Influence of Chip Size Vis-a-Vis Permeability on Pulping Behaviour of Eucalyptus Hybrid (Tereticornis). Part I Effects on Screened Pulp Yield, Kappa Number and Effective Alkali Consumption

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ABSTRACT

Pulp yield showed an increasing trend with increasing permeability when standard sized chips were used. Composite permeability to pulp liquor decreased significantly with increase in chip thickness. Kappa number increased as the chip thickness increased showing a low degree of delignification with decrease in permeability. Lignin free yield was found to be related to composite permeability. At higher premeability, a lower chip thickness resulted in lower yield indicating an over reaction. At 3mm chip thickness yield-permeability curve fwas almost horizontal. A higher permeability thus favoured a higher chip thickness for higher yield.

Pulping process involves the separation of cellulosic fibres by chemical/mechanical action on lignin, action as a bonding agent between the fibres. A major part of the lignin is concentrated in the middle lamella and primary wall and for liberation of fibres in chemical or semichemical pulping the pulping liquors have to penetrate to the middle lamella. Subsequently, the lignin degradation products have to move out to make way for the reagent for further reacton. According to Akhteruzzaman et al (1980) the pulping reaction proceeds in five steps.

- i) transportation of the pulping chemical into chip by bulk flow and diffusion.
- ii) adsorption of the chemical.
- iii) chemical reaction.
- iv) desorption of reaction products.
- v) transportation of reaction products from the reaction site.

The first step is very important and is dependent on the permeability behaviour as well as the available ' voids in the wood. This may be the most crucial stage

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especially in hardwoods, where even the initial penetration/bulk flow may be impeded or retarded if the vessels are plugged with tyloses or other gummy deposits, as the vessels constitute the major flow paths in such types of woods. Such difficulties were encountered in pulping of white oak in which no penetration was possible due to presence of tyloses (Stone 1956).

Even if the vessels are fully penetrated (which will occur quite early prior to actual pulping) it is important that the liquor 1 te transported to other neighbouring cells for better and uniform pulping. This may again occur either by bulk flow through the intercell pits or by diffusion if such chnnels are blocked or non-existent. Because of the heterogeneity in the hardwood structure, in terms of number size as well as types of cells involved, the bulk flow may not be always complete to the extent desired and the process may become diffusion dependent, i e. involving passage of ions through the cell walls under a concentration gradient. In such cases if the temperature of the pulping liquor reaches 130°C before the liquor reaches the middle lamella of every

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cell, the unpenetrated portions are likely to be charred in the presence of heat and pressure and result in loss in yield (Stone and Green 1958,1959). Although the actual pulping may not start during the heating stage, it has been observed that alkalies do react with the wood substance and their flow behaviour is changed due to change in viscosity (Kumar *et al* 1986). Part of this alkali is rapidly consumed in the initial stages of the heating period, due to reaction with carbohydrates and extractives (Hartler 1962;MacDonald & Franklin 1969).

Although it is believed that alkaline solutions diffuse in wood almost equally in transverse and longitudinal directions (Ekman & Fogelberg 1966 Hartler, 1962; Stone and Green 1958; 1959), the diffusion mechanism of alkaline solutions through wood of different densities and having different cell types with different cell wall thicknesses, is not exactly known. If the diffusion stage is not complete before the delignification temperatures are reached, yield losses are likely to occur due to uneven cooking within the chip. Yield losses have been observed in denser woods while pulping to same kappa numbers (Kumar, *et al* 1986).

The last step of transportation of the reaction products from the reaction site is also very important as lignin degraded products undergo condensation reaction and get grafted on to the crystalline cellulose (Kleinert 1966).

The transportation of the pulping liquor into the wood structure and of the lignin reaction products out of the wood chip depends on the permeability behaviour of the wood species. Although the role of wood permeability in pulping was recognised during early fifties, no direct study to correlate pulping characteristics or define pulping parameters vis-a-vis permeability had been attempted. The key factor for ideal pulping being uniform penetration of the pulping liquor, the problem becomes more important when mixed hardwoods of different permeabilities and densities are cooked together. An alternate approach has been to investigate the chip dimensions suitable for pulping in different wood species. Lower chip dimensions give a higher surface to volume ratio and provide better prospects for uniform penetration.

Hatton (1972) introduced the concept of "liquor accessibility factor" which links chip permeability to chip thickness. A chip thickness of 3mm has been recommended as optimal although for some impermeable species like western hemlock, trembling aspen, beech white birch, a chip thickness of 2mm is more desirable (Hatton 1978, 1978 b; Hatton and Keays 1972).

Apart from the problem of inadequate penetration, chip dimensions are critical for energy consumption at the chipper, optimum loading of the digestor, production of fines and plugging of hoppers, feeds bins and pneumatic conveying systems, etc. As chip length and width have not been found to be of much consequence in penetration of pulp liquor, a chip length of 30mm has been recommended as optimum from all points of view (Raggam 1976).

Recently Kumar *et al* (1986) evaluated a number of tropical hardwoods of Indian origin and concluded that low permeability of wood resulted in lower yield, while high permeability always favoured a good pulping with higher yield. Although no direct correlation of axial gas permeability with pulping behaviour could be established, permeability to pulp liquor was found to be strongly cor elated to pulp yield. In addition to permeability of vessels, the lateral movement of fluids to the neighbouring cells played a significant role in better pulping.

Eucalyptus hybrid, a fast growing wood species, has been grown in large scale in the country to augment supplies of fibrous raw material for the paper industry. Studies on preservation of eucalyptus revealed that it is an impermeable species and very refractory to treat. This investigation was undertaken to study the permeability (of *Eucalyptus* hybrid) chip thickness-chip permeability relationships and their overall influence on pulping behaviour of this wood species. Details of permeability studies and their variation were reported in an earlier publication (Bahri & Kumar 1982).

MATERIALS AND METHODS

Materials :

Studies were confined to a single stem obtained from New Forest plantation. While the material from the two ends was used for permeability studies, the middle portion was used for pulping studies.

The remaining log, after obtaining discs for permeability samples, was cross cut into billets of about 50cm. length. Some of these billets were sawn into 3mm and 4mm thick planks. 1mm and 2mm thick sheets were obtained by slicing the billets on a veneer slicing machine. These planks/sheets were then sheared into 1.25cm. wide strips, after demarcation of sapwood, outer heartwood, middle heartwood and inner heartwood portions. The strips belonging to different zones were sorted and were then cut into 3cm long parallelopipedon shaped chips by hand on an inclined platform. The chips from different locations and different thicknesses were stored in separate lots Chips were further stored over a saturated solution of sodium dichromate for one week, so as to regulate the moisture content of the chips to approximately 10%, prior to testing/pulping. Conditioned chips were used for pulping and finding the composite permeability.

Pulping: 200g (oven dry weight basis) of chips were charged idto steel bombs alongwith cooking liquor and were pulped in batches of six, in a series digestor, by the kraft process under the following cooking conditions.

Table-1

Pulping conditions used for pulping *Eucal*, ptus hybrid chips.

1.	Cooking chemicals (as Na ₂ O)	18%
2.	Cooking temperature	162°C
3.	Time to reach maximum temperature	1hr. 30min.
4.	Cooking period	2hr. 30min.
5.	Wood liquor ratio	1:4
6.	Sulphidity	25%

Determination of Composite Permeability of Chips:

Conditioned chips (10% mc) were used for the determination of composits permeability towards water and pulp liquor. A known weight of the chips was placed in a 500ml beaker and a weight was placed over the chips, to prevent them from floating during purging with water/pulp liquor. The chips were covered with water and the beaker was then placed in a pressure cylinder, and subjected to a pressure of 7 kg/cm² for 20 minutes. The water was then drained off and the weight of the wet chips was taken after sponging off the excess water sticking on the surface of the chips. Composite permeability was measured as the percent voids filled and was calculated as follows :

Percent Voids at f s
$$p=(1-Density of Wood) \times 100$$
 (a)
(Density of Wood Substance)

Volume of Chips =
$$O.D.$$
 Wt. of Chips
Density of Wood (b)

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Total Void Volume (fsp) =	
Total Volume (b) \times Percentage Voids (a)	(c)
100	
Total water Absorbed =	
Water Adsorbed + Free water in Wood	
in cell wall Capillaries.	
Water Adsorbed =	
O.D. Wt. of Wood \times (fsp-MC of Wood)	(d)
100	
Water Absorbed =	
(free water in voids).	

Total water Absorbed-Water Adsorbed (d) e)

% voids filled = $\frac{\text{Free water in wood (e)}}{\text{Total voids (c)}} \times 100$

Composite permeability of chips to pulp liquor was obtained in the same manner.

RESULTS AND DISCUSSION

Data on proximate chemical analysis of Eucalyptus hybrid from various locations along the tree radius are reported in Table 2. There are significant variations in the extractive content especially between the sapwood and heartwood. Variation also exists between various locations in the heartwood and the midlle heart h s been found to have the highest amount of extractives. Alcohol-benzene soluble extractives also vary almost in a similar fashion. This not only affects the permeability and pulping reaction (due to increase in alkali demand) but also affects the yield because of lower percentage of total holocellulose (on percentage basis) in extractive rich portions.

Table—2 Proximate chemical analysis of wood from various locations in the (*Eucal*) *ptus* hybrid).

Location	Total extrac- tives %	Alcohal Benzene extrac- tives	Lignin %	Holoce llulose (by diffe rence)
Sapwood	2.34	.44	31.76	65.90
Outer heartwood	5.40	1.41	31.65	42.95
Middle heartwood	12 25	3.71	30.14	51.61
Inner heartwood	9 35	2.57	29 81	60.84

Influence of permeability on Pulp yield :

As has been reported earlier, there is a wide difference in sapwood and heartwood permeability and there is variation within heartwood also although the differences are not statistically significant (Bahri and Kumar, 1982). Fig. 1 shows that the pulp yield (average of 3 mm and 4 mm thickness) obtained from different zonesin the eucalyptus stem is dependent on the permeability, although such proportionality was not observed while analysing data from different species (Kumar *et al*, 1986). A Similar increasing trend in pulp yield was observed with composite permeability to water and pulp liquor (Fig. 2). It may also be noticed that permeability to water in case of eucalyptus whereas a reverse trend was observed in other wood species (Kumar *et al*, 1986).







Effect of chip Thickness on Pulp yield :

The unbleached yield and other properties of pulp obtained at various chip thicknesses are reported in Table 3. Chips from different portions of wood behave in a different manner. While there was an increasing trend in unbleached yield with increase in chip thickness in the sapwood, in the heartwood values reached a maximum at a chip thickness of 3mm, there was a

Table—3.

Pulp yield and pulp properties from different chip thicknesses in different locations.

LOCATION	Chip thickness mm	Unbleached yield %	Kappa number	Lignin content	Alkali consumed g/1
SAPWOOD	1 2 3 4	30.05 36.00 41.35 44.96	21.68 22.04 23.19 23.38	2.39 2.24 2.68 2.98	32 26 31,98 29,30 27,68
OUTER HEARTWOOD	1 2 3 4	35 05 39.69 42 36 38.63 (1.0 SR)*	24.00 24.20 24.91 26.10	2.95 3.05 3.06 3.68	32.75 31.74 30.50 28.90
MIDDLE HEARTWOOD	1 2 3 4	37.20 41.30 42.50 39.04 (-2.5 SR)*	27.29 29.18 30.00 31.39	3.55 3.79 3.45 4.12	33.86 33.42 32.60 31.30
INNER HEARTWOOD	1 2 3 4	35.80 42 44 42.90 40 07 (-2.7 SR)*	29.50 30.90 32.00 32.68	3.37 4.02 4.30 4.70	33.54 32.97 32.03 30.90

*S.R.—Screen rejects %

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sharp fall in pulp yield as the chip thickness was increased to 4mm in the case of heartwood (Fig. 3). Hatton (1978 b) also observed a fall in screened yield with incrcreasing chip thickness from 2-5 to 8mm when the chips were cooked to same kappa number. In our experiment, since constant cooking conditions were maintained, pulps with different kappa number were obtained.



Total yield was also found to be related to the kappa number obtained (Fig. 4). It is well established that the yield decreases when cooking is done to a lower kappa number as cooking to lower kappa number involves a partial removal of the residual lignin. Since residual is more strongly bonded to the fibres (MacDonald, and Franklin 1969) and in addition there may be condensation of dissolved lignin products with the cellulose (Kleinert, 1966), during removal of this lignin, carbohydrate fractions are also likely to be lost. Although there is no evidence of any condensation reaction in the present study (black liquor had a viscosity range of 1.792 to 3.660). The thicker chips (4mm) did tend to have a higher lignin content in the unbleached pulp. This indicates that the chip thickness tends to change the transition point between bulk and residual phases, towards a higher lignin content, although no studies on this aspect are earlier reported. Akhtaruzzaman and

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Virkola (1979) hypothesised that any parameter which retards the delignification reaction (chip thickness being one of them) tends to shift this transition point towards a higher lignin content.



Hartler and Ostberg (1959) obtained almost linear relations between total yield and Roe number. Hatton (1978b) obtained curves almost similer to our curves (Fig. 4). Although pulp yield is related to kappa number the chip thickness has a dominant role to play as the slope of the curve changes with chip thickness. Hatton (1978b) observed a fall in yields for thicker chips when the pulp was cooked to a kappa number of 15.

Effect of Chip Thickness on Alkali Consumption :

Alkali consumed in various cooks from different portions and different chip thicknesses has also been listed in Table 3. It may be seen that alkali consumed falls with the increase in chip thickness (Fig. 5). This is in agreement with the observations made by earlier workers (Hatton 1978a; Hatton and Keays 1973). Akhtaruzzaman and Virkola (1979) also observed decrease in alkali consumption with increasing chip thickness while following a constant cooking schedule. The increase in alkali consumption is attributed to lower removal of lignin at higher chip thickness, since higher kappa numbers were obtained at higher chip thickness (Table 2). If cooking is done to same kappa number alkuli consumption with higher chip thickness usually rises (Hartler, 1962) Extractives are also responsible for the change in alkali consumption. Fig. 6 depicts an almost linear relationship between alkali consumed (average for all chip thicknesses) and extractive content.



The case of sapwood is typical, as only slight increase in kappa number is observed, between cooks obtained from 1mm and 4mm chip thickness. 4mm chip thickness in this case has given the highest unbleached yield (44.96%). The lower alkali consumption points towards a proper delignification process, indicating that the liquor was uniformly distributed. In 1mm thickness there appears to be an over-cooking during which considerable amounts of carbohydrate were also dissolved out. The increase in alkali consumption is probably due to neutralisation reactions with the carbohydrate decomposition products.

The rise in alkali consumption appears to be related to fall in yield when cooking chips of different thicknesses but from the same location in the tree. An opposite trend is however noticed at 4mm thickness in less permeable heartwood. The alkali consumption in this case is low and so is the yield. This is partly due to lower lignin removal (cooks have higher kappa numbers) and partly due to nonuniforn pulps as evidenced by screen rejects. The alkali in such cases is not able to penetrate all through, limiting its action only in the penetrated regions.

Chip Thickness-Permeability-Pulping Relationships :

Permeability according to Darcy's law is a property of the porous medium and is thus independent of the dimensions of the porous medium. It may be seen from the Darcy flow equation that flow rates decrease with the increarsing dimensions (flow rates are inversely proportional to the length of the material). Chip length and thickness will thus play an important role in the penetration of pulp liquor into the chip. Length is important because longitudinal permeability is several times the transverse permeability, whereas thickness is important as it is the smallest dimenion. This has been borne out by many studies on chip dimension vis-a-vis pulping behaviour (Farkos 1965).

Since penetration of p 11p liquor is a time dependent phenomenon, the chip dimensions are likely to infihence the same. Fig. 7 depicts the penetration/absorption pattern of chips of various thicknesses (constant length and width) when impregnated with water. Percent voids filled, represented by composite permeability (an index of longitudinal and lateral flow of the liquid into the chip) falls sharply when chip thickness increases



from 1mm to 4mm. Alkaline solutions may be differently absorbed as the same may be accompanied by some physical and chemical changes. Fig. 8 depicts the absorption behaviour of pulp liquor in chips of different thicknesses. The comparison of the two figures indicates that alkaline solutions penetrate better in chips. Absorption behaviour with pulp liquor with respect to chip thickness was, however, the same as in the case of water, indicating that considerable fall in permeability occurs when the chip thickness increases from 2mm to 3mm to 4mm. In sapwood, the fall in permeability from 3 to 4mm is not much.



Data on composite permeability from various locations at various chip thicknesses and pulp yield obatined are reported in Table 4. The influence of composite permeability on pulp yield can be clearly seen in Fig. 9.

LOCATION	Chip Kappa thickness number (mm)		Composite permeability (% voids filled) Water Pulp liquor		Net lignin free yield	
SAPWOOD	1 2 3 4	21.7 220 23.2 23.4	73 74 43 38	98 80 54 47	27.7 32.8 38 7 43 0	
OUTER HEARTWOOD	1 2 3 4	?4.0 24.2 24.9 26.1	67 51 30 12	81 62 41 21	32.1 36.2 39.1 33.5	
MIDDLE HEARTWOOD	1 2 3 4	27.3 29.2 30.0 31.4	59 43 19 8	68 53 37 16	33.6 37.5 39.1 32.4	
INNER HEARTWOOD	1 2 3 4	29.5 30.9 32.0 32.7	60 40 25 12	79 51 31 17	32.4 38.4 39.0 33.4	

Table—4

Pulp yield and composite permeability of wood chips of various thicknesses from different location

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It is interesting to see that the pulp yield decreases with the increasing permeability of chip at thickness of 1mm and 2mm. At 3mm the curves are almost flat with respect to permeability; and at 4mm chip thickness the pulp yield increases as the composite permeability increases. At lower chip thickness, probably the reaction proceeded too far, resulting in degradation of the carbohydrate fractions. This is also supported by a higher alkali consumption at lower chip thicknesses (Table 2). If the permeability of the wood species is high, higher chip thickness should be used. Although there is a several fold difference in permeability within the heartwood, thickness of 3mm yielded the best results.

The influence of composite permeability chip thickness can also be seen from the degree of delignification. Within the same location a higher permeability yielded a pulp of lower kappa number, or in other words favoured better delignification (Fig. 10). Samples from different locations yield different lines for permeability and kappa number, indicating that some other factors are also involved in the process. This is, however, beyond the scope of this investigation.

When kappa number was plotted against chip thickness, two different sets of curves were obtained (Fig. 11). For sapwood and outer heartwood the curves had a negative slope i.e. kappa number decreased with 62 increasing chip thickness indicating that the reactions proceed uniformly even at higher chip thickness, and there is no difficulty in removal of lignin. In the middle heart and inner heart portions kappa number tended to increase with increasing chip thickness, indicating that the lignin in these portions is more difficult to remove. This opposite relation may be arising from either the low permeability of middle heart and inner heart or from the difference in extractive content.



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CONCLUSIONS

Pulp yield showed an increasing trend with increasing permeability when standard sized chips were used. Composite permeability to pulp liquor decreaced significantly with increase in chip thickness. Kappa number increased as the chip thickness increased showing a low degree of delignification with decrease in permeability. Lignin free yield was found to be related to composite permeability. At higher permeability, a lower chip thickness resulted in lower yeild indicating an over reaction, At 3mm chip thickness yield permeability curve was almost horizontal. A higher permeability thus favoured a higher chip thickness for higher yield.

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