[(3)]
and LEE Tack Wan[(4)]

Summary : To study the effect of the age of tree on pulping characteristics, three trees of different ages (10, 30 and 50 years old) were collected from the same location and their chemical composition, morphological properties, and pulping & paper properties were investigated.

Chemical variation with increasing tree age was indicated by a decrease in holocellulose, hot water and 1% NaOH extractives, and an increase in lignin, pentosan and alcohol-benzene extractives.

Morphological properties showed an increasing trend in fibre width, fibre wall thickness and Runkel ratio, and a decreasing trend in fibre lumen width and in the coefficient of suppleness with the age of tree. No clear trend was exhibited in the variation involving fibre length and felting power.

Although all three samples were easy to pulp using the sulphate process, their screened yields were generally low. The 50 years old tree required more chemicals than the 10 years old or 30 years old tree.

Pulp evaluation indicated that the breaking length and the burst index of the unbleached and bleached pulps generally decreased with tree age. In all cases, high Kappa number pulps had better paper properties compared to low Kappa number pulps. It was suggested that the former had less fibre damage than the latter.

The amount of pitch (yellow pitch specks) deposited on pulp sheets was slightly larger in the older trees, but very small compared to *Dipterocarpus baudii.*

The results indicated that the significant change with wood age of the principal fibre properties had a corresponding effect on the paper properties.

1. Introduction

It has been reported that the *Dipterocarpus* species can be used in chemical pulping, but do not give good paper properties because of their specific fibre morphological properties[1,2]. Furthermore, they are also resinous species[3].

A comparison study on similar species of imported tropical hardwoods in Japan and Malaysian hardwoods showed that there were clear differences in morphological properties and pulping & paper properties between the two materials[4]. The differences were probably caused by differences in the age of trees.

HILLIS reported the relationship between wood age and wood-density, fibre morphology and vessel morphology of *Eucalyptus pilularis*, and it was observed that the diameter of vessel, fibre length and fibre width increased with the age of tree[5].

Received December 24, 1987 林産化学—46 Forest Products Chemistry—46

(1) Forest Products Chemistry Division
(2) (3) (4) Forest Research Institute Malaysia

There are therefore, large differences in chemical components, fibre morphological properties and pulp & paper properties among the same species of tropical hardwoods, due to the influence of wood age. In order to obtain optimal benefits from a material for pulp and paper manufacture it is important to determine the best age for its harvesting, as has been done on *Eucalyptus camaldulensis*, a fast-growing species used in reforestation[6].

In this report, the effect of wood age on the pulping and paper characteristics of *Dipterocarpus crinitus* was investigated.

2. Experimental

2.1 Wood samples

The samples were collected from the Merlimau Forest Reserve, Jasin, Malacca within an area of 1 sq. km. The estimated ages of the trees were taken from the records of the forest station. The 10 years old tree was measured at 6 cm in diameter (breast height) and 7.5 m in height, the 30 years old tree at 24 cm in diameter and 27 m in height, and the 50 years old tree at 39 cm in diameter with no information on its height. The other wood species used for comparison purpose was *Dipterocarpus baudii*.

2.2 Sample preparation

Studies on the pulping and paper properties were performed on 2 cm thick discs taken at breast height and at height level of 50% and 80% of the tree height. Determination of the proximate chemical composition was carried out on the breast height discs which were in turn subdivided into inner and outer zones of sapwood and heartwood.

2.3 Chemical composition

All analyses were made according to Tappi standards.

2.4 Fibre morphology

Wood splints were produced from one disc from the billet at breast height. Wood fibres were prepared by digesting the wood splints through successive additions of acetic acid and 30% H_2O_2 over a water bath, and shaking up the softened wood splints with glass beads in a bottle.

2.5 Pulping procedure

Sulphate pulps were prepared in 6.5 liters stainless steel autoclaves (MK Didester, USA) heated to a temperature of 170°C by a controlled electric heater. The pulps, after washing, were screened on a fractionator using a plate with 0.15 mm slots. Pulping conditions and results are shown in Table 4.

Table 1. Bleaching conditions

Treatment	Pulp consistency (%)	Temp. (℃)	Reaction time (hr)	Chemicals*
First stage (Cl_2)	4	25—27	1	120% (Roe No.)
Second stage (NaOH)	6	60	1	2.5
Third stage (ClO_2)	6	60	2	1.0
Fourth stage (NaOH)	6	60	1	1.5
Fifth stage (ClO_2)	6	60	2	1.0

Note) * : Pulp basis, Roe No. is calculated from Kappa No. ×0.16.

2.6 Bleaching

Pulps were bleached by a five-stage (C-E-D-E-D sequence) process. The bleaching conditions used are shown in Table 1. The brightness and color reversion of the bleached pulps were determined according to ISO 2470 and Tappi UM 200, respectively. The numbers of resin spots on the paper sheets were counted under a stereo microscope (× 15).

2.7 Evaluation of pulps

Pulps (equivalent to 24 g o.d. per charge) at 3% consistency were beaten in the Lampen mill. Sheet making and testing were carried out according to appropriate Appita methods.

3. Results and discussion

The results of the chemical analysis of wood samples from trees of 3 different ages (10, 30 and 50 years old) are shown in Table 2. The 10 years old tree was distinguished from the others by having relatively high contents of ash, hot-water extractives and 1% NaOH extractives and low contents of alcohol-benzene extractives and lignin. There was a slightly decreasing trend in holocellulose and alpha-cellulose contents except in the case of the sapwood of the 30 years old tree. Lignin, pentosan and alcohol-benzene extractives, however, showed an increasing trend with wood age.

These chemical compositions of *D. crinitus* were broadly similar to those of softwoods[1,7]. The increase in alcohol-benzene solubles with wood age, and from the sapwood to the heartwood in the 30 years old and 50 years old trees, tallies with the fact that as the wood matures it tends to accumulate extractives like resins, waxes etc.[8] The young tree which was devoid of any heartwood gave the highest hot-water solubles. It is interesting to note that with increasing age, corresponding to an increased development of the heartwood, there was an accompanying drop in hot-water solubles. Although a general decrease in 1% NaOH solubles was exhibited as the tree aged, the difference in this component between the sapwood and the heartwood of the same sample was distinct.

It would have been expected that the sapwood would contain more short-chained carbohydrates, being of a more juvenile structure as demonstrated in the high alkali solubility of the 10 years old sample. In a separate determination, it had been observed

Table 2. Chemical components of the different age of tree samples of *Dipterocarpus crinitus*.

Sample	Ash %	Extractives		1% NaOH %	Holocellulose %	Alpha-cellulose %	Pentosan %	Lignin %
		Alc/ben %	Hot-water %					
Y	0.48	1.76	3.45	9.69	71.14	47.70	12.32	25.76
M−H	0.21	2.53	0.59	8.51	70.76	47.69	13.12	28.14
M−S	0.23	2.88	1.15	6.89	72.53	48.13	12.12	26.59
O−H	0.28	4.25	0.32	8.42	68.70	46.32	14.73	30.72
O−S	0.23	2.48	0.73	6.73	67.70	44.33	12.88	28.86

Note) All results are based on o.d. pre-extracted wood meal.
Y, M and O are 10, 30 and 50 year-old trees, H and S are heartwood and sapwood, respectively.

that the lignin of the sapwood had a significantly lower solubility in alkaki than that of the heartwood of the same sample.

In the case of the 10 years old samples, however, the low alkali solubility of the lignin (2.79% as opposed to 6.13% and 4.24% of the heartwood and sapwood lignin of the 50 years old tree, for example) could not be considered as a significant cause of the high alkali solubility of the wood (9.69%).

The latter, as reflected in the high hot water solubility (3.45%) is more probably an indication of the presence of a relatively large amount of low molecular weight carbohydrates. The results of the cellulose, pentosan and lignin determinations suggest that as the tree grow older the decline in the cellulose content (especially in the heartwood) was compensated by relative increases in the amounts of pentosan and lignin.

Density and morphological properties of the samples are shown in Table 3. The results indicate a high wood density ranging from 0.72 to 0.83 g/cm^3. The fibre length varied from 1.33 to 1.45 mm. There were increasing trends with wood age in fibre width (21.62–25.24 μ),fibre wall thickness (9.28–10.40 μ) and Runkel ratio (6.1–9.5), and decreasing trends in fibre lumen width (3.06–2.20 μ) and coefficient of suppleness (14.0–8.7). The thick cell-wall and the low coefficient of suppleness suggest that the fibres would be less flexible during the beating operation. A look at the density trend reveals an apparently anomalous higher density in the 10 years old tree. That the 50 years old tree should have a denser wood than the young 30 years old tree is understandable in view of the cell-wall thickening and increased lignification with maturity. However, it is not valid to explain for the highest density in the youngest sample in terms of its fibre dimensions. In fact, the sample had the most slender fibre with the thinnest cell-wall. The reason behind the high density was nevertheless revealed on a close examination of its anatomical features.

In contrast to the older samples, the 10 years old wood had a more compact structure on account of the abnormally small vessels which were found to be a third or less the size of those of either the 30 years old or 50 years old samples. Another significant factor to account for the higher density of the 10 years old wood was the relatively high contents of extraneous substances. The hot-water solubility was the largest in this sample. The ash content was about twice that of either of the older samples.

The conditions used for pulping and properties of the pulps are shown in Table 4. To obtain the pulps of Kappa number 20 and 40, several conditions in active alkali (10 to 17%) were carried out on the samples by the chemical sulphate method. At 18% active alkali, the pulps were found to be overcooked resulting in low Kappa number from 11.4 to 14.5 with screened pulp yield of 41 to 47%. Khoo and Peh[9] inferred from their study of the

Table 3. Morphological properties of the different age of samples of *D. crinitus*.

	L	D	l	W	COS	R.R	L/D	Density
Y	1.33	21.62	3.06	9.28	14.01	6.1	61.5	0.826
M	1.44	23.06	2.46	10.30	10.70	8.4	62.5	0.730[a)]/0.709[b)]
O	1.45	25.24	2.20	10.40	8.7	9.5	58.2	0.748[a)]/0.736[b)]

Note) L : fibre length (mm), D : fibre width (μ), l : fibre lumen (μ), W : fibre wall thickness (μ), COS : coefficient of suppleness (%), R.R : Runkel ratio, L/D : felting power, Density : g/cm^3.
a) heartwood, b) sapwood.
Y, M and 0 are 10, 30 and 50 year-old trees.

Table 4. Pulping conditions and properties of sulphate pulps of *Dipterocarpus crinitus*.

Sample		A.A (%)	Kappa No.	Pulp Yield (%)			Brightness		Bleach yield (%)
				Screened	Screening	Total	Unbleached	Bleached	
Dipterocarpus crinitus	Y−20	14	20.5	43.6	4.5	48.1	24.8	89.8	95.5
	Y−40	10	42.0	27.2	30.8	58.0	17.1	86.6	93.2
	M−20	14	21.7	44.1	5.6	49.7	21.3	86.7	97.0
	M−40	11	40.5	31.2	25.8	57.0	14.8	86.5	92.9
	O−20	17	18.1	43.6	1.0	44.6	20.0	86.3	95.4
	O−40	12	41.7	36.4	16.6	53.0	14.0	87.3	92.7
Dipterocarpus baudii		14	23.2	39.9	4.4	44.3	23.1	86.9	94.8

Note) Sulphidity ; 25% (based on active alkali), Liquor to wood ratio ; 3.5 : 1.0, Schedule ; 1.5hr to 170℃, and 1.5hr at 170℃, Y, M and O are 10, 30 and 50 year-old trees. 20 and 40 are Kappa number of pulps.

chemical composition of some Malaysian hardwood in relation to their pulping properties that an indication of the expected chemical pulp yield could be obtained from a consideration of both the holocellulose and aplha-cellulose contents. On this basis, the trend in the total pulp yield of the *D. crinitus* with wood age could be explained. Differences in the yields of both the 10 years old and 30 years old samples were marginal due to the close values of their holocellulose and alpha-cellulose contents. That the 50 years old sample produced the lowest pulp yield was a reflection of the lowest contents of these components in the sample. The results indicate that the wood samples required low amounts of chemicals for pulping, although the wood had high lignin content, density and thick cell-wall. Consequently,further pulping trials were carried out at lower chemical concentrations to achieve the desired Kappa numbers. By using 14% active alkali, the 10 and the 30 years old samples gave a pulp yield of 44% with a Kappa number of around 20. Pulping of the 10, 30 and 50 years old samples at 10, 11 and 12% active alkali, respectively, resulted in Kappa number around 40.

These samples gave pulp yields of less than 40% but high shiver contents. The amounts of active alkali to obtain the required pulps seemed to be correlated with the chemical composition of the samples. The active alkali required increased with increasing extractives and lignin contents.

The results indicated that the younger tree was more pulpable compared to the older ones.

The bleaching conditions and the properties of the bleached pulps are given in Tables 1 and 5. All pulps produced clear and white pulps, which ranged from 86.3 to 89.8% in brightness (ISO basis). Bleached yields ranged from 92.7 to 97%.

Resins in bleached pulps are considered to be the cause of pitch problems and to have an effect on color reversion[10].

Table 5 shows the post color number of the samples. In the case of the pulps of Kappa number 20, post color number increased with tree age from 1.84 to 2.53. The results show that the *D. crinitus* had rather higher post color numbers than the average Japanese hardwoods[11]. A large number of resin spots were observed on the pulp sheets from *Shorea resinosa* and *Dipterocarpus baudii*. On the other hand, the appearance of resin spots on the sheets from *D. crinitus* was much less, but showed an increasing trend with the age of tree (Table 6).

Evaluation data for the unbleached and bleached pulps are shown in Tables 7 and 8

Table 5. Brightness and color reversion tests of *Dipterocarpus crinitus*

Pulp no.	ISO Brightness of pulp			P.C. No.
	Before aging	4 hrs in oven (105℃)	18 hrs in oven (105℃)	
Y—20	89.8	84.1	80.3	1.84
Y—40	86.6	80.1	75.9	2.79
M—20	86.7	81.6	77.6	2.21
M—40	86.5	79.3	75.5	2.92
O—20	86.3	79.5	76.5	2.53
O—40	87.3	79.7	75.9	2.90
Diptero. baudii	86.9	81.8	78.4	1.99

Note) P.C.No. was determined after 18 hrs in 105℃ oven.
$P.C.No. = [(1-R)^2/2R - (1-R_0)^2/2R_0] \times 100$, where R and R_0 show the reflection ratio of pulp sheets after and before aging, respectively.
Y, M and O are 10, 30 and 50 year-old trees.
20 and 40 are Kappa number of pulps.

Table 6. Numbers of resin spots (yellow pitch specks) on the bleached sulphate pulps (*D. crinitus* and *D. baudii*)

Sample		Size of pitch I	II	III
Dipterocarpus crinitus	Y—20	0	0	6
	Y—40	0	0	3
	M—20	0	0	9
	M—40	0	0	22
	O—20	0	2	88
	O—40	0	2	54
Dipterocarpus baudii		6	82	852

Note) Resin spot area ; I = 0.5 mm², II = 0.5—0.1 mm², III = 0.1 mm², Pulp sheet area is 200 cm².
Y, M and O are 10, 30 and 50 year-old trees, respectively. 20 and 40 are Kappa number of pulps.

Table 7. Evaluation of unbleached sulphate pulps of *Dipterocarpus*

Species		Thickness (microns)	Density (g/ml)	Breaking length (km)	Tear Index (mNm²/g)	Burst Index (kPam²/g)	Folding endurance	Air resist. (sec.)
Dipterocarpus crinitus	Y—20	98.2	0.61	6.32	10.9	3.3	63	18.1
	Y—40	96.4	0.60	6.80	11.6	3.5	88	20.6
	M—20	104.1	0.58	5.85	11.4	3.3	66	12.1
	M—40	103.2	0.58	6.25	11.0	3.5	101	17.1
	O—20	103.4	0.58	5.49	10.4	2.8	50	6.0
	O—40	107.4	0.55	6.07	10.6	3.3	77	6.1
Dipterocarpus baudii		95.6	0.63	7.74	11.5	4.5	286	25.3

Note) Basis weight ; 60 g/m², Y, M and O are 10, 30 and 50 year-old trees, respectively.
20 and 40 are the Kappa number of pulps.

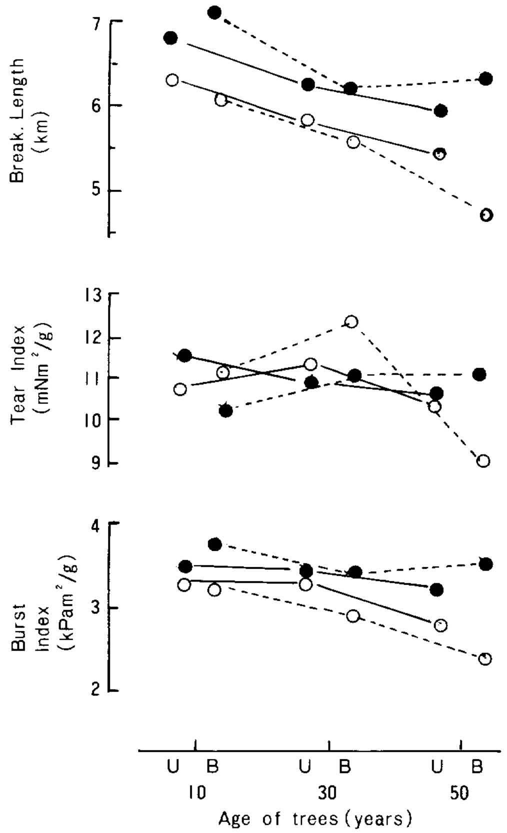

Fig. 1. Relationship between physical properties of pulps (—UPK, --- BKP, ○ Kappa no. 20, ● Kappa no. 40) and wood-age.

crinitus

Stretch (%)	TEA Index (mJ/g)	Freeness (ml)
3.9	1,248	235
3.8	1,303	245
3.8	1,140	250
4.1	1,317	265
3.5	992	250
3.8	1,181	240
4.1	1,636	250

and Fig. 1. The results indicate that breaking length, burst index, folding endurance and TEA index of the unbleached and the bleached pulps generally decreased with tree age. These pulps also gave lower hand sheet density and higher thickness than those of Japanese hardwood[11]. It has been established that the thickness of the fibre wall has an important bearing on most paper properties, and that the thick-walled fibres give bulky, open sheet with rather rough surface and poorly-formed sheet[12].

The main reason is that these thick-walled fibres do not collapse readily when formed into sheets giving rise to small contact area and poor interfibre bonding. The results show that the paper strength properties of *D.*

Table 8. Evaluation of bleached sulphate pulps of *Dipterocarpus*

Species		Thickness (microns)	Density (g/ml)	Breaking length (km)	Tear Index (mNm²/g)	Burst Index (kPam²/g)	Folding endurance	Air resist. (sec.)
Dipterocarpus crinitus	Y—20	97.3	0.61	6.10	11.2	3.27	61	21.9
	Y—40	96.9	0.61	7.06	10.3	3.83	106	17.6
	M—20	98.4	0.60	5.57	12.5	2.94	63	9.3
	M—40	103.2	0.57	6.25	11.0	3.44	89	9.4
	O—20	98.8	0.60	4.67	9.0	2.44	30	10.5
	O—40	101.1	0.59	6.40	11.3	3.64	84	28.1
Dipterocarpus baudii		94.6	0.63	7.36	12.6	4.12	198	32.6

Note) Basis weight ; 60 g/m², Y, M and O are 10, 30 and 50 year-old trees, respectively.
20 and 40 are the Kappa number of pulps.

crinitus were adversely affected by an increase in fibre wall thickness.

Evidence of similar behavior of thick-walled fibres has also been observed by KOEPPEN and COHEN[13] in their comparison of the pulp properties of the heavy and lighter density mangrove species. As in the case of *D. crinitus*, the heavy density species, which had almost similar densities and fibre length/fibre diameter ratios, generally displayed good tearing resistance but much poorer values in other strength properties[14]. PEH[15] found among the 116 tropical hardwoods studies that those with a basic density of over 650 kg/cm³ were normally associated with thick-walled fibres giving pulps of poor strength except tear. It was also reported that the *Dipterocarpus* group (Keruing spp.), although readily pulped to give satisfactory yields, tended to produce resinous pulps and papers which were bulky and weak[1].

In all cases, high Kappa number pulps had better paper properties compared to those with a low Kappa number. It was suggested that the former had undergone less fibre damage than the latter. The same observations had been noted among the heavy density Malaysian dipterocarps where higher Kappa number pulps were found to develop higher strength properties on beating than pulps of lower Kappa number[15].

In conclusion, it should be emphasized that there are significant relationships between pulping and paper-making characteristics and the age of tree. In addition, it is necessary to develop a method for removing or reducing the presence of pitch in pulps prepared from such resinous woods since pitch problem in papermaking, especially for fine papers, is undesirable.

4. Acknowledgements

The authors would like to express their gratitude to Mr. YONG Fook Onn and Ms. YEO Moi Wah for their valuable support in wood analysis and paper testing, and to the officers of the Merlimau Forest Reserve for the collection and identification of the *Dipterocarpus crinitus* samples.

crinitus

Stretch (%)	TEA Index (mJ/g)	Freeness (ml)
3.9	1,237	220
3.9	1,382	250
4.0	1,179	225
3.9	1,258	185
3.7	927	245
3.9	1,273	200
4.2	1,616	213

References

1) KAYAMA, T. *et al.* : Bull. For. & For. Prod. Res. Inst, 221, 175 (1969) and 207, 166 (1967)
2) PEH, T.B. *et al.* : Pulp & Paper Industry and Research in Peninsular Malaysia, in press, (1985)
3) For example, Low, C.K. *et al.* : Malay. Forester, **45** (2), (1982)
4) SHIMADA, K. : Unpublished
5) HILLIS, W.E. : J. Tappi, **29** (7), 341, (1975)
6) TAKANO, I. : Bull. For. & For. Prod. Res. Inst, 347, 170, (1987)
7) USAMI, K. : Times of Paper Technology, **6**, 1, (1987)
8) HALE, J.D. : Structure and physical properties of pulp-wood in the pulping, Vol. 1, McGraw Hill, USA, (1969)
9) KHOO, K.C. and PEH, T.B. : Malay. Forester, **45**, 244, (1982)
10) RUDRA, P. Singh : The Bleaching of Pulp, Tappi press, (3rd edition), 357, (1979)
11) KAYAMA, T. : J. Tappi, **22** (11), 581, (1968)
12) HILLIS, W.E. : Wood Extractives, Acad. Press, NY, (1962)
13) KOEPPEN, K. *et al.* : Holzforschung, **16** (1), 18, (1956)
14) KENNETH, W.B. : Handbook of Pulp and Paper Technology, 225, Van Nostrand Reinhold Company, (1970)
15) PEH, T.B. : Malay. Forester, **41**, 253, (1978)

（研　究　資　料）

クルイン（*Dipterocarpus crinitus*）材のパルプ化および製紙特性に及ぼす樹齢の影響

島　田　謹　爾[1]，KHOO Kean Choon[2]
Mohd. Nor bin Mohd. YUSOFF[3]
and LEE Tack Wan[4]

要　旨

熱帯材の樹齢がパルプ化および製紙特性に及ぼす影響を明らかにする目的で，同じ環境下で成長した，10，30および50年生のクルイン材（*Dipterocarpus crinitus*）を供試料とし，化学成分，繊維形態，パルプの性質等を比較検討し，以下の結果を得た。

1. 化学成分組成は樹齢の増加に伴って，ホロセルロース，熱水抽出物，1% NaOH 抽出物が減少し，リグニン，ペントザン，アルベン抽出物が増加した。
2. 繊維形態は樹齢とともに繊維幅（D），膜厚（W），2 w/1 (Runkel ratio) が増加し，ルーメン幅（1），1/w × 100 (Coefficient Suppleness) が減少した。
3. いずれもクラフト法で十分にパルプ化されるが，蒸解薬液消費量は樹齢とともに増加した。
4. パルプの強度的性質では，裂断長，破裂および耐折強さは樹齢とともに低下した。
5. パルプシートの色戻り（PC No.）および樹脂斑点の量は樹齢とともにわずかに増加するが，後者は比較に用いたクルイン材（*D. baudii*）より非常に少なく，樹脂障害の影響は少なかった。
6. 樹齢により，成分組成，繊維形態が大きく異なるため，結果として製紙特性に及ぼす影響が大きいことが認められた。

---

(1)　林産化学部
(2) (3) (4)　Forest Research Institute Malaysia