

# INFLUENCE OF PERMEABILITY ON PULPING BEHAVIOR OF TROPICAL INDIAN HARDWOODS

*Satish Kumar*<sup>1</sup>

Senior Research Officer  
Forest Research Institute  
Dehra Dun, India

*Man Mohan Singh*

Director, R&D  
Orient Paper Mills (formerly Senior Research Officer, Forest Research Institute)

*P. B. Dobriyal*

Research Assistant  
Forest Research Institute  
Dehra Dun, India

and

*Sonya Bahri*

Former Research Scholar  
Forest Research Institute  
Dehra Dun, India

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## ABSTRACT

In this paper, an attempt has been made to correlate pulping of six tropical Indian hardwoods with their permeability and penetration behavior.

A poor correlation existed between pulping suitability indices and directional permeability. Pulping suitability indices were found to be poorly correlated even with the composite permeability of wood, indicated by loading of cooking chemicals into wood under pressure. Composite permeability of wood was found to have a good correlation with pulp yield. High composite permeability, an index of good penetration of the cells, always resulted in lower yield losses, e.g., in udal and kokko. A medium permeability resulted in medium yield (maharukh). A poor composite permeability resulting from poor penetration of the cells, despite high gas permeability in some cases (chilauni, eucalyptus, toon) resulted in higher carbohydrate losses.

**Keywords:** Hardwoods, permeability, diffusion, composite permeability, suitability indices, wood structure, pulping.

## INTRODUCTION

Various pulping processes are aimed at freeing the carbohydrate fractions of wood from lignin through selective dissolution of the latter from the middle lamella as well as the cell walls of the fibers. Pulp yield and quality are the two major criteria for deciding the suitability of any cellulosic material for making

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<sup>1</sup> Present address: Department of Forest Products, Oregon State University, Corvallis, OR 97331 (until March 1988).

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pulp. Low yield materials may be uneconomic to pulp, while low strength pulps will need blending with stronger pulps for getting requisite strength in the paper sheet.

The available information on molecular association of various wood components indicates that lignin and hemicelluloses form a three-dimensional interpenetrating matrix surrounding the cellulose microfibril. Selective action of pulping chemicals on lignin is therefore not practical without affecting the cellulose and hemicelluloses, resulting in lower yield and degradation in strength. Major losses, however, occur if the wood is not uniformly penetrated by the pulping liquor during the cook.

Although the role of wood permeability in pulping was recognized during the late fifties, no detailed study to correlate it with pulping behavior has been undertaken. Stone (1956) evaluated a number of softwoods and hardwoods for air flow characteristics and penetrability to pulp liquor, but did not quantify the suitability limits with respect to permeability. The broad conclusions indicated that Englemann spruce and white oak were practically impossible to penetrate by pulp liquor. Tylose-free hardwoods provided rapid flow paths through the vessels and were found suitable for production of pulp (Stone and Green 1958).

The importance of chip thickness, recognized about 40 years ago (Backman 1946) is an indirect approach to account for permeability behavior of wood. On the basis of studies on various softwoods and hardwoods, Hatton (1972) proposed a new term called "Liquid Accessibility Factor" as a guide to uniform pulping.

The Indian paper industry uses both bamboo and mixed hardwoods as furnish for various paper grades. Although a bleached yield of 31–57.5% has been obtained in laboratory studies from various wood species, statistics on production of paper and raw materials used show that the average screen yield in industrial pulping does not exceed 33% (Kumar 1980). Also, more than 70% of Indian hardwoods are difficult to impregnate or are only partially penetrable (ISI 1982). This appears to be one of the main reasons for low screen yields of mixed hardwood pulps. Some efforts have been made in the past to classify various hardwoods on the basis of their papermaking characteristics (Guha 1969; Misra 1972). The approach followed by Singh and Bhola (1976), however, seemed to be more appropriate as it included the cost of pulping based on total chemicals used in addition to pulp yield and pulp properties.

In this paper, an attempt was made to correlate pulping behavior to the wood directional permeability, penetrability by water/pulp liquor, and anatomical features.

#### MATERIALS AND METHODS

The species selected for study were chilauni (*Schima wallichii* Choisy), eucalyptus (*Eucalyptus* hybrid), kokko (*Albizia lebbeck* Benth.), maharukh (*Ailanthus excelsa* Roxb.), toon (*Toona ciliata* Roem) and udal (*Sterculia villosa* Roxb.). Samples 20 mm in diameter were obtained in the three structural directions with a plug cutter. Samples were obtained from heartwood portions. Samples were cross-cut to 25-mm length in case of axial flow. Smaller lengths were used for transverse flow because of very low flow rates encountered in transverse direction even at high pressure drops in some wood species. Both ends of the samples were microtomed to eliminate differences due to surface roughness (Choong et al. 1975).

All samples were conditioned to about 9% moisture content by storing over a saturated salt solution of sodium dichromate in a desiccator at 25 C.

Nitrogen flow rates were measured across the dowel samples using gas permeability apparatus fitted with air flow meters (rotameters) (Kumar 1979). Flow rates were plotted against pressure (pressure drop  $\times$  average pressure), and linear regressions were run for points falling within the streamline flow region. These regression equations were used to compute flow rates at three pressure drops (500-mm water column, 200-mm and 400-mm mercury column) for all the samples. Average permeability coefficients were calculated from these data using Darcy's flow equation for compressible fluids under steady-state conditions (Kumar and Chaubey 1987). At least ten samples were used for each species in each direction of flow.

Permeability samples used for axial permeability measurements were divided into two sets of five samples, so as to cover the entire permeability range. One set was impregnated with water to obtain composite permeability. The samples were submerged in water in a beaker using weights, and a pressure of 7 kg/cm<sup>2</sup> was applied for 20 minutes in a pressure cylinder. Samples were wiped with filter paper and weighed to obtain the water uptake. Air voids present in each sample were obtained from the specific gravity and swollen volume. The other set was treated with pulp liquor following a similar procedure. Percent voids filled with water and pulp liquor after making adjustments of water adsorbed by the cell wall (from 9% to fiber saturation point) was taken as the composite permeability to the respective fluid.

Suitability indices for pulping were obtained from pulping standard-sized mill run chips under identical cooking conditions. Cooking pressure (5.6 kg/cm<sup>2</sup>), time (4 hours), wood : liquor ratio (1:4), kappa number ( $25 \pm 1.5$ ) and sulphidity (25%) were kept constant for all cookings. Active alkali used was varied between 18–28% for different species in order to get pulps with a kappa number range of  $25 \pm 1.5$ . Total chemicals consumed and the pulp yield were used to calculate the pulping costs (principal basic property). Breaking length, burst factor, and tear factor were used as auxiliary properties to evaluate the suitability factor (SF) as described by Singh and Bhola (1976).

$$SF = \frac{P_0 W_0 + \sum_{x=1}^{x=n} P_x W_x F_x}{W_0 + W_1 + W_2 \dots W_n}$$

where  $P_0$  is the principal basic property;  $W_0$  weightage factor for  $P_0$ ;  $P_x$  is the auxiliary basic property and  $W_x$  is the weightage factor and  $F_x$  is the adjusting factor for the various auxiliary properties.

Suitability indices were obtained by dividing the SF values obtained for individual wood species by S'F obtained for bamboo, a standard material used by the Indian industry:

$$S.I. = \frac{SF}{S'F} \times 100$$

The suitability indices and the properties used to arrive at the same have been listed in Table 1.

Blocks of the size 35  $\times$  35  $\times$  15 mm of all the species were prepared from

TABLE 1. *Comparative suitability indices of hardwoods studied.*

| Species          | Unbleached pulp yield, %                | Strength properties of unbleached sheets |                                    |                                   | Comparative suitability index<br>$\frac{SF}{S'F} \times 100$ |
|------------------|-----------------------------------------|------------------------------------------|------------------------------------|-----------------------------------|--------------------------------------------------------------|
|                  | Total chemicals, %<br>(P <sub>0</sub> ) | Breaking length<br>m(P <sub>1</sub> )    | Burst factor<br>m(P <sub>2</sub> ) | Tear factor<br>m(P <sub>3</sub> ) |                                                              |
| Weightage factor | 2                                       | 2                                        | 1                                  | 1                                 | —                                                            |
| Adjusting factor | 1,000                                   | 0.000323                                 | 0.0468                             | 0.0164                            | —                                                            |
| Bamboo*          | 41.0/20.0                               | 6,340                                    | 43.8                               | 125.0                             | 100.00                                                       |
| Chilauni         | 36.2/28.0                               | 7,150                                    | 44.4                               | 101.2                             | 89.03                                                        |
| Eucalyptus       | 37.0/18.0                               | 5,110                                    | 37.0                               | 100.5                             | 88.00                                                        |
| Kokko            | 45.0/22.0                               | 9,000                                    | 57.8                               | 115.0                             | 117.84                                                       |
| Maharukh         | 48.2/22.0                               | 7,200                                    | 34.4                               | 74.2                              | 96.40                                                        |
| Toon             | 44.5/25.0                               | 8,420                                    | 54.1                               | 86.4                              | 105.26                                                       |
| Udal             | 49.2/22.0                               | 8,500                                    | 50.0                               | 83.0                              | 111.08                                                       |

\* Standard raw material used by the industry.

heartwood portions. Five percent solution of silver nitrate was impregnated into the submerged blocks under vacuum in a desiccator, followed by an air pressure of 3.5 kg/cm<sup>2</sup> for 30 minutes in a pressure cylinder. Blocks were further treated with a 2% solution of hydrazine dihydrochloride solution and kept in bright sunlight for nearly two hours for rapid fixation of silver ions to avoid any lateral movement due to diffusion. The treated blocks were split in the middle, and 12 to 15 micron sections were prepared on a microtome. The sections were mounted temporarily in glycerine for examination under the light microscope to study the penetration pattern followed in various tissues in each species.

## RESULTS AND DISCUSSION

*Influence of directional permeability on pulping behavior*

Permeability and pulping data, along with comparative suitability indices for pulping (Singh and Bhola 1976) are tabulated in Table 2. Longitudinal flow in hardwoods primarily controlled by vessels varied accordingly depending upon their number, size, and condition, i.e., whether open or filled with tyloses, gum deposits, etc. (Kumar 1979). Although it is believed that highly permeable timbers should be most suitable for pulping, suitability indices obtained for various hardwoods showed a very poor relation (0.62) with the magnitude of axial gas permeability in this study (Fig. 1). It has been observed in some hardwoods that the

TABLE 2. *Directional permeability and pulping behavior of six tropical Indian hardwoods.*

| Wood species | Total holo-cellulose content, % | Apparent nitrogen permeability |           |               | Composite permeability (% voids filled) |             | Suitability index* |                            |
|--------------|---------------------------------|--------------------------------|-----------|---------------|-----------------------------------------|-------------|--------------------|----------------------------|
|              |                                 | d Axial                        | md Radial | Tangential md | Water                                   | Pulp liquor | Yield wood basis   | Yield holo-cellulose basis |
| Chilauni     | 70.7                            | 0.394                          | 0.014     | 0.08          | 47                                      | 40          | 89.0               | 89.1                       |
| Eucalyptus   | 66.9                            | 0.006                          | NA        | NA            | 21                                      | 25          | 88.0               | 88.9                       |
| Kokko        | 62.7                            | 0.331                          | 0.002     | 0.05          | 93                                      | 70          | 117.8              | 122.3                      |
| Maharukh     | 71.3                            | 0.0175                         | 1.020     | 0.47          | 63                                      | 60          | 96.4               | 96.3                       |
| Toon         | 75.9                            | 3.926                          | 0.060     | 0.04          | 31                                      | 42          | 105.3              | 103.2                      |
| Udal         | 65.7                            | 16.200                         | 0.300     | 0.60          | 93                                      | 69          | 111.1              | 114.1                      |

NA = Not available.

\* = Calculated with respect to bamboo taken as 100.

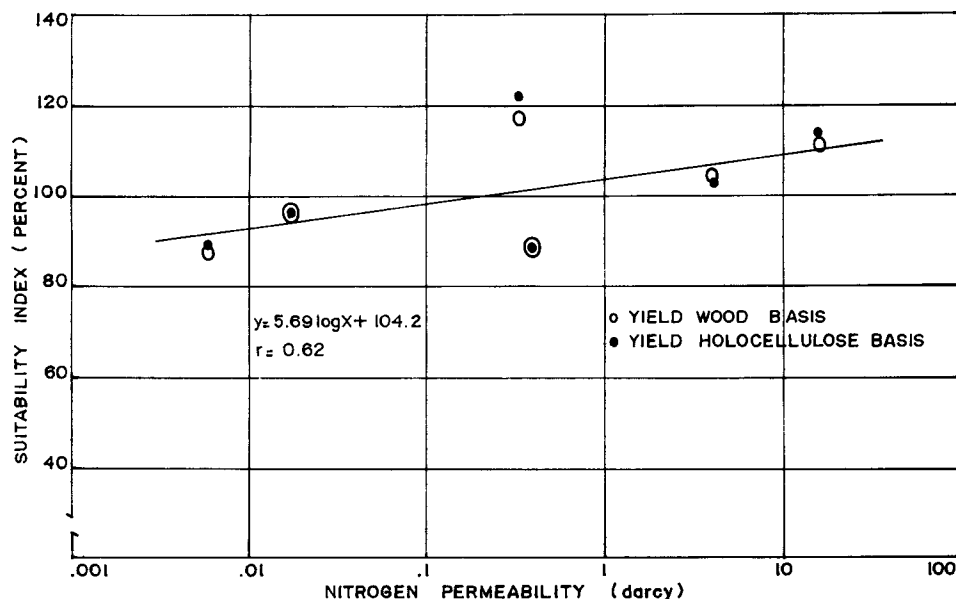


FIG. 1. Influence of axial permeability on pulping suitability of different wood species.

gaseous flow is not totally viscous as assumed in application of the Darcy equation and there is a varying amount of slip flow component, whereas in flow of liquids, no slippage occurs (Kumar 1979). Even if the effects of slippage are eliminated, the correlation is not affected significantly.

Since vessels play a key role in fluid penetration in hardwoods, earlier workers concluded that hardwoods free from tyloses would behave better during pulping (Stone 1956). While it may be qualitatively true, quantitatively the presence of a single large vessel or a cluster of medium-sized tylose-free vessels would contribute enormously to longitudinal permeability, while other cell tissues may not be penetrated at all, thus leading to nonuniform delignification reaction within the chip. This may be one of the reasons for the absence of any correlation between axial permeability and pulping indices or pulp yield (Figs. 1 and 2).

A higher holocellulose content in wood would result in a higher pulp yield. There was a variation of up to 13% in the total holocellulose content among the wood species studied (Table 2). In evaluating suitability indices, this factor was not taken into consideration by earlier workers (Singh and Bhola 1976). However, suitability indices recalculated from pulp yield expressed as percent of total holocellulose content also did not improve correlation with axial permeability (Fig. 1). No clear trend was evident between transverse permeability and pulp yield.

If pulp yield was taken as the only criterion for suitability of wood, the relationship between axial permeability and pulp yield was not improved (Fig. 2). Deviations from linearity were attributable to species differences in terms of overall penetrability of the various cell tissues. The importance of permeability in pulping behavior has been elucidated by comparing results of pulp yield from wood samples from different permeability zones within the same wood species. Figure 3 shows the relationship between wood permeability and pulp yield as obtained from *Eucalyptus* hybrid from different permeability zones viz. sapwood,

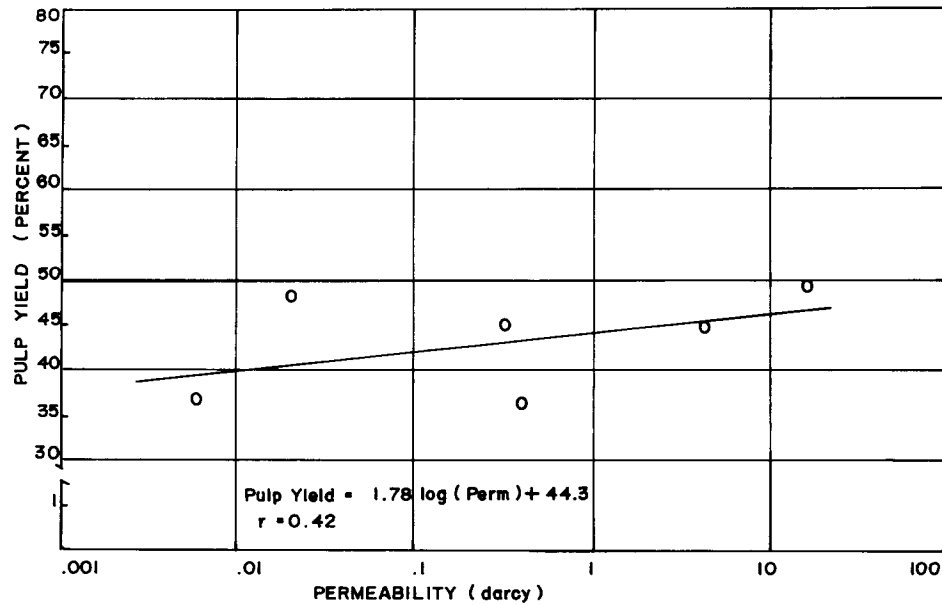


FIG. 2. Pulp yield as affected by permeability in different hardwood species.

outer heartwood, middle heartwood, and inner heartwood. The straight line relation clearly demonstrates the role of permeability in obtaining higher pulp yields.

#### Diffusion

Penetration of pulping liquor in wood is complex, and both pressure and temperature play important parts. In the initial stages of pulping, the penetration of pulp liquor is usually slow, as it enters only the most permeable structures such

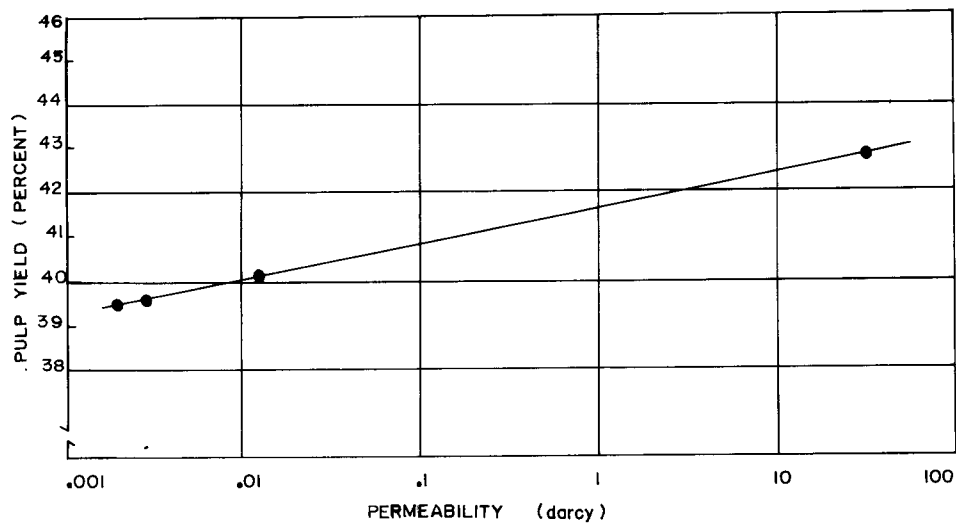


FIG. 3. Pulp yield as affected by permeability in different zones of eucalyptus. From left to right the points refer to inner heartwood, middle heartwood, outer heartwood, and sapwood.

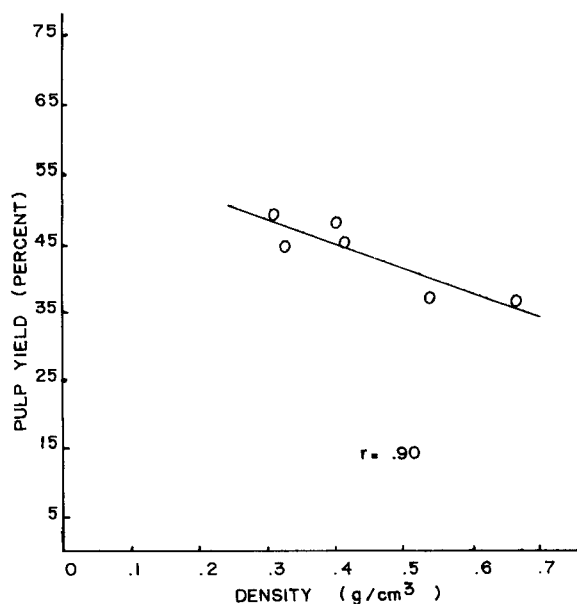


FIG. 4. Effect of wood density on pulpwood yield.

as vessels and a few contiguous fibers in case vessel-fiber pits are permeable. Rays, if void of any deposits, may also be filled, but in tropical hardwoods rays are usually occluded. This partial penetration is, however, not said to affect the pulp yield adversely as the process of delignification starts only at 130–140 C. With an increase in temperature and pressure, the penetration is supposed to be complete due to softening of tissues, dissolving of gums, etc. and lowering of viscosity (Paranyi and Rabinovitch 1955). Lateral penetration is mostly accomplished by diffusion of cooking liquor through the cell wall, and cooking liquors being alkaline diffuse through wood at almost equal rates in all directions, so that anisotropy of wood has little significance at this stage (Hartler 1962; Stone and Green 1958).

No data are, however, available for diffusion of pulping liquor through woods of different densities. Diffusion coefficients for water vapor have been found inversely proportional to the square of density and an increase in temperature favors a faster diffusion (Bains and Kumar 1979; Kumar and Purushotham 1972; Yao 1966). The delignification reaction is thus diffusion controlled. Plot of pulp yield versus density of wood depicts a decreasing trend with increasing density (Fig. 4). The correlation ( $R = 0.90$ ) is significant at the 5% level. Diffusion of alkaline pulping solutions is thus also controlled by the density of wood. Slow diffusion in denser woods results in a prolonged contact at the penetrated sites and thus lowers the pulp yields. Toon is the only exception where despite the low density, the pulp yield (%) is low. This behavior is probably due to its semi-ring-porous nature (all other species are diffuse porous). Because of this structural difference, most of the chemicals remained confined to pockets of larger vessels at the growth ring, causing a higher degradation of carbohydrate and higher chemical utilization. This is also proved by low filling of voids under pressure and the penetration pattern depicted in Fig. 5.

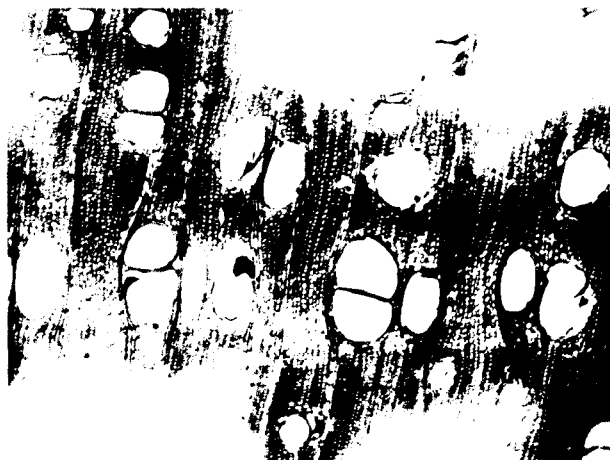


FIG. 5. Toon treated with silver nitrate. The stain is concentrated around vessels on growth ring (25 $\times$ ).

It is, therefore, essential that wood be uniformly saturated with the pulp liquor before the start of delignification so that diffusion may occur evenly in all directions resulting in uniform delignification. Diffuse-porous hardwoods were thus found to have an edge over ring-porous hardwoods, the latter requiring longer periods for pulping because of uneven distribution of the pulping liquor (Stone and Green 1959).

#### *Composite permeability versus pulping behavior*

Overall penetration under pressure occurs from all directions, and it is difficult to pinpoint the contribution of axial, tangential, or radial permeability in such impregnation. Since the entering fluid follows the path of least resistance, it is expected that axial permeability, being several thousandfold the transverse permeability (in many cases), will play a major role. This is especially so in chips where length-to-thickness ratio is not very high as compared to treatment of wood in lumber form. However, even in chips of some species like aspen or birch, a higher thickness has been found to lead to insufficient penetration to the core (Hatton 1978a, b). It is therefore the composite permeability of the material that controls the impregnation from all directions. Such permeation occurs under nonsteady-state conditions, and in the case of pulping liquor this is accompanied by deformation of the material as well, which is likely to further complicate the penetration process. The overall penetration may not thus be merely permeability-controlled. In the initial stages in the absence of pressure, there is capillary penetration, which is accompanied with swelling of the cell wall. Swelling has been found to reduce the permeability significantly (Kumar and Chaubey 1987). At higher pressures, the flow rates increase and there is every possibility of having turbulence in the wood structure so that the actual permeability of the wood further falls (Kumar 1979). Flow rates may further be affected because of an increase in viscosity of the pulp liquor as the reaction proceeds. Because of the complexity of the processes involved, it was but expected that no correlation of pulping (suitability index and pulp yield) with gas permeability would become

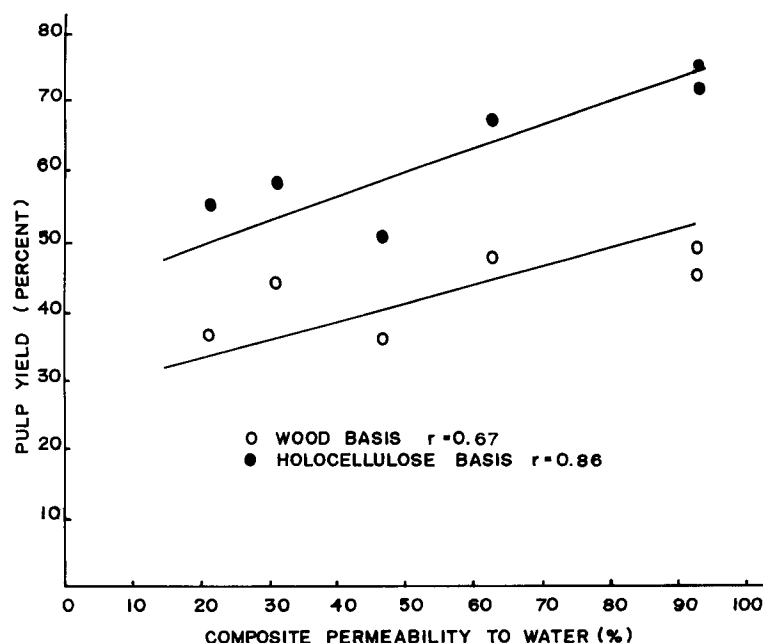


FIG. 6. Pulp yield as related to composite permeability to water.

apparent (Figs. 1 and 2). The absence of a good correlation, however, does not indicate that permeability is not at all a factor in pulping.

The amount of liquor that can be forced into wood under pressure gives an estimate of the composite permeability as well as the internal geometry of the wood structure. However, it has not always been possible to correlate this to absolute permeability even in nonreacting fluids. Most of the deviations are due to the existence of parallel paths of low and high permeability; larger capillary forces being required to fill low permeability tissues (Galvao et al. 1974). Particularly in hardwoods, the overall loading largely depends on the vessel volume and the permeability of the vessel-vessel, vessel-fiber, and fiber-fiber pit system. Water and pulp liquor absorption by different wood species (percent voids filled) have been listed as composite permeability in Table 2. Pulp yield expressed as percentage of wood weight showed a correlation of 0.67 with the percent wood voids filled with water (Fig. 6). The correlation improved to 0.86 (significant at 5% level) when the yield was expressed as percent of the total holocellulose content of wood. However, Kraft liquor due to its swelling nature may be absorbed differently in wood. Treatment with Kraft white liquor was therefore done under similar conditions (7 kg/cm<sup>2</sup> at ambient room temperature for 20 minutes). The correlation coefficient of pulp yield (wood basis) with alkali solution retention improved to 0.83 (significant at 5% level), while that of pulp yield (holocellulose basis) came to 0.92 (significant at 2% level) (Fig. 7). It may be seen that with a higher swelling effect of pulp liquor on wood, the alkali absorption was generally less than water absorption. Although impregnation was carried out at ambient room temperature so that no delignification was expected, the absorptions were retarded by a decrease in permeability due to swelling and also by an increase in viscosity of pulp liquor due to partial dissolution of wood components. White

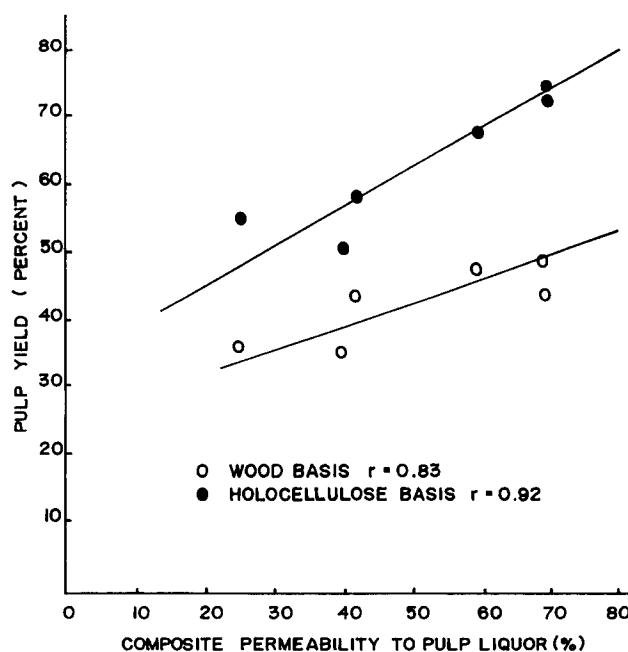


FIG. 7. Pulp yield as related to composite permeability to pulping liquor.

liquor changed to a brown color, and in some cases, it was quite viscous. Pulping suitability indices were, however, found to be poorly correlated even with the composite wood permeability to water or alkali. Suitability indices obtained from purely arbitrary parameters thus do not seem to be appropriate for evaluation of hardwoods.

#### *Penetration pattern vis-à-vis pulping behavior*

In hardwoods many individual factors are responsible for overall impregnation with pulping liquors. These factors are wood density, percentage of vessel area, vessel-vessel, vessel-fiber, and fiber-fiber communication systems and penetrability of rays. Penetration pattern in different wood species along with other data are presented in Table 3. Although low density provides a higher void volume for absorption of pulp liquor and thus may lead to higher yield coupled with low chemical consumption (e.g., udal), the results can be otherwise if the penetration pattern is not uniform as observed in the case of toon (Fig. 5). Vessel area becomes more important in denser woods. A higher vessel area can result in higher absorptions of the liquor and in the case of diffuse porous woods, such liquor being uniformly distributed leads to a uniform pulping, resulting in a higher pulp yield or low carbohydrate losses at low chemical consumption (kokko, maharukh—Table 3) irrespective of the penetrability of the various tissues. Vessel-fiber, fiber-fiber, and ray communication systems assume greater importance, in case vessel volume is low. Udal, with a low vessel volume but a good communication system, lost only 16.5% carbohydrates, whereas toon with a poorer communication system lost over 30% carbohydrates. Toon being semi-ring-porous, the distribution of the chemicals was limited to growth ring area, where these vessels were concen-

TABLE 3. Penetration pattern of different cell types and carbohydrate loss in different hardwood species as affected by density and vessel area.

| Wood species | Density<br>g/cm <sup>3</sup> | Vessel area<br>on cross-<br>section (%) | Penetration of different tissues <sup>a</sup> |        |      | Permeability<br>to pulp<br>liquor <sup>b</sup> (%) | Chemicals<br>used (%) | Loss in car-<br>bohydrate<br>content (%) |
|--------------|------------------------------|-----------------------------------------|-----------------------------------------------|--------|------|----------------------------------------------------|-----------------------|------------------------------------------|
|              |                              |                                         | Parenchyma                                    | Fibers | Rays |                                                    |                       |                                          |
| Udal         | 0.307                        | 13.9                                    | +++                                           | +      | +++  | 69 (H)                                             | 22                    | 16.5                                     |
| Toon         | 0.326                        | 10.8                                    | ++                                            | +      | ++   | 42 (L)                                             | 25                    | 31.5                                     |
| Maharukh     | 0.400                        | 23.8                                    | +                                             | —      | —    | 60 (M)                                             | 22                    | 23.1                                     |
| Kokko        | 0.412                        | 21.2                                    | ++                                            | —      | +    | 70 (H)                                             | 22                    | 17.7                                     |
| Eucalyptus   | 0.541                        | 6.3                                     | +++                                           | +      | +    | 25 (L)                                             | 18                    | 29.9                                     |
| Chilauni     | 0.655                        | 15.4                                    |                                               |        |      | 40 (L)                                             | 28                    | 34.5                                     |

<sup>a</sup> +++ Heavily penetrated, almost all cells showed penetration.

++ Moderately penetrated.

+ Scarcely penetrated, only a few cells showed penetration.

— Not penetrated.

<sup>b</sup> Permeability rating: <50 Low (L), 51–60 Medium (M), 61–70 High (H).

trated. Conversely denser woods, with low vessel volume and poor communication from vessels to other cells end up with poor yield and high losses in carbohydrate content (chilauni and eucalyptus).

#### CONCLUSIONS

Although no direct correlation between pulping suitability indices and gas permeability could be established, pulp yield has been found to be directly correlated to the composite permeability to the pulp liquor. Pulp yield has also been found inversely correlated to the density of wood.

Diffuse-porous hardwoods have been found to have a slight advantage over semi-ring-porous woods as the chemicals are uniformly distributed in the former, leading to more uniform pulping. A better communication system between vessels and other cells favors better pulping with less loss in carbohydrate content. A large vessel volume, however, may compensate for poor communication between other cells, especially in denser woods.

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