MICROSCOPICAL ASPECTS OF HARDWOOD REFINER PULPS

by

HECTOR A. CISNEROS

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Department of Forestry

The University of British Columbia
Vancouver, Canada

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ABSTRACT

In order to gain insights into ultrastructural changes taking place during the conversion of hardwoods into mechanical pulps, refiner pulp fibres were studied in detail using several microscopical techniques. Aspen (*Populus tremuloides* Michx.) and white birch (*Betula papyrifera* Marsh.) wood chips were used to produce thermomechanical (TMP), chemithermomechanical (CTMP) and chemimechanical (CMP) pulps. Following the hypothesis that there are fundamental differences in the surface and state of the fibres due to species and processing conditions, four pulps for each species and process were analyzed. Trends in fibre characteristic development were obtained within each group, based on the detailed optical analysis of 300 fibre cross-sections for each pulp.

Fibre surface quality was the most important aspect of this study. Retention of middle lamella and of the S_1 layer, as well as the extent of exposure of the S_2 layer were evaluated. It was found that TMP processing of wood chips produced fibres with more exposure of the S_2 layer. Chemical pretreatment did not improve the extent of S_2 layer exposure nor the extent of fibrillation. However, the TMP fibres remained stiff, producing pulp sheets of low density and strength.

Birch fibres showed a marked tendency to produce separation at or near the S_1/S_2 boundary. This resulted in high exposure of S_2 layers in TMP fibres, but produced a sheath

of S_1 and ML around fibres from chemically-treated chips. This sheath was sometimes rolled back, exposing the fibre S_2 layer. Aspen TMP pulps showed high proportions of fibres with partially exposed S_2 layer. The application of chemical pretreatments to aspen chips resulted in fibres of similar levels of S_2 exposure than those achieved by TMP processing of this species, but only after reaching freeness levels of about 100 mL CSF.

Fibres that showed radial failure were frequent in TMP but not in CTMP nor CMP pulps. The breakdown pattern of tension wood fibres (G-fibres) was also studied. TMP processing showed preferential breakdown of G-fibres, from which the G-layers were freed. This was not the case in the G-fibres from chemically-treated chips, in which the G-layer generally remained inside the fibres. Other categories discussed in the analysis of fibre cross sections included fibres with delamination of the S_2 layer and proportion of fibres distorted due to chemical impregnation.

The breakdown of vessel elements (VE) was studied by comparing VE size frequency distributions and the proportion of whole VE that survived refining. TMP reduced VE into small fragments showing virtually no whole VE, while wood softening due to chemical pretreatment was responsible for a high proportion of whole VE in CTMP and CMP pulps. The VE from birch tend to be destroyed more easily than those from aspen, due to the intervessel pitting arrangement of the former.

It is concluded that despite superior bonding potential of TMP fibres due to:

- large S_2 exposure in fibres on account of separation at or near the S_1/S_2 boundary,
- increased fibrillation,
- longer fibrils in fines, and
- release and exposure of highly cellulosic G-layers from tension wood in the case of aspen,

the lack of conformability of TMP fibres, which translates into low sheet density, negates the promising benefits that otherwise would be obtained.

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I. INTRODUCTION

The use of hardwoods for pulp and paper products has increased considerably in recent years, primarily because of the decreasing availability of traditional old-growth coniferous raw materials, and also because of recent developments in demonstrating the suitability of hardwoods for the manufacture of printing and writing papers. For example, the world's total capacity for the manufacture of hardwood pulps for their conversion into paper and paperboard products has increased from 32.6 million air-dry metric tonnes per year in 1982 to 39.3 million adt/year in 1988. It is estimated that by 1993 the capacity will grow to 47.4 million adt/year (Anon. 1983, 1989a).

Printing papers are often manufactured with a high-yield mechanical pulp as the major pulp component. This provides a furnish with the proper balance of optical and strength properties. Hardwood mechanical pulps, and alternative furnishes offered by a plethora of refiner mechanical pulping systems, are becoming important members of the family of mechanical pulps available today. It is anticipated that their use will continue to grow in Canada and elsewhere.

The manufacture of high-yield pulps requires the application of mechanical energy to separate the fibres from the wood. Due to the nature of the process, these pulps contain a variety of particle sizes and shapes. These particles can

range from fibre bundles to very fine fragments of the cell wall. The pattern of breakdown of the wood into fibres and, in turn, into small fragments and fines, is important from the standpoint of the ultimate quality of the pulp. Fibres or tracheids are the major component in the wood material so that a large portion of the smaller fractions will originate from these. The quality of the different fibre fractions is determined by the structure of the fibres themselves which, in turn, may be modified by the conditions of pulping. It is anticipated, then, that examination of the long fibre component of a pulp will provide an indication, not only of the breakdown pattern, but also of the quality of the smaller fractions, and the overall quality of the pulp.

Almost any tree species can be reduced to mechanical pulp by the refiner process (Sugden 1967). In general, the resulting strength properties of hardwoods pulps are low and not generally satisfactory for inclusion in printing papers, unless some type of chemical treatment is applied during the pulping process (Atack and Heitner 1982, Marton *et al* 1979). Hardwood fibres appear to have a more rigid structure and a relatively thicker S_1 layer than softwood tracheids. This is thought to prevent the peeling-off of the S_1 layer and fibrillation of the S_2 layer (Marton *et al* 1979) during mechanical pulping. The S_1 layer is considered to impede flexibility of hardwood fibres (Law *et al* 1985). However, upon application of a mild chemical treatment to the chips, the wood is softened, the fibres become more flexible, and

the primary wall and S_1 layer are peeled off in a manner similar to what is observed in spruce TMP fibres (Giertz 1977). The implication is that, in order to access the S_2 layer in hardwood fibres and to promote fibrillation and production of fibrillar fines, a chemical treatment is necessary. Thus, the retention of the S_1 layer appears to be detrimental to pulp strength. The exposure of the S_2 layer is desirable because of its higher ratio of cellulose to lignin content compared to the layers external to it. Therefore, it possesses the highest interfibre bonding potential. This is particularly true in the case of hardwoods, in which the relative concentration of carbohydrates in the secondary wall is higher than in the case of softwoods (Rydholm 1965).

However, the extent of retention of the S_1 layer --with or without the compound middle lamella-- and the resulting exposure of the S_2 layer has not been studied in a quantitative manner for a large number of isolated hardwood fibres after their separation from the wood matrix. Of specific interest are the effects of mechanical pulping alternatives, wood species and refining energy consumption levels on the pattern of removal of the outer layers of the cell wall. This can be best studied by examination of fibres in cross section.

One technique for assessing the surface quality of a fibre was developed to study lignin distribution patterns after a certain pulping treatment. The process uses ultra-violet

light microscopy of fibre cross sections (Wardrop *et al* 1961, Kerr and Goring 1976, Gadda *et al* 1981, Bruum and Lindroos 1983). Analysis of middle lamella retention has also been done using staining techniques for softwood mechanical pulp fibres in cross section (Kibblewhite 1983, Williams 1989). A second technique examines the exposed fibre surface under a transmission electron microscope. For this technique the sample preparation is laborious, and the number of fibres which can be analyzed is limited (Iwamida *et al* 1980a, Wardrop *et al* 1961). The method makes use of the orientation of microfibrils on the fibre surface to define the identity of the exposed layer.

The analysis of fibre cross-sections, although not an entirely new tool, has not been used for the quantitative analysis of hardwood mechanical pulps. The use of this technique could potentially provide answers to questions of fibre surface quality, and also of other fundamental features useful in elucidating aspects of hardwood refiner pulping. The possibilities offered by the examination of fibre cross-sections in terms of fibre quality and fibre breakdown appear to be worth exploring.

Examination of fibres in cross-section, in addition to those features already mentioned regarding elucidation of fibre surface development, can provide information on the manner in which the outer layers separate from the fibre; on the extent of damage to fibres, including cell wall delamination and radial failure; on the proportion of chemically-treated

fibres in a refiner pulp produced from chemically-treated wood chips, as well as the extent of the treatment itself; and on the presence and breakdown pattern of tension wood fibres commonly present in some hardwood species. These features, combined with the use of other microscopy techniques, should provide reliable explanations for the behavior of hardwoods in refiner pulping and set the basis for process improvements leading to better utilization of these species.

The amount of microscopical detail provided by such a study is expected to contribute to the understanding of the behavior of hardwood mechanical pulp fibres under intense mechanical shear stresses which, ultimately, determine the properties of these fibres.

Another aspect of interest in the mechanical pulping of hardwoods is the breakdown of vessel elements (VE). Vessel elements are conducting cells, characteristic of the wood of angiosperm trees, generally shorter and much wider than the fibres of the parent wood. Thus, their shape is not conducive to bonding within a paper sheet and can cause serious problems during the printing process by picking in the printing press (Marton *et al* 1979, Colley 1973). This is particularly troublesome in offset printing which consists of the application of high-viscosity inks to the paper surface. Thus, fast printing requires papers of high surface strength and integrity, and good resistance to picking. Previously, about 50% of the whole vessel elements present

in the wood have been estimated to survive mechanical pulping despite the refining action (Marton *et al* 1979). Since the fibres and, presumably, the VE of high yield pulps are more rigid in nature than those of chemical pulps, VE can be a problem when increasing proportions of these pulps are used in the manufacture of printing papers.

The size-distribution of VE in hardwood mechanical pulps under different mechanical pulping conditions, and their survival as entire entities upon refining, can provide basic information of the potential picking problems when large components of hardwood mechanical pulps are used. Vessel element size modification after mechanical pulping is an important task in eliminating the picking problem associated with printing papers made of hardwood mechanical pulps.

With these questions in mind, the following objectives were established in this thesis:

a) To investigate the response of two important North American hardwoods to refiner mechanical pulping conditions,

b) To characterize such hardwood refiner mechanical pulps by detailed examination of the fibre cross-sections as to the state of their cell walls,

c) To study the breakdown of vessel elements upon refining.

In order to achieve these objectives, the following hypotheses were tested in this study:

a) Different hardwoods respond differently to mechanical defiberization.

b) The optical analysis of fibre cross-sections can reveal fundamental species' responses, and provide quantitative differences on fibre structural changes as a result of differences in species and application of different processes and refining energy levels. Surface properties of the fibres should prove to be dependent on species as well as on chip processing conditions. Other relevant fibre cross-sectional categories may be explored and developed based on the optical observations. These include, but are not limited to, fibre damage, the proportion of fibres chemically affected by liquor penetration, and the breakdown characteristics of tension wood fibres (G-fibres).

c) A wide range of features could be recorded by fibre cross-sectional analysis, and the statistical analysis of the data collected is expected to uncover some relationships with properties of the pulp. In particular, quantitative surface properties observed for the fibres are expected to relate to the strength properties of the pulp.

d) The breakdown pattern of vessel elements and the number of whole vessel elements that survive the refining of

wood chips is largely dependent on chip processing conditions. However, due to species differences in the dimensions and structure of the vessel elements, a species effect may also be important.

II. LITERATURE REVIEW

2.1. Importance of Hardwoods

Canada's pulp and paper economy is primarily based on softwood market pulps and newsprint. Utilization of hardwoods is only on a relatively small scale. This is particularly true for medium to high density hardwoods, not only in Canada and other temperate zones but also in the tropics.

Today, however, with the increasing acceptance of hardwoods, opportunities abound for the utilization of this resource. Hardwoods in Canada comprise 5.32 billion m^3 of merchantable timber (Anon. 1989b) of which *Populus* species comprise 56%. Aspen (*Populus tremuloides* Michx.) is the major species, growing throughout the forested areas of Canada (Hosie 1979). In B.C., aspen forests account for a total net volume of over 220 million m^3, a resource still virtually untapped (Zak 1989). Birch is the second most abundant hardwood in Canada, with a merchantable volume of over one billion m^3 distributed throughout most of Canada.

In northern Alberta, there is much current activity with new projects that will make use of the large available hardwood resource (Sims 1989). In eastern Canada, where substantial losses of softwood fibre have resulted in the 70s and 80s from spruce budworm infestation, a raw material supply gap is foreseen for the pulp and paper industry between the harvest of sound old-growth timber and that of second-growth

timber (Bird 1985). Hardwoods have the potential to fill this supply gap provided they can be processed satisfactorily. These factors are stimulating interest and incentive in the production of mechanical pulps from hardwoods.

The successful conversion of hardwood species into mechanical pulp requires somewhat different processing conditions than those used for softwoods. Hardwood pulps are suitable for the manufacture of printing and writing papers for which the demand continues to grow at an approximate rate of 3% per annum. It is estimated that the total world capacity for newsprint and other printing and writing papers will increase from 100.9 million adt/year in 1988 to 112.5 million adt/year by 1992 (Anon. 1989a).

Hardwood pulps can impart good opacity, and provide good formation and surface finish to the final paper product. It is, therefore, anticipated that the utilization of hardwoods in pulp and paper products will continue to increase in the near future. Specifically for mechanical pulps an increase in the utilization of hardwood pulps has already been manifested. In the last mechanical pulping surveys (Leask 1989, 1990) it was reported that four new systems will be installed in Canada for the manufacture of mechanical pulps from hardwoods, adding 2120 adt/day of pulp to the world's mechanical pulp capacity. Furthermore, the production of hardwood mechanical pulps for the manufacture of printing papers is likely to continue to increase (Wood and Karnis

1989, Moldenius and Jackson 1989).

2.2. Refiner Mechanical Pulping

In the 1930's, Asplund, in Sweden, developed a method for the production of coarse grades of mechanical pulp for use in the manufacture of resin-bonded hardboard (Atack 1985). The method involved the steaming of wood chips for a few minutes at a pressure of about 800 kPa and a temperature of 170 ^{o}C, followed by defiberization in a pressurized single-rotating disc refiner. Then, in the late 1940's, atmospheric discharge disc refiners were used to produce semichemical pulps and to treat rejects from mechanical pulps produced by stone grinding. The small additional step required to disc refine wood chips and residuals was taken in the early 1950's and, in 1962, this system, known today as refiner mechanical pulping (RMP), was first used commercially to produce newsprint-grade pulps. In the refiner mechanical pulping process, wood chips are fed between two metal disks with at least one of them rotating. Defibration occurs by mechanical action as the chips are first broken by the large breaker bars located near the eye of the refiner. The coarse pulp produced in this manner is further refined as it passes through the middle zone and refining zone of the plate towards the periphery of the refiner.

In 1964, the first commercial pressurized refining system was installed in eastern Canada. Chips were steamed for a few seconds prior to being fed into a pressurised refiner.

After 1968, several new pressurised systems were installed in Sweden and the United States based on the original design of the Asplund Defibrator system. Pulps produced in this type of system were named thermomechanical pulps (TMP). This system involves steaming wood chips under pressure in a steaming vessel for 1-3 minutes, at a temperature between 105 and 130^{o}C, followed by pressurized refining. The casing of the refiner is maintained at a pressure between 200 and 300 kPa. The pulp is then refined to its final state in an atmospheric discharge refiner.

Today, many refiner pulping systems have added an impregnation stage to the process, so that the chips are chemically treated prior to refining. The overall production of refiner pulps has now surpassed that by the conventional stone grinding process.

The modification of some of the important variables in the disc refining system have resulted in the current major processes for the production of refiner mechanical pulps according to Atack *et al* (1980):

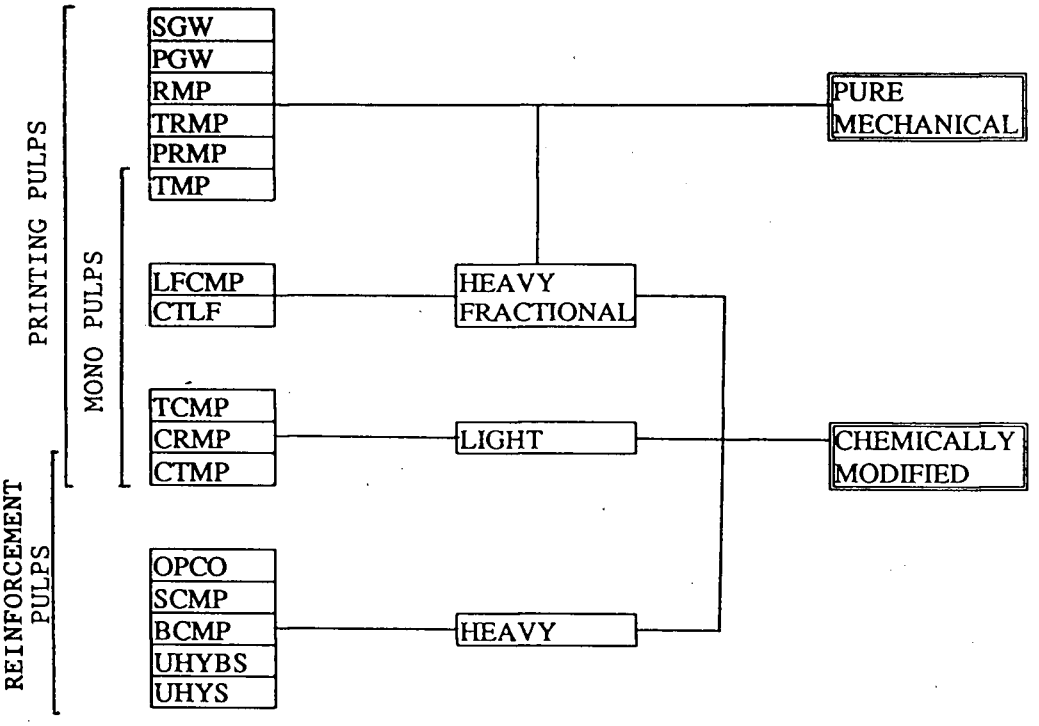
a) RMP: refiner mechanical pulping; atmospheric discharge refining of untreated chips,
b) TMP: thermomechanical pulping; presteaming and pressurized refining of untreated chips,
c) CTMP: chemithermomechanical pulping; presteaming and pressurized refining of treated chips, and
d) CMP: chemimechanical pulping; atmospheric

discharge refining of treated chips.

There are, however, many variations of these methods since refining can be, and usually is, done in more than one stage. Mild chemical treatments can be applied to the chips, to the first-stage pulp, or to the finished pulp. Thus, alternatives include chip treatment (Jackson 1988, Prusas *et al* 1987), inter-stage treatment (Barnet *et al* 1980), post-treatment, and refining of pulp rejects (Leask 1987). There are several patents in existence related to the specific processing conditions of these so-called "alphabet" pulps (Mackie and Taylor 1988). Figure 2.1 shows a more detailed classification of most mechanical pulping processes.

Reasons for the rapid expansion of mechanical pulps manufactured from chips, as opposed to stone grinding of bolts, include the possibility of using wood waste, such as residual chips and lower quality raw material like sawdust and shavings (Leask 1977); production of a substantially stronger pulp; simpler and more effective refiner and pulp quality control; and the potential of using low-level chemical treatments to produce pulps which can approach the strength of chemical pulps (White 1969).

The main criticism of disk refining has been its relatively high specific energy consumption (White 1969). There are, however, several ways of reducing energy usage in refining such as the application of chemical pretreatment to the wood chips, chip destructuring, uniform refining consistency,



MONO PULP:
Pulp that, because of its combination of optical and strength properties, can constitute 100% of a newsprint furnish.

PRINTING PULP:
Pulp that constitutes the bulk of a newsprint furnish but requires a strong pulp for reinforcement.

REINFORCEMENT PULP:
Strong high-yield pulp that can replace chemical pulps in a newsprint furnish.

SGW	STONE GROUNDWOOD
PGW	PRESSURIZED GROUNDWOOD
RMP	REFINER MECHANICAL PULP
TRMP	THERMO REFINER MECHANICAL PULP
PRMP	PRESSURIZED REFINER MECHANICAL PULP
TMP	THERMO MECHANICAL PULP
LFCMP	LONG FIBRE CHEMIMECHANICAL PULP
CTFL	CHEMICAL TREATMENT LONG FIBRE
TCMP	THERMO CHEMI MECHANICAL PULP
CRMP	CHEMIMECHANICAL PULP (ALSO AS CMP)
CTMP	CHEMI THERMO MECHANICAL PULP
OPCO	ONTARIO PAPER COMPANY PROCESS
SCMP	SULFONATED CMP (CIP PROCESS)
BCMP	BISULPHITE CMP
UHYBS	ULTRA HIGH YIELD BISULPHITE
UHYS	ULTRA HIGH YIELD SULPHITE

Figure 2.1. Mechanical pulping processes according to Franzen (1986).

improved plate design, and computerized control systems (Allan *et al* 1968, Atack 1980, Gavelin 1982b, Hartler 1980). The most important means to offset energy consumption is probably by heat recovery when refining under pressure (Gavelin 1982b).

Although most of the mechanical pulps produced are used in newsprint manufacture, a large variety of products contain substantial amounts of mechanical pulps including printing and writing papers, tissues and towelling, diapers, liquid-packaging boards, wallpaper, coated paper and other specialty papers (Leask 1982).

2.3. Characterization of Mechanical Pulps

A model of the layered structure of a fibre is given in Appendix A for a better understanding of the tranformation of a wood chip into fibres and fibre particles that takes place during refiner mechanical pulping.

When wood is subjected to mechanical attrition, the resulting pulp is composed of particles of different shapes and sizes. The different particles can be classified into five categories as shown in Figure 2.2, according to Mohlin (1982a).

Today's fractionation techniques are unable to separate completely the particles according to Figure 2.2. When using a Bauer-McNett Classifier, for example, shives and fibre bundles are found in the coarse fibre fraction (R50 mesh), fibre fragments and ribbon-like lamellae mainly in the

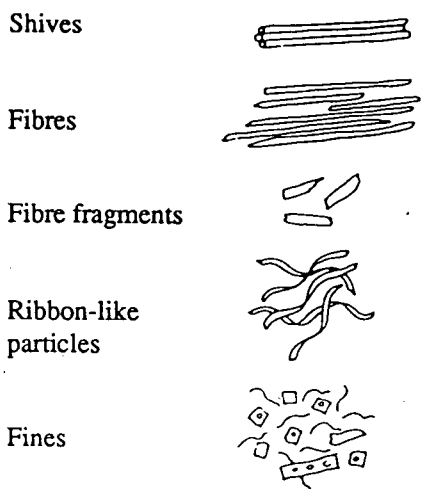


Figure 2.2. Schematic representation of the particles present in pure mechanical pulps (Mohlin 1982a).

middle fraction (50/200 mesh), and fine particles in the fine fraction (P200 mesh). The relative amounts and quality of the different particle types are the most important characteristics of mechanical pulps.

As an indication of the bonding ability of these particles, tensile index of handsheets of separate fractions has been used as a criterion. Each type of particle contributes to the properties and performance of a mechanical pulp in a non-additive way (Mohlin 1982a). The influence of the different particles on some important pulp properties is summarized in Figure 2.3.

In general, there are two fundamentally different approaches to mechanical pulp characterization . One is to imitate the papermaking process in the laboratory and, based on test

	Strength Properties (Tensile Index)	Optical Properties (Light scatt. Coefficient)	Surface Properties (Smooth-ness)	Runnability (Drainage)
Shives	-		-	
Fibres	+ Reinforcing bars	-	-	+
Fibre fragments	-			
Ribbons	+ Bonding	+	+	-
Fines	+ Bonding	+ +	+	-

Figure 2.3. Influence of different mechanical pulp particles on some important pulp properties. (Mohlin 1982b).

results obtained from handsheets, evaluate the pulp quality for a certain application. The other is to measure a few basic pulp properties and, from experience, translate these results into relevant information about the expected quality and suitability of the pulp tested for certain paper applications (Mohlin 1982a). A combination of these two approaches will probably render the most information about a pulp, particularly when assessing new raw materials or process alternatives for which experience might be insufficient to predict pulp quality.

Among the methods used to characterize mechanical pulps, the specific surface of a pulp or of its fractions has been used to determine the bonding potential, since it measures the extent of fibre surface development. Apparent density of handsheets, on the other hand, provides a useful measure of the ability of the wet-formed web to consolidate under an applied load. This property represents a complex index of fibre flexibility, degree of fibre packing and extent of interfibre bonding between the fibres after drying (Jackson and Williams 1979). These authors also used the swollen volume of fractions as a measure of the degree of dimensional swelling of the fibres as it related to fibre flexibility.

The fines, with a large specific surface as compared to the long fibre fraction, have an important role in producing paper with good strength, optical properties and printability. These particles could be divided into cellulose-containing fines with good bonding ability (slimy, swollen fibrils and lamellae) and a lignin-containing flour-like fraction consisting of non-swelling fines with poor bonding potential (Honkasalo *et al* 1983). It is difficult and laborious, however, to prepare handsheets from this material and the direct permeability method does not work for fines.

Pulp freeness is perhaps the most important test used to characterize mechanical pulps. Freeness measurement enjoys widespread use in spite of criticism of this test as an

indication of pulp quality. Freeness testing is done both to assess dewatering characteristics of the pulp and to estimate the strength of the paper to be made. However, for both purposes, the test is less than ideal (Gavelin 1982a). For a given mill, freeness can be a very useful indicator of pulp strength, but one cannot use freeness alone to calculate strength accurately. The test is best used in a restricted situation where the wood supply is uniform. Pulps produced by different processes at the same freeness can show very different properties (Fahey 1987).

Freeness is affected differently by the different pulp fractions. Fines, for instance, affect freeness much more than strength, whereas fibre length has comparatively little effect on freeness. Thus, a test value of freeness for one pulp may not mean the same for another, particularly if species or processing conditions are changed.

A full characterization of a mechanical pulp will probably never be complete without a microscopic study. Microscopy provides an opportunity to visually examine the whole pulp or each individual fraction, and to determine the surface quality of fibres, the extent of fibre cutting and unravelling, and the degree of fibrillation. It also provides an opportunity to apply counting techniques to determine frequency of various structural modifications on fibres, especially in the long-fibre fraction (Laamanen 1983). Furthermore, microscopy can be applied to pulp samples and to bonded paper sheets to visually assess their

papermaking potential. With regard to studies on cross sections of fibres, microscopy coupled with ultra violet illumination has been used to assess lignin distribution on fibres after different pulping or pretreatment alternatives (Wardrop *et al* 1961, Bruun and Lindroos 1983, Kerr and Goring 1976). Kibblewhite (1983) has used staining techniques on cross sections of softwood fibres to evaluate fibre damage and retention of middle lamellae.

2.4. Effect of Wood and Fibre Characteristics in Mechanical Pulping

The influence of wood specific gravity (SG) and morphology on the physical properties of pulps in general, has been the subject of numerous investigations. In general terms, SG depends upon (1) the diameter of the cells and (2) the thickness of the cell walls, and the relationship between the number of cells of various kinds in terms of (1) and (2) (Panshin and de Zeeuw 1980). The relationships obtained between wood characteristics and pulp properties have been derived mostly from chemical pulps, and often on a relatively small number of softwood species. On the other hand, the greater variability in morphology for hardwoods, both within and between species (particularly when tropical hardwoods are considered), makes it difficult to develop relationships between wood parameters and pulp quality, even for chemical pulps.

Specific gravity has probably been the wood property most

widely used to explain or predict pulp strength. Generally, it has been considered that woods of high SG, produce pulps with low bonding ability. Although SG may provide a general indication of pulpability for a certain raw material, it may not serve as a good predictor of pulp quality since it provides little clue as to the relative numbers, form, structure and distribution of the various cell types that make up the wood of different species (Scurfield 1976).

For mechanical pulp properties, there is no direct relationship *per se* between wood SG and pulp strength properties (Bueno 1978, De Montmorency 1965, Marton *et al* 1979). Relationships between wood SG and pulp bonding strength that appear to be clear for chemical pulps, may not apply to mechanical pulps. In fact, some woods resulting in strong mechanical pulps can yield chemical pulps ranking low in tensile and burst strength (De Montmorency 1965). For example, black spruce with a higher density than balsam fir produced mechanical pulps of much greater strength, but chemical pulps of lower strength, than did balsam fir. For hardwoods, Marton *et al* (1979) showed that white birch (SG=0.472) produced a much better refiner mechanical pulp in terms of tensile strength than did *Eucalyptus viminalis* Labill. (SG=0.465). Similarly, sugar maple (SG=0.607) gave stronger pulp than red oak (SG=0.548) even at higher yield values of the former. Thus, the statement by Giertz (1977) that pulp yield should be lower for higher density species to produce acceptable mechanical pulp strength, does not

universally apply.

Since a great deal of cutting and fragmentation takes place during mechanical pulping, initial fibre length of the wood material has not been considered an important limiting factor (Giertz 1977, Marton *et al* 1965). In fact, the middle and fines fraction have higher bonding ability than the long fibre fraction. For example, for a softwood TMP pulp, Jackson and Williams (1979) reported that the 100/200 fraction had 8 times the tensile strength of the R14 fraction, and twice that of the 48/100 fraction. However, long fibres are important to distribute stresses, and most strength properties correlate with the size of the long fibre fraction i.e., pulp strength properties depend on the added length of the fibres in the pulp, not on the length of individual fibres (Forgacs 1963, Mohlin 1982b).

Giertz (1981) indicated that a paramount task in mechanical pulping must be to remove the surface layers quantitatively without unnecessarily shortening the fibres. On the other hand, it is essential that the fines produced in mechanical pulping be of good quality, i.e. fibrillar fines capable of improving bonding and sheet consolidation, rather than chop. In fact, the bonding level of a pulp depends largely on the fine material (fines and ribbon-like particles) because it acts like bridges between the stiff fibres. These fibres, however, at the same time become more flexible as the fibre surface layers are removed, indicating that one cannot vary the fines content without also influencing the fibre

characteristics (Gavelin 1982a). It is clear that both fibres and fines are important for mechanical pulp quality. The quality of these fines will largely depend on the way in which the fibre loses its surface layers during refining. In general, the bonding ability of both fractions will improve as refining proceeds (Giertz 1977, Simmonds and Hyttinen 1964) since more of the cellulose-rich S_2 layer of the fibres is exposed, and the fines fraction will also have a higher proportion of fibrillar-type fines originating from the S_2 layer. Access to this layer of the fibre wall seems to be a very important consideration in mechanical pulping.

On the other hand, formation of ribbon-like particles is of significance for the quality of a mechanical pulp. Forgacs (1963) suggested that the success of mechanically pulping a particular species might be associated with the ability to promote unravelling of the fibres. He showed how development of initial splits or cracks on the S_2 layer turned into fibre unravelling and ribbon formation. Because of the large specific surface of these ribbons, particularly when fibrillated, they had increased bonding ability.

The prime limitation of raw materials for mechanical pulps appears to be the ability of their fibres to unravel and produce good quality fines. Thus, it seems that the internal fibre morphology, rather than specific gravity or fibre length, affects the strength and bonding characteristics of mechanical pulps. The internal fibre morphology refers not only to the relative thicknesses of the cell wall layers,

particularly the S_1 layer that may restrict access to the S_2 layer, but to their structure and chemical composition.

2.5. Mechanical Pulping of Hardwoods

In general, hardwoods have a more heterogeneous wood anatomy than softwoods. A greater number of cell types is found in hardwoods. Vessel elements are characteristic of this group of species. These cells are short, nonfibrous and are joined end-to-end in a vertical series to form tube-like structures (vessels) which are seen as pores on the wood cross section. The arrangement of pores in the wood can vary considerably between species, with both diffuse-porous and ring-porous species being represented in North America. Hardwoods have little or no radial alignment of the fibres or vessels, except for short radial chains of vessels in a few species (Parham 1983). The rays bend around the pores, and large vessel elements crowd other cells out of line. Also, in hardwoods, rays are more variable in width, with most species having multiseriate rays with or without uniseriate rays. Further, hardwoods usually contain more strand parenchyma with a large variety of arrangements.

Thus, it is not surprising that many hardwood species present difficulties in their conversion into mechanical pulps of acceptable quality. Consequently, the resulting pulp is often low in strength and not generally satisfactory for its inclusion in printing papers. For most species, the application of a chemical pretreatment is essential for the

successful refining of wood into fibres and fibre bundles without serious fibre damage (Marton *et al* 1979, Leask 1982). Although this is particularly true for higher density hardwoods, even low density, light coloured species may also require pretreatment (Atack and Heitner 1982). The improvement in strength and brightness with different combinations of chemicals is substantial. The use of a mixture of NaOH and Na_2SO_3 is common and effective as pretreatment for hardwood chips. While the NaOH addition is related mainly to pulp strength, Na_2SO_3 improves brightness (Higgins *et al* 1977). Although it has a negative effect on pulp brightness, alkali consumption increases swelling of the fibre walls with accompanying plasticization that enhances the ability of the fibres to conform and bond to one another during sheet making (Katz *et al* 1981). In many cases, hardwood mechanical pulps can be upgraded to a level where they can be substituted for softwood mechanical pulp, or partially replace chemical pulps in printing papers.

Hardwood fibres appear to have a more rigid structure than softwood tracheids, making the peeling off of the S_1 layer and fibrillation of the S_2 layer difficult, if not impossible (Marton *et al* 1979). It has also been found that the S_1 layer of many hardwoods is disproportionally thick, resulting in poor response to fibrillation upon refining. They suggested that the thicker the S_1 layer, the more difficult it would be to remove it. The thinner S_1 layer was associated with good fibrillation upon refining.

However, when chemical treatments are applied and the wood is softened resulting in more flexible fibres, the primary wall and S_1 layer are said to be peeled off in a manner similar to what was observed with spruce TMP fibres (Giertz 1977). For these softwood fibres, Giertz proposed that there was a possible weak bonding between the S_1 and S_2 layers and, upon refining, the primary wall and S_1 layer rolled back. At the same time the S_1 layer cracked and fibrillated to give access to the S_2 layer. By mixing fines from spruce TMP and birch high yield bisulphite pulps with spruce TMP fibres and measuring the strength properties of the resulting pulp sheets, Giertz concluded that the quality of the fines of hardwood mechanical pulps was inferior to that of softwoods. Although this conclusion may still hold, it should be mentioned that the fines involved in the experiment were removed from spruce and birch pulps which had different freeness values (57 ^{o}SR or about 125 mL CSF for spruce and 44 ^{o}SR or over 200 mL CSF for birch). This freeness difference may account for part of the difference in the fines quality reported. On the other hand, the fact that hardwoods contain abundant ray cells and vessel elements, which produce poor quality fines in terms of bonding (Marton *et al* 1979), may be the cause of this quality difference. A different conclusion was arrived at by Levina *et al* (1987). They reported an improvement in the pulp strength of cotton sulphite pulp when aspen CTMP fines (removed from a pulp of 26 ^{o}SR or about 400 mL CSF) were

added.

It is clear that more work needs to be done to understand the role of fines in hardwood mechanical pulps. For example, the quality of the fines from different mechanical pulping alternatives requires study.

Presumably, if the S_2 layer is exposed, the fibres will have more bonding potential because of their high percentage of carbohydrates compared to the compound middle lamella (CML), which is rich in lignin and relatively poor in cellulose and hemicelluloses. Cellulose, the main carbohydrate in wood, has at least 3 times the bonding potential of lignin (Rydholm 1965). Hardwoods have lower concentrations of lignin in the secondary wall compared to softwoods, and the effect of CML removal should be even more beneficial in terms of bonding than in the case of softwood fibres. In order to access the S_2 layer, the lignin-rich layers external to it have to be removed. In softwoods, the S_1 layer is removed and at the same time it fibrillates (Giertz 1977). In hardwoods, however, it seems that in order to remove the S_1 layer, alkaline treatment is required. For Eucalyptus species, it was reported that the S_1 and primary wall separated as a lignified morphological complex under cold soda pulping conditions (Wardrop *et al* 1961).

Sulphonation of lignin is a common practice in the pretreatment of chips for production of mechanical pulps from softwoods. It results in lower shive content, a larger long fibre fraction and more flexible fibers, thereby

indirectly increasing pulp strength. However, the extent of sulphonation of hardwood lignin is generally lower than that for conifers (Beatson *et al* 1985).

The basic chemical structure of lignin consists of phenylpropane units. The addition of one methoxyl group to the phenol ring produces a **guaiacyl** unit, while the addition of two methoxyl groups results in a **syringyl** unit. Almost all gymnosperms contain guaiacyl lignin, and all hardwoods have a guaiacyl-syringyl lignin, which is a copolymer of guaiacyl and syringyl residues (Panshin and de Zeeuw 1980). Evidence has been found that during chemimechanical sulfite pulping of aspen, the syringyl lignin units do not sulphonate. Thus, the distribution of sulphur followed the distribution of guaiacyl units (Beatson 1986). It was also found that the middle lamella cell corner region and the vessel wall were guaiacyl rich, whereas most of the syringyl units are located in the fibre wall. This statement also applies in the case of birch wood (Fergus and Goring 1970). On the other hand, Beatson *et al* (1985) reported that the degree of sulphonation of aspen lignin was only 50 per cent that of spruce lignin.

It is generally accepted that the low degree of sulphonation of the fibre wall lignin of hardwood chemimechanical pulps results in a lower strength gain relative to that usually observed for softwoods. It is actually the alkalinity of the treatment which is largely responsible for the increase in strength for hardwood mechanical pulps.

2.6. Tension Wood in Pulp

Tension wood (TW) is a form of reaction wood in angiosperms. It is thought to be formed as a mechanism for restoring leaning tree stems to their normal vertical orientation, or for maintenance of a preferred angular orientation of branches (Panshin and de Zeeuw 1980). Thus, it forms typically on the upper side of leaning trunks or branches, although it may occur in a diffuse arrangement in the stem cross-section, or be present with little evidence of stem eccentricity, particularly in species of *Populus*.

Tension wood is characterized by the presence of a singular type of fibre known as gelatinous fibre (or G-fibre). These G-fibres differ from normal wood fibres in that the inner portion of the cell wall consists of a unique layer, known as the gelatinous layer (or G-layer), which is composed of over 98% cellulose (Norberg and Meir 1966) and is loosely attached to the inner secondary wall. Thus, contrary to the reaction wood in softwoods, TW is low in lignin and pentosans compared to normal wood, but has usually higher density due to the presence of G-fibres. Despite it being labeled gelatinous, the G-layer contains no pectins. The cellulose in the G-layer is highly crystalline; the microfibril orientation is nearly parallel to the fibre axis and has a better developed microcapillary system than the cell walls of normal wood. It has also been shown that the G-layer has a lamellar structure with weak lateral bonds

(Cote and Day 1965, Norberg and Meier 1966). The thickness of this G-layer varies considerably depending on the species and degree of development of tension wood (Panshin and de Zeeuw 1980). Even within a single fibre, the G-layer is much thicker at the centre along the fibre length than toward the tips (Okumura *et al* 1977). The amount of TW is also variable depending on species and growth conditions. Isebrands and Parham (1974) reported up to 40 % G-fibres in short rotation *Populus*, and more than 70 % has been indicated for other species (Rao *et al* 1983).

The presence of G-fibres affects not only wood specific gravity, but also the size and number of other wood elements. Kaeiser and Boyce (1965) indicated that non-G-fibres of reduced diameter and increased cell wall thickness were present in TW, and that the diameter of VE decreased, although their frequency increased. Fibre length remained unchanged in TW compared to normal wood, although wood specific gravity had increased (Scaramuzzi and Vecchi 1968). High proportions of TW are undesirable for lumber and veneer products. Woolly surface appearance, buckling problems and dulling of the cutting tools are all caused by TW.

Chemical pulps prepared from TW have much lower strength than those from normal wood. This has been attributed to the lower hemicellulose content of TW fibres, thus affecting adversely the swelling and beating characteristics (Perem and Clermont 1961). More recently, weak sheets from TW have been related to the minimal conformability of the G-fibres

and their lack of collapse (Robinson 1977, Isebrands and Parham 1974), as shown by higher bulk values and cross sections of pulp handsheets.

Neutral sulphite semichemical (NSSC) pulping of aspen showed that pulps produced from TW required more beating to reach the same freeness level compared to those from normal wood. For the same beating times, however, TW pulps exhibited lower density, lower tensile strength and higher tear index (Clermont and Bender 1958). This suggests better preservation of fibre length in TW NSSC pulps compared to those from normal wood. However, the lower sheet density and higher tear index indicate a lower degree of collapse of the G-fibres.

Little information is available on the effect of tension wood on mechanical pulp properties. Dadswell *et al* (1957) found that the highest TW contents were associated with the highest pulp strength when refining *Eucalyptus nitens* and *E. regnans* chips under RMP conditions. The ease of fibre separation reported was attributed to the lower lignin content and a more "rubbery" nature of G-fibres. Work done on chip refining of *Populus deltoides* tension wood showed that reduction of freeness occured more quickly for tension wood than for normal wood. Handsheets prepared from TW RMP showed a significant increase in density and strength properties. The pulp presented individualized and long elements accompanied by abundant ribbon-shaped and fibrillar material (Scaramuzzi and Vecchi 1968). Evidence of exposure

of G-layers separated from the parent fibres was also presented for this pulp. Conversely, adjacent wood presented predominantly bundles of fibres and chippy material, and produced pulps of low strength.

It is interesting to note that pulping processes of a chemical nature had detrimental effects on TW pulp strength. On the other hand, pure mechanical pulps from TW are stronger than those from normal wood. The increased density of these pulps indicates that the stripping of G-layers out of the inside of the fibres might be the cause for stronger mechanical pulps from TW. Assessment of the breakdown pattern of G-fibres can provide fundamental details of the beneficial or detrimental effects of tension wood on pure mechanical pulping and other chemical modifications of this process. Nothing has been reported on the effect of TW in chemimechanical pulps.

2.7. The Role of Vessel Elements in Pulp

Vessels are tube-like structures of undetermined length, composed of individual cells called vessel elements (VE). These cells, present only in wood of deciduous trees, are specialized for conduction and are generally much larger in diameter, shorter in length and have thinner walls than the associated fibres (Haygreen and Bowyer 1982).

In the manufacture of printing paper products, VE can cause difficulty with respect to the surface quality of the paper since their shape is not conducive to development of strong

bonds between pulp elements.

The chemical composition of VE differs from that of the fibres. While the lignin in the secondary wall of fibres is rich in syringyl units, that of the VE is mostly of the guaiacyl type (Fergus and Goring 1970, Hardell *et al* 1980). The bonding ability of VE is generally lower than that of fibres. Handsheets formed from unrefined VE of kraft pulps gave strength properties inferior to those of the fibres (Marton and Agarwal 1965). However, when VE are present in quantities not exceeding those found in nature, they contribute to the strength of hardwood kraft pulps. The potential adverse influence of VE in a paper sheet therefore arises not from a reduction of bonding between elements, as measured by conventional pulp strength tests, but because of their presence in the paper surface. Vessel elements present on the surface of a printing paper sheet may reduce its surface resistance. This can cause problems during the printing process.

During offset printing, vessel picking or lift-off can occur, causing the deposition of these elements on the printing surface. This will reduce the quality of printing and can lead to periodic shutdowns of the press to clean the printing blankets (Smook 1982).

Vessel picking has been studied for a number of hardwood species, including tropical woods, but always for chemical pulps. From experiments carried out by Colley (Colley 1973, Colley 1975 and Colley and Ward 1976), it was found that

both fibre and vessel element morphology affected vessel picking. For a given species, a reduction of vessel/fibre ratio, an increase in beating time, and the reduction of vessel element size, all caused a reduction of vessel picking tendency. Nanko *et al* (1988) reported that high consistency refining could reduce vessel element size. However, this was only effective before a freeness level of 400 mL CSF was reached, beyond which further refining gave only small changes in size and numbers of VE, regardless of the method of refining. Mukoyoshi *et al* (1986) found that a bleached pulp prepared by kraft cooking of chemimechanically prefiberized chips had lower vessel picking tendency than conventionally bleached kraft pulp. This indicates that there might be a reduction of VE size upon chip refining.

Several other means of suppresing vessel picking have also been reported. Perhaps the most common one is the application of proper surface sizing (McGovern 1977, Colley and Ward 1976). Also, partial removal of large VE from the pulp using hydrocyclones (Ohsawa 1987) was indicated as useful, although it was not as effective as using stratified sheet formation techniques to cover the VE in the paper surface with a very thin layer of fibres (Nanko *et al* 1987). Although vessel picking has not been reported for mechanical pulps, it is a potential problem, particularly if the use of hardwood mechanical pulps increases for the production of printing papers. Marton *et al* (1979) noted the existence of whole VE in TMP pulps, and even more so in CTMP pulps. Since

these mechanical pulp fibres do not have the bonding potential of kraft pulps, and the vessel elements are possibly more rigid and less conformable in mechanical pulps, vessel picking is likely to occur. It is anticipated, however, that mechanical defibration will cause considerable reduction of vessel element size. The effect of different mechanical pulping processes on the breakdown pattern of vessel elements has not been reported in the literature so far. However, Marton *et al* (1979) estimated that about one-half of the whole vessel elements present in the wood were disintegrated into fines during TMP pulping. Under alkaline sulphite CTMP refining conditions, the number of whole VE per gram of pulp almost doubled for white birch, but surprisingly decreased for Eucalyptus. The number of VE fragments was also counted, but the minimum particle size was not specified. One would expect, however, that very small particles would be increasingly difficult to identify. Contrary to these findings, Giertz (1977) indicated that VE were reduced to fine material which mostly ended up in the 100/200 fraction of a Bauer McNett Classifier.

The question of the way in which VE breakdown occurs in chip and pulp refining has not been solved. Estimation of the survival of whole VE as well as particle size distribution upon refining would provide basic information of the pattern of breakdown of VE. Of specific interest is the study of VE breakdown under conditions of wood softening and refining.

III. METHODOLOGY

This chapter describes the methods followed to characterize the wood species used for pulping, to produce and evaluate the pulps, and the microscopic techniques involved in the analysis of fibres and vessel element breakdown during mechanical pulping. A general outline of the experimental procedures used is presented in Figure 3.1.

Specifically, the effects of three mechanical pulping processes (TMP, CTMP and CMP) were studied on the defibration characteristics of aspen and birch woods. Four pulps were produced by each of these processes for each species, over a broad range of refining energy consumption levels. A total of 24 pulps was analysed for the two species selected. Pulp identification codes and properties are presented in Table 4.9.

3.1. Wood Procurement

Two hardwood species were selected for this study because of their importance to Canada: aspen (*Populus tremuloides* Michx.) and white birch (*Betula papyrifera* Marsh.). One tree of each species was felled at the Alex Fraser Forest at Williams Lake, B.C. Both trees were 110 years of age and had a diameter of 22 cm at breast height (DBH). The trees were cut into logs of about 1.20 m in length and debarked by hand. Any visible decay was removed from the logs by splitting them in 4 quarters along the grain and cutting out

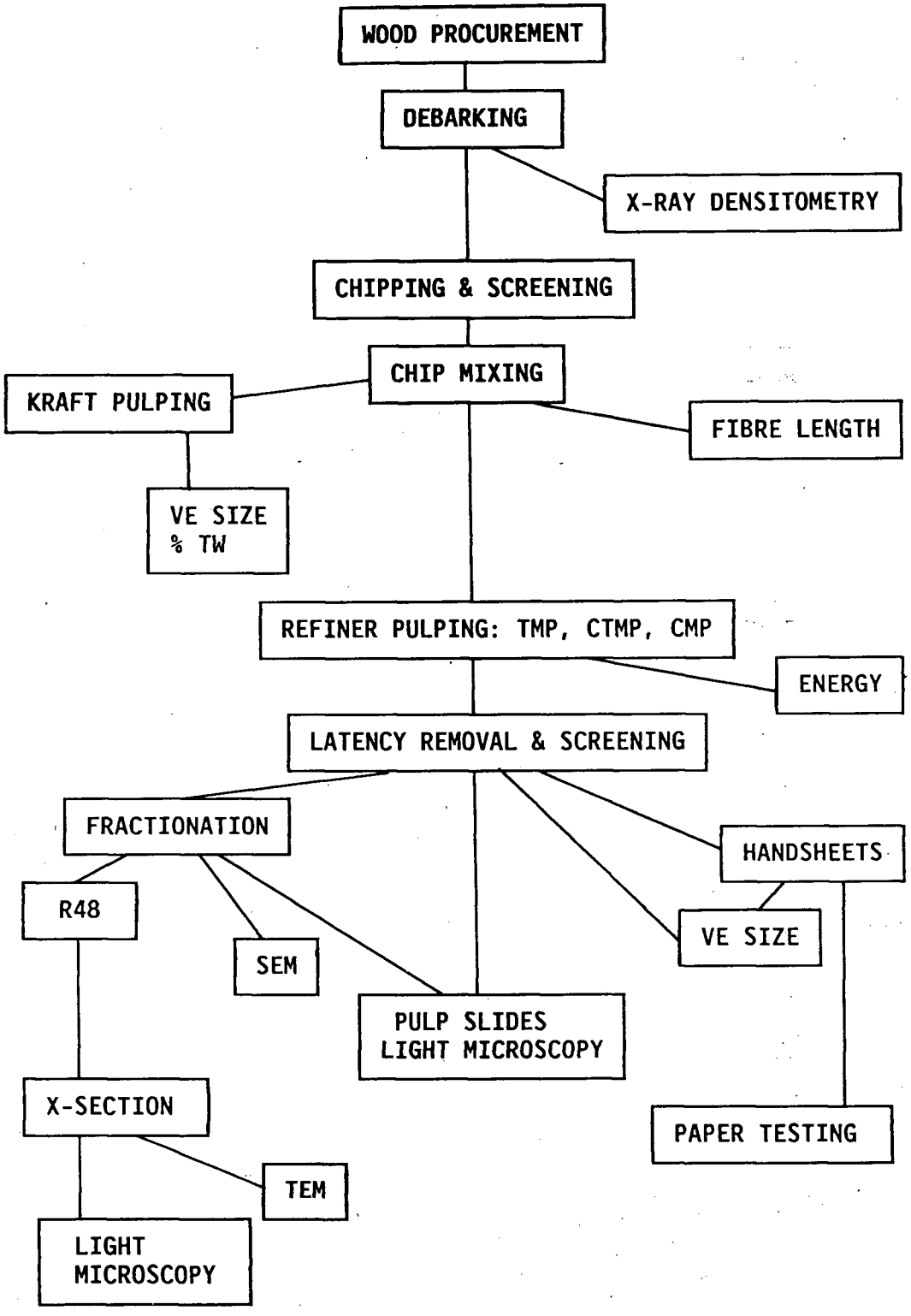


Figure 3.1. General Outline of Experimental Procedures.
VE:vessel elements; TW:tension wood fibres;
SEM:scanning electron microscope;
TEM:transmission electron microscope.

the affected areas with a band-saw before chipping. Since a considerable portion of the aspen tree was affected, and therefore removed, a second aspen tree (80 years of age, DBH=30 cm) was taken from an area near Lytton, B.C., in order to ensure a sufficient amount of material to carry out this investigation.

The trees were characterized in terms of wood density by means of direct reading X-ray densitometry as described by Jozsa and Myronuk (1986). Wood disks 1 in (25 mm) thick at DBH, and at heights of 20, 40, 60 and 80% up the tree stem length were cut and processed. The extracted thin cross sections analysed by densitometry provided wood density values that corresponded to basic relative density (oven dried weight per green volume) according to the calibration procedure developed by Jozsa *et al* (1987).

3.2. Production and Characterization of Chips

Only sound wood pieces were used in the production of chips. Some of the logs had to be split further along the grain to comply with a maximum of 9x9 in (23x23 cm) cross-section required by the shape and size of the chipper spout. The wood pieces of each species were chipped in a 60 H.P., six-knife disk chipper. The chips were collected in plastic bags and stored in a freezer at about -7 ^{o}C before further use. In order to remove oversized material and fines, the chips were then screened in a continuous Burnaby Mill and Machinery Equipment Ltd pilot scale chip screen with two

decks: an upper deck with 1.25 in (32 mm) holes and a lower deck with holes of 0.25 in (6 mm). The solids contents of the aspen and birch chips were 61.1% and 59.4%, respectively.

Accept aspen chips from the two trees cut were thoroughly mixed. This was done by forming a pile of chips and shovelling them into a doughnut-shape arrangement. The chips were then shovelled back into the centre to form a new pile. This operation was repeated 5 times.

The proportion of chips from Williams Lake and Lytton in the mix was approximately 0.625:1 . Birch accept chips were also mixed together to provide a uniform chip furnish. A representative sample of accept chips from each species was characterized by thickness classification (Table 3.1) using a Wennberg Chip Classifier.

TABLE 3.1. Accept chip thickness classification

	SPECIES	
THICKNESS	ASPEN	BIRCH
< 2 mm	0.8%	2.8%
2-6 mm	94.1%	95.5%
> 6 mm	5.2%	1.7%

These chips were used in the manufacture of thermomechanical pulps (TMP) and chemithermomechanical pulps (CTMP).

For the manufacture of chemimechanical pulps (CMP), a portion of these chips was screened by thickness and only the 2-6 mm thick accepts were used for pulping. Kraft pulping trials were also done using this material.

Fibre length was measured from the chips produced for the two species. Matchstick-like pieces were cut from randomly selected chips, macerated in a mixture of hydrogen peroxide and acetic acid (50/50) and gently boiled until fibre separation occurred, as described in CPPA Standard B.2P. Fibre length was then measured by projecting slides of fibres stained with toluidine blue, and tracing these with a probe of an NEA Fibre Length Counter. Each length measured was automatically entered into a corresponding class interval. These intervals were 80 *um* apart.

3.3. Preparation of Pulps

This study is based on twenty-four hardwood refiner pulps selected from several pulp runs. Twelve pulps were prepared for each species. The processes by which these pulps were produced encompassed TMP, CTMP and CMP conditions and each covered four points over a wide range of refining energy levels and pulp freeness, as shown in Figure 3.2. The pulps produced are given an identification code in Table 4.9, and their corresponding pulp properties are presented in Tables 4.9 through 4.11.

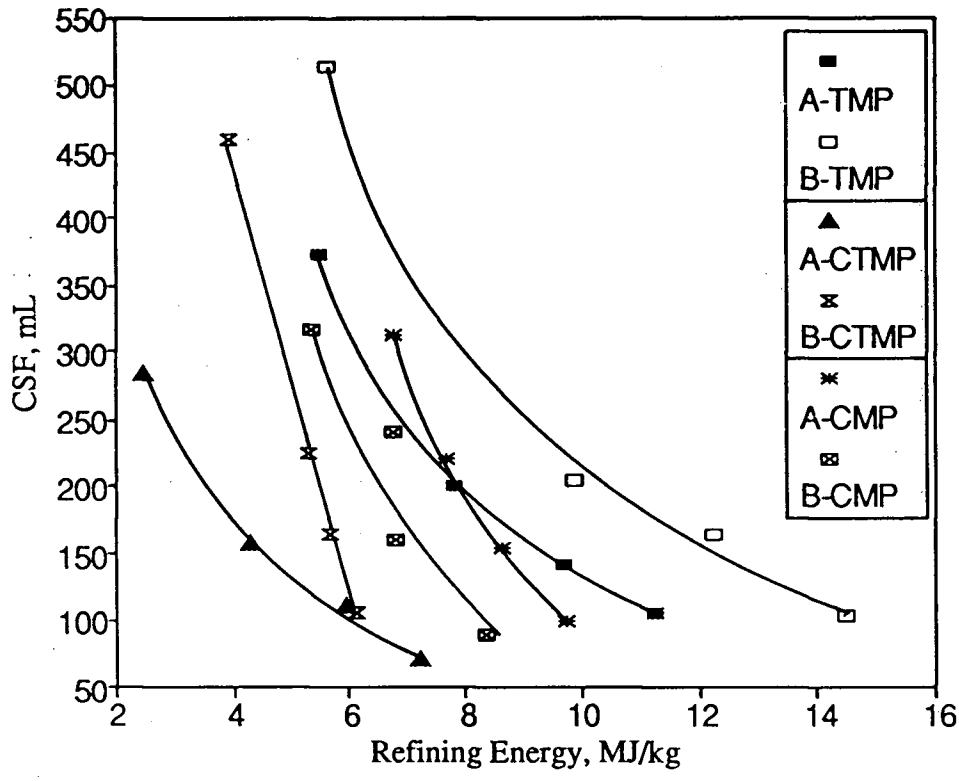


Figure 3.2. Relationship between specific energy consumption and unscreened freeness for the hardwood refiner pulps produced.

Before any pulping took place, the frozen chips were thawed to room temperature.

The refining energy consumption was calculated from charts recording a watt meter output attached to the refiner.

The pulps produced were tested for unscreened freeness after hot disintegration as described in Section 5.4. The pulps were stored in sealed plastic bags at about -7 ^{o}C if not processed immediately.

3.3.1. Production of Thermomechanical Pulps (TMP)

In the primary refiner pulping, a Sunds Defibrator TMP 300 pressurized laboratory refiner (300 mm diameter) was used for the production of TMP from aspen and birch, under the following conditions:

- Atmospheric Presteaming: 15 min
- Preheating: Temperature: 125 °C
  Pressure: 16 psi (110 kPa)
  Time: 5 min
- Refiner Housing: Temperature: 128 °C
  Pressure: 18 psi (124 kPa)
- Prex Impregnator Compression Ratio: 1:3
- Refiner Plate Pattern: Rotor R3809
  Stator S3804
- Refining consistency: approx. 20%

These conditions were similar to those reported in the literature for the production of hardwood TMP (Jackson 1982, Marton *et al* 1979, Law *et al* 1985, Koran 1988).

The gap between the plates was decreased stepwise during a refining run to increase the refining energy. In this manner, a number of pulps with different freeness values was collected at the outlet of the refiner system. However, only one pulp was selected from each species for secondary refining.

The pulp obtained from the primary system was further refined in a 12 in (300 mm) diameter Sprout-Waldron

laboratory refiner with atmospheric discharge. To obtain different levels of refining energy, the number of passes was increased and/or the gap between the refiner plates was decreased. The refining consistency was kept between 15 to 20 % and the refining temperature was controlled at 85°C. Refiner plates with a designated D2A507 pattern were used.
In an effort to measure pulp yield, all the pulp material was collected during the refining process. However, because of the large amount of pulp produced in any single trial and the difference in consistency within this material, the values obtained were not considered to be accurate.

3.3.2. Production of Chemithermomechanical Pulps (CTMP)

The same primary system which was used for TMP production was also used for the CTMP runs. The same conditions of pre-steaming, compression ratio, pre-heating, refiner housing and refiner plate pattern were maintained, as before.
The chemical pre-treatment of the chips was carried out as follows. After pre-steaming, the compressed chips were allowed to expand in a solution of sodium hydroxide and sodium sulfite of known concentration within the pre-heating vessel. The concentration of these reagents was adjusted to provide a liquor consumption by the chips of 2% Na_2SO_3 and 3% NaOH based on the amount of oven-dried chips. The liquor pH was 13.6 for both species.
The selected pre-treatment represented typical conditions used to optimize hardwood CTMP quality (Higgins *et al* 1978,

Sinkey and Charters 1977, Jackson 1982, 1987, Law *et al* 1985, Valade and Law 1988).

For aspen, it was possible to obtain low levels of pulp freeness by decreasing the plate gap. This allowed pulps to be made within the desired range of 100-300 mL CSF freeness. In the case of birch, energy input could not be increased beyond a certain value, without causing excessive darkening of the pulp. Thus, only pulps at high freeness levels (above 400 mL CSF) were produced, and additional passes through the atmospheric discharge refiner were required in order to obtain birch pulps of lower freeness. The conditions used for the secondary refining of birch CTMP were similar to those for TMP. As in the case of TMP, yield determination was also attempted for CTMP.

3.3.3. Production of Chemimechanical Pulps (CMP)

A more severe pre-treatment of the wood chips was carried out to assess differences in fibre development upon refining. For the production of pulp with this process, the chips were pre-treated under the following conditions:

- Atmospheric Pre-steaming: 15 min
- Chemical charge: 2% Na_2SO_3 based on oven-dried chips
  5% NaOH based on oven-dried chips
- Initial pH of liquor: 13.0
- Liquor to Wood Ratio: 5:1
- Maximum Temperature: 135 ^{o}C
- Time to Maximum Temperature: 50 min

- Time at Maximum Temperature: 30 min

These conditions are more severe than those generally reported in the literature, particularly in terms of the time and temperature of pre-cooking (Higgins *et al* 1977, 1978, Sinkey and Charters 1977, Allan *et al* 1968, Marton *et al* 1979, Valade and Law 1988, Leask 1968). More drastic conditions were used here to see if the pre-treatment could cause complete removal of the compound middle lamella and S_1 layers from CMP fibres, and if the cell wall was weakened enough to produce fibres that failed radially.

A known weight of chips was placed in a basket inside the digester for yield evaluation. After the cook, these chips were soaked in water for 5 days with frequent water changes. Washing was complete when no further change in the color of the water was observed.

Immediately after the cook, the treated chips were refined in the Sprout Waldron atmospheric discharge refiner. Four batches from several pulps, produced from each species by adjusting energy input levels, were selected for evaluation.

3.3.4. Production of Kraft Pulps

Initially, conventional kraft pulps were produced to assess the presence of tension wood (TW) fibres by analysis of fibre cross sections. The presence of tension wood fibres is illustrated in Figure 3.3. The proportion of TW fibres was measured from kraft fibre cross sections, as shown in Figure

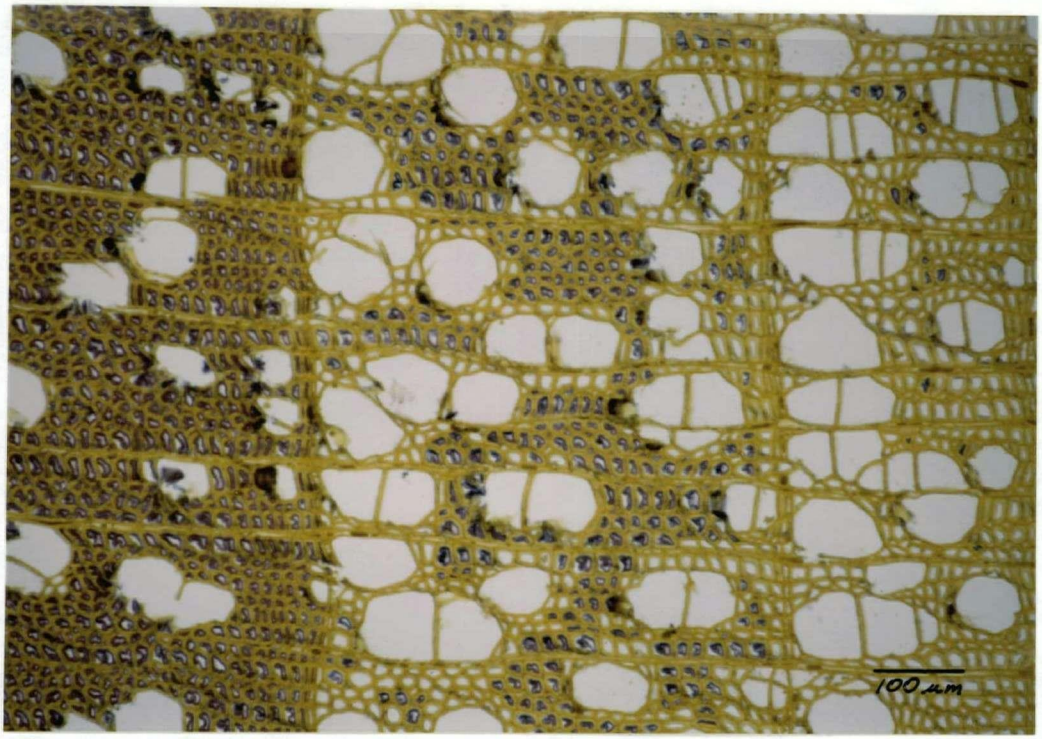


Figure 3.3. Cross section of aspen tension wood. A Herzberg reagent stained purple the G-layers, while the lignified material turned yellow.

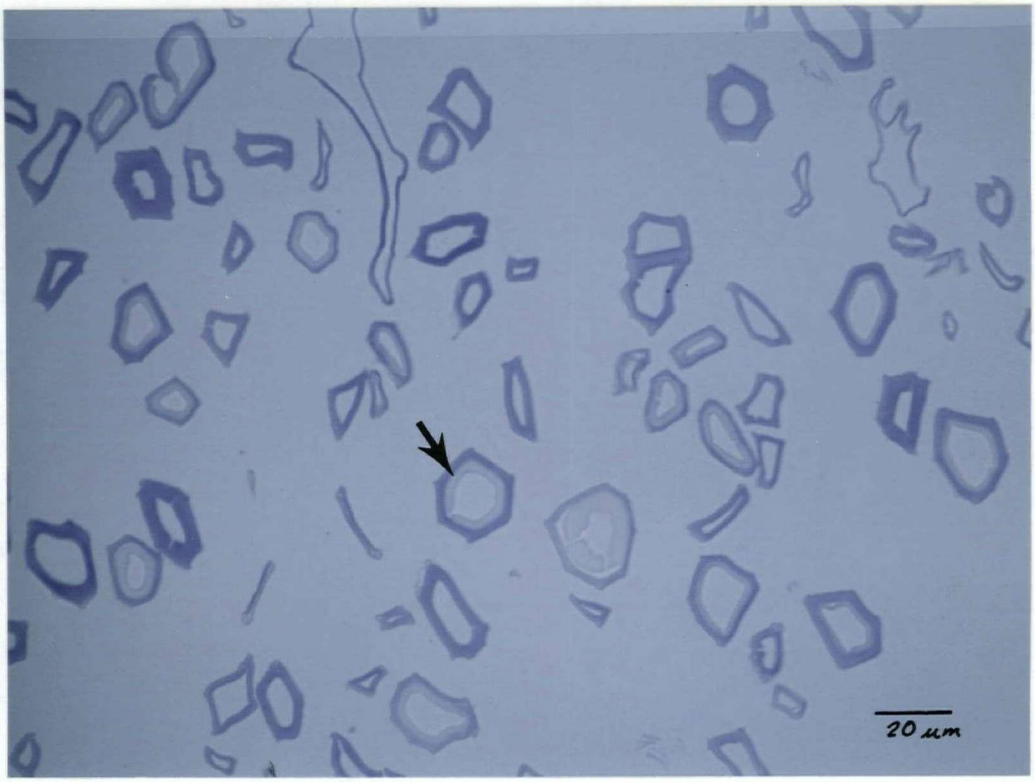


Figure 3.4. Aspen kraft pulp fibres stained with Toluidine Blue O, showing G-fibres in cross section.

3.4.

Kraft pulps were also used to gain information on the separation of cell wall layers following pulping for the two species under study. Further, these pulps were also used in measuring the original size of vessel elements for determining the possible vessel element breakdown during mechanical pulping.

Representative samples of aspen and birch chip furnishes were pulped to Kappa Numbers of 15.8 and 15.5, respectively (approximate yields of 56 and 52%), for which H factors of 955 and 1110 were required. The pulps were then washed and screened through an 0.008 in (0.2 mm) slot flat screen before further processing.

3.4. Pulp Processing

Latency was removed by hot disintegration before the refiner pulps produced were evaluated. It is important to remove latency prior to the characterization of mechanical pulps. Latency is a stress condition within the fibre wall and is largely caused by mechanical pulping. In latency removal, the lignin-hemicellulose matrix of the mechanical pulp fibre softens and the cellulose is released from its stressed condition. This results in a pulp showing lower freeness and higher strength values than the untreated fibre (Fahey 1987). Latency removal was done by disintegrating the pulps with boiling water for 5 minutes (1500 revolutions) at a consistency of 1.2% in a British Standard Disintegrator. The

pulp was then further diluted with cold water and screened according to PPRIC Standard Testing Procedure PS-3 (Method B) in a flat screen with slots 0.006 in (0.15 mm) wide. Fines were recirculated and eventually collected with the rest of the pulp.

3.5. Pulp Testing

The following tests were performed on the wet mechanical pulps:

- Canadian Standard Freeness: CPPA Standard C.1
- Shive Content: Pulmac Shive Analyser
- Bauer-McNett Classification (28, 48, 100, 150 and 200 mesh screens): TAPPI Standard 233 cm-82

Pulp handsheets were prepared according to CPPA Standard C.4, with fines recirculation. The fines material is a very important part of mechanical pulps in terms of bonding, light scattering ability and surface quality of the sheet (Mohlin 1982b). Five handsheets were discarded before a test specimen was prepared in order to reach a constant proportion of fine material retention for all subsequent handsheets. The prepared handsheets were conditioned according to CPPA Standard A.4 before the following tests were performed:

- Basis Weight: CPPA Standard D.12
- Bulk: CPPA Standard D.5H
- Density: CPPA Standard D.5H

- Brightness: CPPA Standard E.2
- Light Scattering: CPPA Standard E.2
- Opacity: CPPA Standard E.1
- Tear Strength: CPPA Standard D.9
- Burst Strength: CPPA Standard D.8
- Tensile Strength: CPPA Standard D34.P
- Tensile Energy Absorption: CPPA Standard D.34P
- Stretch: CPPA Standard D.34P
- Porosity: CPPA Useful Method 524
- Smoothness: CPPA Standard D.29P

3.6. Microscopy

A repeat set of Bauer-McNett fractionations was performed on the pulps so that the fractions could be collected. For the collection of the P200 fraction, a sample of about 15 L of suspension passing the 200 mesh screen was collected between the third and sixth minute after the test had started. The pulp material was then dewatered by filtration through a cloth, but not allowed to dry.

Light microscopy was carried out in a Nikon Microphot-FX Universal Microscope, while scanning electron microscopy was done using a JEOL JSM-840A scanning microscope. A ZEISS High resolution electron microscope EM-10C was used for transmission electron microscopy.

3.6.1. Pulp Slides

A portion of each fraction and each whole pulp was sampled

to prepare microscopic slides. A suspension of the material was uniformly spread and dried onto a warm slide. The pulp material was subsequently stained with Toluidine Blue O, a dye which has been extensively used in biological studies (Clark 1981, Green 1978). Permount was applied as mounting medium before affixing the cover slip. A few slides were left unstained so that they could be used with a solution of an appropriate refractive index for observation under phase contrast.

Because of the presence of tension wood fibres in aspen, other reagents were also used to distinguish the highly cellulosic G-layer from the rest of the fibre wall. These included Basic Green (TAPPI Routine Control Method RC-221), which stained the G-layer yellow and the lignified fibre wall green in chemically treated fibres, and the Herzberg Stain (CPPA Standard B.3P) which produced a purple color on the G-layer and gold for the lignified material.

3.6.2. Study of Fibre Cross Sections

Fibre cross-sectional analysis is a technique that allows the study of different features of pulp fibres based on their cross sections. Of particular interest in this study were the fibres produced by mechanical pulping processes; their examination included the assessment of fibre surface quality, and cell wall damage. The Bauer-McNett R48 fraction (fibres retained on the 48 mesh screen) was prepared for analysis of fibre cross sections. The R28 fraction was

considered too small to provide information over the entire pulp. On the other hand, fractions passing the 48 mesh screen were considered to be largely attrition products of the long fibre fraction. The R48 fraction always accounted for a large portion of a screened pulp, as shown in Figure 3.5. Twenty-four mechanical pulps were studied and, for each pulp, two samples, consisting of 150 fibres each, were analyzed.

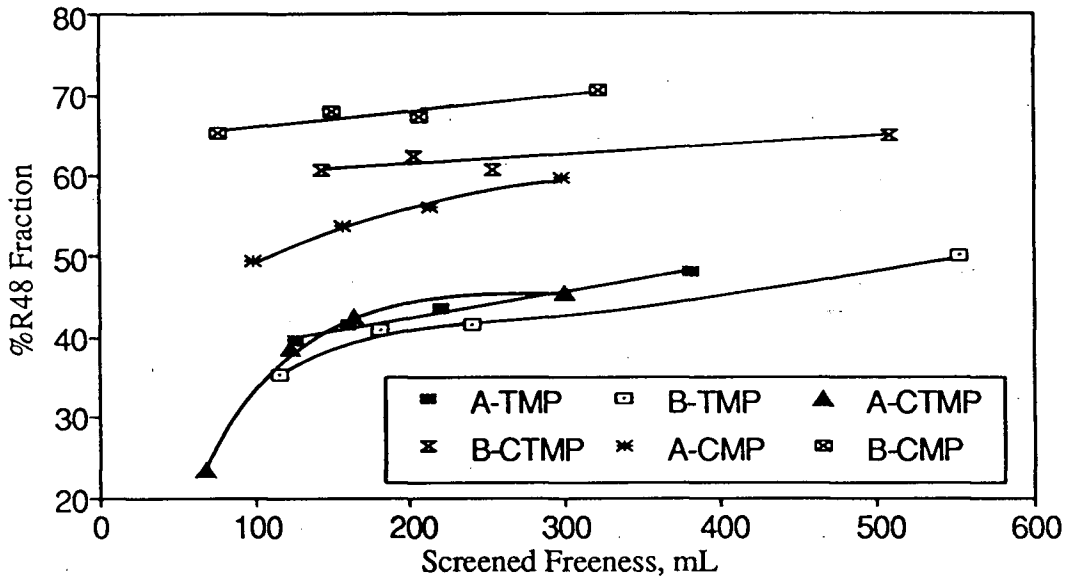


Figure 3.5. Relationship between freeness and the percent of fibres retained on a 48 mesh screen.

3.6.2.1. Preparation of fibre cross sections

The following steps were completed in the preparation of fibre cross sections:

a) Standard Bauer-McNett tests were performed on the 24 pulps selected; the R48 fractions were collected and not allowed to dry.

b) The samples were placed in a continuous solvent exchange apparatus. Solvent exchange was done for about one hr until the fibres were gradually embedded in 100% tert-butanol, following the method by Goldfarb *et al* (1977).

c) The fibres suspended in butanol were placed in a special flask and this, in turn, introduced into methanol at about -45 ^{o}C in a Neslab Cryobath CB-60 apparatus. By slowly rotating the flask, the material froze.

d) The fibres were dried under vacuum at a temperature of -55 ^{o}C in an Edwards Freeze Dryer Super Modulyo apparatus.

e) The dry, fluffy fibre material was kept under vacuum in a desiccator in small labelled vials to avoid hydration.

f) A portion of these fibres was placed over a glass slide with a few drops of Spurr embedding medium previously prepared. Spurr resin was chosen for its low viscosity and clear color. The standard Spurr preparation formula was used according to a J.B.EM Services Product Note (Anon. 1988b). To ensure good penetration of the resin into the fibres, the fibres were subjected to several

cycles of gentle vacuum to remove air in the fibres.

g) The impregnated fibres were then carefully aligned with fine tweezers into fibre bundles of at least 200 fibres. Alignment was done for a few fibres at a time until a bundle of enough material was produced. Each bundle was then picked up and rolled gently between fingers. Plastic gloves were used to prevent skin contact with the resin.

h) With fine tweezers, the roll of fibres was placed into the tip of a silicone capsule. Hemi-hyperboloid tip JBS #301A BEEM embedding capsules were used. The capsules contained a drop of Spurr resin and had to be previously evacuated to remove the air bubble usually trapped in the tip. When the fibre roll was inside the tip of the capsule, it was filled with resin and subjected to several cycles of gentle vacuum until no bubbles were observed.

i) Finally, the capsules were labelled, closed and placed in a constant temperature oven (TAAB Embedding Oven MK-11) maintained at 70 ^{o}C. The resin was cured for at least 16 h.

j) Six capsules were prepared for each pulp sample. However, only two of them were selected for the actual analysis of fibre cross sections. The blocks, after stripping the silicone capsules, containing the best fibre bundle alignment were selected after observation under a stereo microscope.

k) The cured blocks were trimmed down to just above the middle of the fibre roll length. This was done with a Dremel Moto-tool 395 with variable speed under a stereo microscope. A subsequent cut with a sharp razor blade provided the final block surface before sectioning with glass knives.

l) A LKB 2178 Knifemaker II was used to prepare glass knives following the instructions in the corresponding manual.

m) Using freshly prepared glass knives, the blocks were cut in a microtome into sections of 1.5 um in thickness. Either a Sorval Porter-Blum Ultramicrotome MT-2 or a Reichert-Jung 2050 Supercut were used for cutting at very slow speeds.

n) The sections were picked up and placed in a watch glass containing the staining solution. A JBS #197 tissue stain (Anon. 1988a) was selected for staining after trying several other options. It contained a mix of Toluidine Blue O and Basic Fuchsin. Toluidine Blue O is known to selectively stain lignin and to give good contrast between middle lamella and secondary wall in a single fibre (O'Brien *et al* 1964, Green 1978, Clark 1981). However, optimum staining contrast differed from softwoods in which pH is kept at around 10 (Williams 1988). On the other hand, Basic Fushin was found to stain the G-layer of tension wood fibres with a pinkish color since it has affinity for cellulose (Clark 1981).

The commercial staining solution was diluted 10 times before it was used to stain the sections for an approximate time of five minutes. The pH of the staining solution was neutral.

o) The stained sections were then picked up with a small loop, immersed briefly in distilled water for washing and placed on a block of Teflon to prevent adhesion, while removing the rest of the dye with the corner of an absorbent paper.

p) Once the sections were dry, they were carefully picked up with fine tweezers and placed on a glass slide on top of a drop of Polybed embedding medium (Anon. 1987) previously prepared. Polybed was used here as a mounting medium because it does not cause reticulation with Spurr resin and does not bleach the stain out of the stained fibres (Williams 1988). Before placing the cover slip, the sections were totally submerged in Polybed to avoid formation of air bubbles.

q) The slides prepared in this way were clamped overnight to ensure section flatness. Every slide contained several sections, but only one was selected for the analysis of fibre cross sections after inspection under the light microscope. The sections chosen provided the best combination of stain contrast and flatness.

3.6.2.2. Definition of Categories Studied

With the objectives in mind and after careful observation of

several sections, it was decided to confine the analysis of fibre cross sections to the following categories:

a) Retention of Compound Middle Lamella,

b) Retention of S_1 layer,

c) Exposure of S_2 layer,

d) Mode of separation of outer layer,

e) Radial failure,

f) Delamination of the S_2 layer,

g) Breakdown of tension wood fibres, and

h) Proportion of distorted fibres.

Only completely separated fibres were considered for every one of these categories, i.e., no fibres within a fibre bundle or shive were analysed. Also, since it was not possible to cut all the fibres at the exact centre of their length, no restriction was placed on minimum fibre diameter, except that the lumen be visible. A sample chart used for recording the above-mentioned categories is presented in Appendix B. The abbreviations used to identify fibre surface categories are listed in Appendix F.

Figures 3.6 to 3.10 show examples of the fibre cross sectional features recorded. Data was obtained for each category by counting the number of fibres that presented the feature of interest. The categories were established and defined as follows:

a) Retention of Compound Middle Lamella (MLr):

Fibres in cross section could be classified as having or

lacking a middle lamella around it. This is defined by the sharp contrast between compound middle lamella and secondary wall due to their difference in lignin concentration. Toluidine Blue O has great affinity for lignin and does not stain polysaccharides such as cellulose (O'Brien *et al* 1964, Green 1978).

Because of the difficulty of distinguishing the true middle lamella from the primary wall, the term middle lamella (ML) is considered here to include the primary wall. Thus ML was treated as the compound middle lamella, which actually includes the primary wall. Within this category, several subdivisions were considered:

- No retention of middle lamella {ML(r=0)}: when no light absorption was evident around the fibre cross section,i.e., zero retention.
- Less than 50% Retention {ML(r<50)}: when there was some retention of the ML but it was less than 50% of the perimeter of the fibre cross section.
- More than 50% Retention {ML(r>50)}: when the retention was not total, but was greater than 50% of the perimeter of the fibre cross section.
- Total Retention {ML(r=100)}: when the ML was present along the entire perimeter of the fibre cross section, i.e., 100% retention.

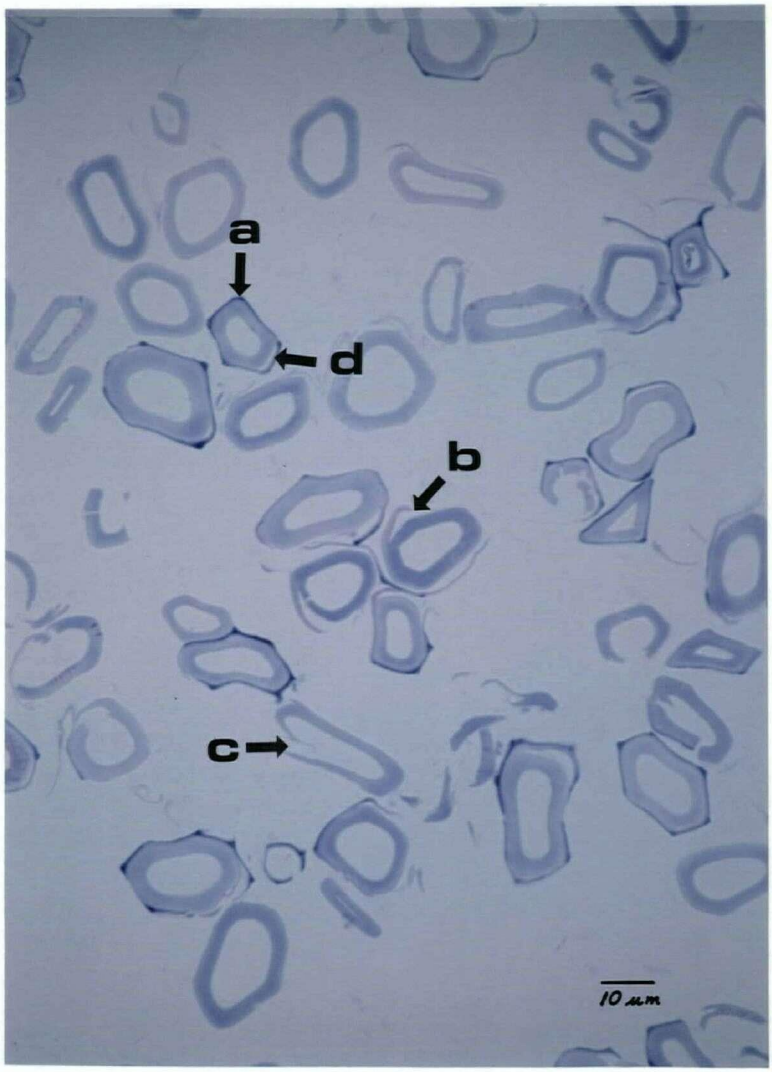


Figure 3.6. Hardwood refiner pulp fibres in cross section showing: (a) ML retained, (b) delamination of the S_2 layer, (c) radial failure, and (d) separation of the outer cell wall or "out/in" effect.

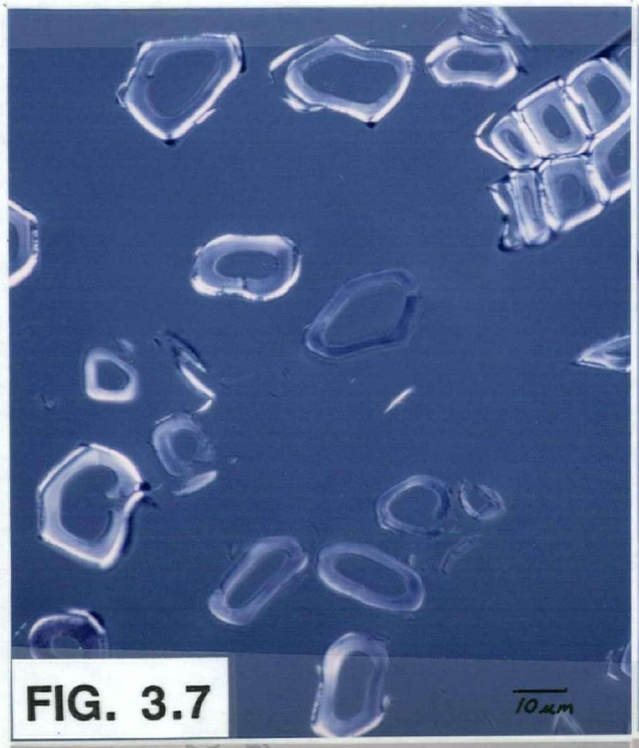


FIG. 3.7

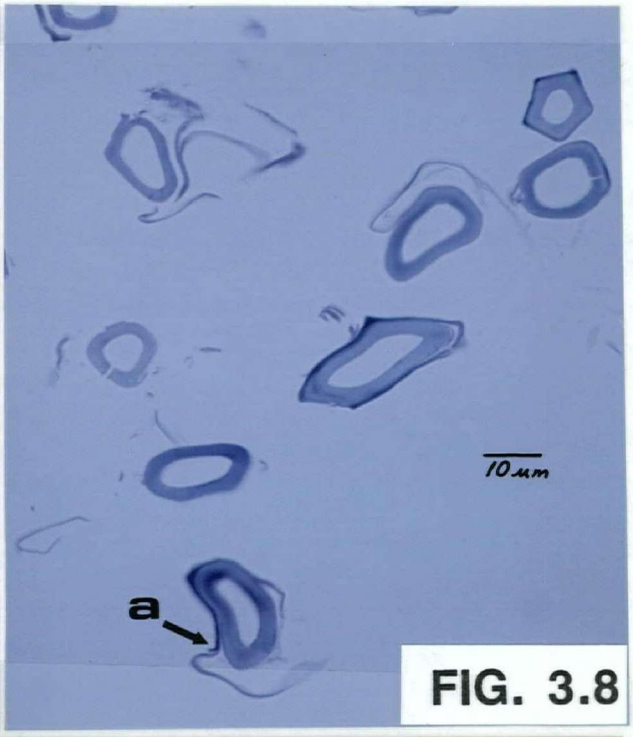


FIG. 3.8

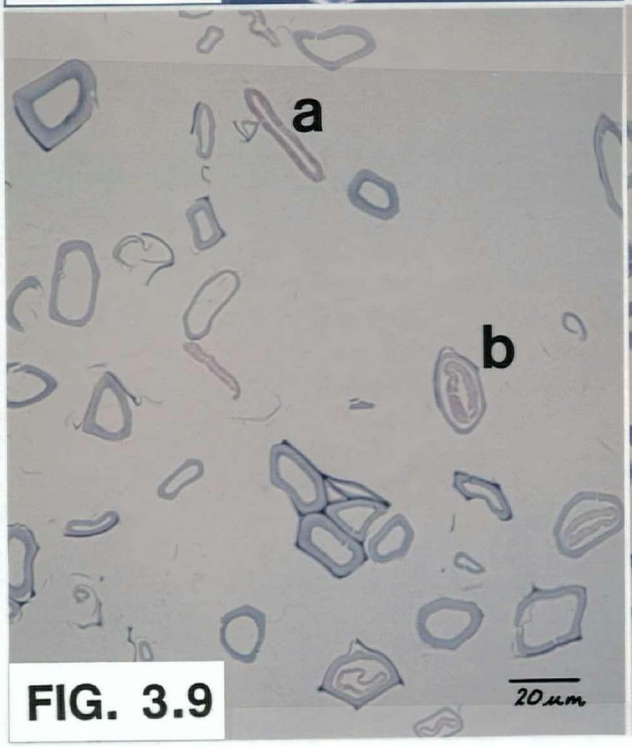


FIG. 3.9

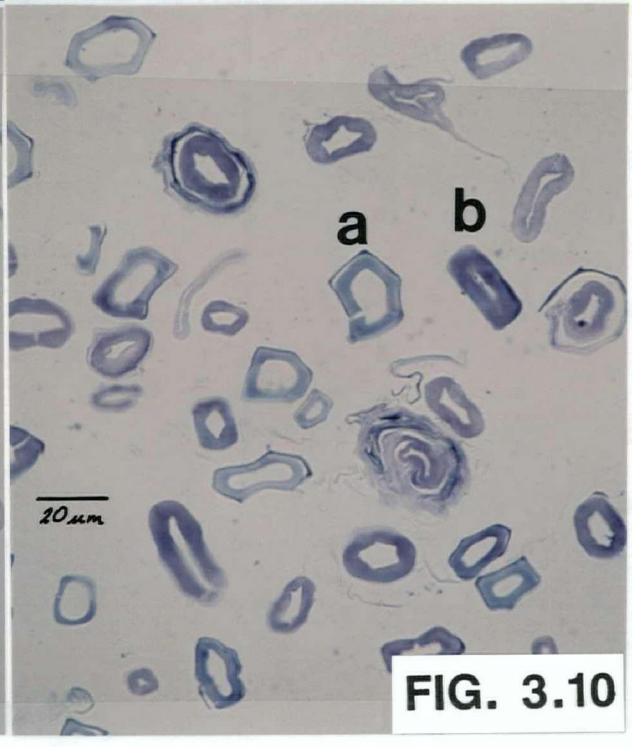


FIG. 3.10

Figure 3.7. Retention of S_1 layers shown as bright lines around fibres in cross section under polarized light.

Figure 3.8. (a) Peeling of the ML.

Figure 3.9. Aspen pulp fibres showing (a) G-layer stripped from the lignified cell wall, (b) G-layer inside the fibre.

Figure 3.10. Pulp produced from chemically-treated chips showing (a) Non-distorted fibre, and (b) distorted.

An Index for ML retention (MLrI) was conceived. It provided an indication of ML retention for an average fibre of a particular pulp, according to the following formula:

$$MLrI = ML(r<50)x0.25 + ML(r>50)x0.75 + ML(r=100)$$

b) Retention of the S_1 layer (S_1r):

Under transmitted light microscopy using polarized filters, the change in fibril angle of the S_1 with respect to the S_2 layer provides an excellent means of establishing the presence or absence of the former. The S_1 layer has a large fibril angle with respect to the axis of the fibre. Thus, the cellulose microfibrils are arranged almost perpendicular to the light path when a fibre is viewed in cross section. Due to this particular arrangement and to the crystalline nature of cellulose, light is polarized by the S_1 layer and it appears, when present, as a bright line around the fibre (Cote 1981). By rotating the stage in the light microscope, the best polarization for a particular portion of the fibre cross section perimeter could be obtained.

As in the case of ML retention, the following subdivisions were considered in this category:

- No retention of S_1 layer {$S_1(r=0)$}: when there is no evidence of ML retention and no light polarization due

to the S_1 layer, i.e., zero S_1 retention.

- Less than 50% Retention {$S_1(r<50)$}: when there is retention of the S_1 layer but this is less than 50% of the perimeter of the fibre cross section.
- More than 50% Retention {$S_1(r>50)$}: when retention is not total but is more than 50% of the perimeter of the fibre cross section.
- Total Retention {$S_1(r=100)$}: when the S_1 layer is present along the entire perimeter of the fibre cross section, i.e., 100% retention.

The S_1 retention Index (S_1rI) was calculated in accordance with the equation to provide indication of S_1 layer retention for an average fibre:

$$S_1rI = S_1(r<50)x0.25 + S_1(r>50)x0.75 + S_1(r=100)$$

It is clear that when the ML was present, the inner S_1 layer was also present. However, when the ML had been removed, the S_1 layer may or may not have been left behind still attached to the rest of the fibre wall.

c) Exposure of the S_2 layer (S_2e):

Example of S_2 layer exposure can be seen in Figures 3.6 through 3.10. However, observation through crossed Nicols is essential for assessing the possible presence of the S_1 layer on top of S_2 (Figure 3.7). This category is

actually defined by the previous one, i.e., retention of the S_1 layer. For instance, when there is no retention of S_1, there is total exposure of the S_2 layer and *vice versa*. More than 50% retention of the S_1 layer means less than 50% exposure of the S_2 layer. This category was subdivided in a similar manner than the previous two: No exposure $\{S_2(e=0)\}$, less than 50% exposure $\{S_2(e<50)\}$, more than 50% exposure $\{S_2(e>50)\}$ and total exposure $\{S_2(e=100)\}$. The S_2 exposure Index (S_2eI) was calculated as follows:

$$S_2eI = S_2(e<50)x0.25 + S_2(e>50)x0.75 + S_2(e=100)$$

d) Mode of Separation of the Outer Layer:

When the compound middle lamella, with or without the S_1 layer attached to it, was removed from the rest of the fibre wall, separation was observed to occur in three different modes:

- ML Peeling: when there was ML still attached to the fibre surface, but part of it was actually hanging loose at one end, as shown in Figure 3.8.
- ML separated but not broken: when at least a portion of ML, with or without the S_1 layer attached, separates from the perimeter of the fibre. The ends of this portion, however, remain attached to the fibre, i.e., portions of the outer layer are separated but not

removed. This category has been designated here as the "out/in effect". Figure 3.6 illustrates some typical situations found under this category.

e) Radial Failure:

This occurred when the fibre cross section was ruptured radially. Examples of this category are presented in Figure 3.6.

f) Delamination of the S_2 layer:

When there was evidence of separation or delamination within the S_2 layer, whether or not this layer was exposed, as shown in Figure 3.6.

g) Breakdown of Tension Wood Fibres:

Because of the relatively high proportion of TW fibres found in aspen, it was decided to include these in the analysis of cross sections. G-layers can be differentiated even if they are no longer inside the fibre wall, i.e., when the G-layer has been stripped from an opened fibre. Two distinct subdivisions were made for this category:

- G-layer inside a fibre: when TW fibres appeared with the G-layer inside, as in the wood fibres.
- G-layer exposed: when the G-layer has been stripped from the surrounding fibre wall.

Both subdivisions are illustrated with examples in Figure 3.9.

h) Proportion of Distorted Fibres:

While examining fibre cross sections of CTMP and CMP fibres, it was observed that many of these fibres were swollen, more rounded, and even coloured differently from the rest of them, particularly when compared with TMP fibres. Thus, these were called distorted or "d" fibres. Such fibres existed within CTMP or CMP pulps. Examples of distorted in comparison with non-distorted fibres are shown in Figure 3.10.

3.6.2.3. Repeatability

Two different samples of 150 fibres each were processed for each pulp. A Z-test for difference of proportions was performed for each category for every pulp, according to procedures outlined by Walpole (1982).

3.7. Scanning Electron Microscopy (SEM)

The SEM was used as a complement of the light microscope. It was very useful for qualitative examination of surfaces of freeze dried fibres or fractions, analysis of paper surface composition and paper cross sections.

Since samples in SEM can be viewed at very high magnifications, an attempt was made to prepare samples of fibres in cross section for SEM observation. The technique

involved attaching a section to a glass slide and then dissolving away the Spurr resin, following a technique conceived by Maxwell (1978), before normal preparation of SEM samples. A similar principle was applied in the preparation of cross sections of handsheets, where the sheets were embedded and cured in cyanoacrylate glue, cut with a sharp razor blade and finally the glue was dissolved with a mixture of solvents (Williams 1989).

Before a sample could be observed under the SEM, it had to be coated with gold-palladium under a HUMMER IV Sputtering System for 5 min.

3.8. Transmission Electron Microscopy (TEM)

TEM was used in this study not only to confirm the observations made on fibre cross sections in the light microscope, but also to gain additional information in terms of details in the separation of cell wall layers upon mechanical pulping.

Only four samples were chosen for observation under the TEM. These were the high freeness TMP (ATMP-1 and BTMP-1) and low freeness CMP (ACMP-4 and BCMP-4) pulps from both species. Blocks of Spurr embedded fibres were cut into thin sections of 900 A using an ultramicrotome equipped with a diamond knife. The sections were stained in a solution of 1% $KMnO_4$ for 40 minutes before washing in distilled water (Parham 1974). The permanganate reacts with the lignin, thus providing the high electron density contrast means for

differentiating the layers of the cell wall. Because of the differences in fibril angle between the S_1 and S_2 layers, a difference in shade and the perception of a different texture is evident between these two layers, as shown by Panshin and de Zeeuw (1980). Several photomicrographs were taken for each sample.

3.9. Vessel Element Breakdown

The study of VE breakdown during mechanical pulping was done by measuring element particle size in the slides prepared from whole pulps. After measuring VE fragments from each pulp with a 10X objective, starting at a minimum size of 50 um , it was decided to increase the minimum particle size measured to 125 um and to switch to a 4X objective. This is a size range in which the VE fragments could be identified with more certainty. Also, the size distribution in this case included a larger percentage of relatively large fragments; these represent a potential concern for printing papers in terms of surface strength and picking tendency during printing. To investigate this, attempts were made to process these samples by Image Analysis. This system would have provided not only the linear dimensions of vessel fragments but also area values of each particle. Insufficient contrast was achieved for a successful analysis because some fragments retained only one cell wall layer and the staining density was not sufficient even after extensive staining.

Comparisons of size distribution were carried out using Contingency Tables following the Chi-Square test criterion (Steel and Torrie 1980). Differences due to species, processes and refining energy were tested. Whole VE size from kraft pulps was also measured for comparison.

The knowledge provided by the size distribution of VE fragments was complemented by data on the number of whole vessel elements per gram of pulp, which supplied information on the survival of these elements upon refiner pulping.

The number of whole VE was measured as follows: one handsheet was taken from each of the 24 pulps under study and also from the kraft pulps prepared from each species. Eight circles of one cm in diameter were punched out randomly from each sheet. Six of these circles were selected at random, oven-dried, and weighed. The other two circles were soaked in water overnight, stained with toluidine blue and disintegrated according to procedures outlined in Tappi Standard T401, but ensuring that no pulp particles were lost in the process. The disintegrated material was taken to 50 mL and, from this uniform suspension in motion, 2 mL were drawn with a plastic pipette. The pulp material was enough to prepare two 75x25 mm pulp slides. The number of whole VE were counted under a light microscope across the entire area of each slide with the aid of a tally counter. The weight provided by 6 circles allowed these results to be taken to represent the number of VE in one gram of pulp. Then, by using the measured yield values, or assuming appropriate

yields for those pulps from which yield values could not be measured accurately, the number of whole VE per gram of wood was calculated.

Separation of large VE fragments was attempted following the techniques developed by Jacquelin (1966). His technique is based on fibre flocculation. This principle was further applied by Colley (1973) for separation of VE in hardwood chemical pulps. Several stock concentrations and speeds were tried in the Jacquelin Apparatus for refiner pulps produced from chemically treated chips. Even with the sole use of fractions rich in large VE fragments, coherent flocs did not form in quantities large enough to allow separation. The method of Marton and Agarwal (1965), also applied to hardwood chemical pulps, was attempted. However, due to the nature of the mechanical pulps and the relatively small aspect ratio of hardwood fibres, no satisfactory results were obtained. The influence of whole VE on printing properties could have otherwise been tested, provided that handsheets could be produced at similiar levels of surface smoothness. Surface strength and VE picking measurements could have then been attempted before and after removal of whole VE.

IV. RESULTS

4.1. Analysis of Fibre Cross Sections

The results and discussion on the repeatability of the techniques employed in this study for each category of the analysis of fibre cross sections, are presented in Appendix C.

4.1.1. Retention of Compound Middle Lamella and S_1 Layer

Table 4.1 shows the percentage of fibres with different degrees of ML and S_1 retention, including their MLr and S_1r indices, for every pulp studied. The results indicate that the ML and S_1 layer followed similar patterns of removal from the rest of the fibre wall in refiner pulping. TMP pulps presented more fibres with ML or S_1 removed than did either CTMP or CMP pulps. Retention was generally more pronounced in the fibres of CTMP and CMP pulps, but additional removal of the outer layers was accomplished with smaller reductions in freeness than in the case of TMP pulps.

Table 4.2 displays the results of Chi-square tests (Contingency Tables) performed to determine differences in distribution patterns of ML and S_1 removal. The data reveal that the distribution patterns of removal changed significantly for any given refiner pulping process or species as the refining proceeded.

Table 4.3 compiles data for the percentage of fibres which show peeling of the outer layer and that of fibres showing

partial separation of the outer layer (designated as the out/in effect). TMP fibres presented a larger percentage of fibres with outer-layer peeling. The out/in effect was mainly a characteristic of birch refiner pulp fibres and it appeared only occasionally in aspen pulp fibres.

4.1.2. Exposure of the S_2 Layer

Table 4.4 presents the proportion of fibres that were classified under the different categories of exposure of the S_2 layer, as well as their index of exposure. TMP fibres showed higher S_2 layer exposure than CMP or CTMP fibres. However, upon refining, the S_2 layer of the chemically-treated fibres became exposed more quickly, particularly in the case of aspen fibres.

Since the S_2e distribution pattern, when inverted, is identical to the one for the S_1 layer, the results from Table 4.2 also apply, indicating that the S_2e pattern changed significantly upon refining.

4.1.3. Cell Wall Damage

The percentages of fibres presenting radial failure as well as of those showing delamination of the S_2 layer, are summarized in Table 4.5. Radial failure occurred more frequently in TMP than in either CTMP or CMP fibres, whereas delamination was generally more pronounced in the fibres from chemically-treated wood chips and more pronounced for aspen than for birch.

4.1.4. Distorted Fibres

Table 4.6 lists the percentage of distorted fibres from CTMP and CMP pulps. CMP pulps contained a larger proportion of distorted fibres than did CTMP pulps, and aspen, in general, appeared to produce more distorted fibres than did birch.

4.1.5. Tension Wood Fibres

The proportion of aspen tension wood fibres (G-fibres) in the chip supply, as measured from kraft pulp, averaged 31% as presented in Table 4.7. Table 4.8 shows the presence of G-layers in the R48 fraction of aspen refiner pulp fibres, whether in isolation (G-layer stripped from the rest of the fibre wall, i.e., "G ONLY") or inside the fibre, as in the original wood fibre. TMP pulps not only showed a high percentage of G ONLY fibres compared to CTMP and CMP, but also a much lower overall percentage of G-fibres in the R48 fraction.

4.2. Pulp Properties

The pulp characteristics evaluated before handsheets were prepared are presented in Table 4.9. Included are results on refining energy, freeness, shive content and Bauer-McNett fractionation. In general, increasing refining energy was accompanied by reductions in freeness, shive content, and long-fibre fraction (also shown in Figure 3.5), and increases in fines content. TMP pulps consumed more energy

to a given freeness level (also shown in Figure 3.2) than did pulps from CTMP and CMP processes, and produced higher contents of fines.

Handsheet properties are presented in Tables 4.10 and 4.11. Sheet bulk, density and strength properties are shown in Table 4.10. For any given pulp, refining caused increases in sheet density and strength properties, except for tear strength in CTMP and CMP pulps where it followed a more complex trend. The effect of chemical pretreatments on strength properties is evident. CMP sheets were denser and stronger than those from CTMP and both, in turn, had much higher strength than those from TMP. Table 4.11 shows porosity, sheet surface and optical properties. Porosity was reduced dramatically as a result of chemical pretreatment and increased refining energy. The effect of species is also evident with birch giving higher porosity values than aspen. Surface roughness, on the other hand, did not show large variations due to refining energy nor chemical pretreatment when the test was performed on the rough surface of the sheet. However, roughness as measured on the glazed side of the sheet, decreased considerably due to chemical pretreatment and higher refining energy application. Birch produced rougher sheets than did aspen pulps.

As expected, pulp brightness decreased with increasing alkali concentration in the chemical pretreatment, as did the light scattering coefficient. At comparable alkali concentration, aspen always gave brighter pulps than birch.

While brightness stayed constant on further refining, scattering showed an increase for TMP pulps but relatively little change in CTMP or CMP pulps. Opacity showed little variation due to refining. For both species TMP and CMP pulps provided the most opaque sheets, although birch exhibited higher opacity values than aspen.

4.3. Breakdown of Vessel Elements

Table 4.12 presents the average size of VE from aspen and birch kraft pulps. Birch VE were found to be significantly longer than aspen VE with an average of 904 um. The average VE size for aspen was 625 um. Tables 4.13 and 4.14 show VE size frequency distributions in aspen and birch pulps, respectively. For both species, TMP pulps had the larger concentration of small VE fragments, whereas both CTMP and CMP contained fragments of larger sizes, as well as whole VE. Chi-square test results for distribution pattern differences in VE size are shown in Table 4.15. It indicates significant differences among refiner pulps for a given species. These differences were largely due to the different refiner processes under study. When comparing species under the same process, TMP and CMP did not show significant VE size distribution pattern difference.

Results on the number of whole VE per gram of pulp, and estimates for VE per gram of original wood, are presented in Table 4.16. TMP produced virtually no whole VE, whereas their structure was preserved to a large extent in CTMP and

even more so in CMP pulps. Within these last two processes, further refining reduced the number of whole VE.

4.4. Fibre Length and Wood Specific Gravity

Table 4.17 displays the results of fibre length measurements for aspen and birch pulps, showing an average of 1.33 mm for birch. This is significantly higher than the 0.90 mm for aspen fibres.

Wood specific gravity variations from pith to bark at breast height are presented in Figure 4.1 for the three trees involved. Aspen showed a wide range of density values, with an average of approximately 0.45. Birch gave density values which were narrowly distributed around 0.55 with no definite trend in density variation from pith to bark.

Appendix D shows the variation of wood density from pith to bark at different heights up a tree for each of the three trees pulped.

Table 4.1. Percentage of fibres under the categories defined for ML and S_1 retention.

	%ML RETENTION					%S1 RETENTION				
PULP ID	MLr=0	MLr<50	MLr>50	MLr=100	MLr INDEX	S1r=0	S1r<50	S1r>50	S1r=100	S1r INDEX
A-TMP1	33	32	21	14	38	28	17	27	28	52
A-TMP2	40	24	22	14	37	36	22	19	24	43
A-TMP3	37	33	16	14	34	30	25	19	26	47
A-TMP4	47	26	19	8	29	41	13	28	18	42
A-CTMP1	5	15	22	58	78	4	8	22	66	84
A-CTMP2	29	17	19	35	54	24	16	17	43	60
A-CTMP3	39	10	21	31	49	37	6	18	38	54
A-CTMP4	40	23	17	20	39	37	17	17	29	46
A-CMP1	17	20	14	49	65	14	14	15	56	71
A-CMP2	40	19	17	23	41	32	21	14	33	49
A-CMP3	51	14	9	26	36	46	11	11	31	43
A-CMP4	46	21	19	14	33	42	14	17	27	43
B-TMP1	40	11	11	38	49	35	10	14	40	53
B-TMP2	44	12	12	32	44	43	9	13	35	47
B-TMP3	51	7	8	34	41	49	7	7	38	44
B-TMP4	63	9	10	18	28	60	7	9	24	33
B-CTMP1	16	5	5	74	79	16	3	3	78	81
B-CTMP2	33	5	12	50	60	30	4	7	59	65
B-CTMP3	32	8	8	52	60	28	5	6	61	66
B-CTMP4	37	6	12	45	55	35	4	6	55	61
B-CMP1	22	6	9	63	71	22	2	6	70	75
B-CMP2	36	9	9	46	55	31	6	4	59	63
B-CMP3	30	7	10	53	62	26	5	7	62	69
B-CMP4	43	7	1	48	51	31	6	5	58	63

Table 4.2. Dependency of MLr, and S_1r or S_2e, on energy input. Chi-square values compared to a critical value of 16.92 (α=0.05, 9 d.f.).

	MLr		S1r or S2e	
PULP TYPE	ASPEN	BIRCH	ASPEN	BIRCH
TMPs	23.1	46.1	37.9	47.0
CTMPs	164.6	66.8	159.0	42.7
CMPs	144.2	52.9	109.9	20.2

CONCLUSION: Distribution pattern depends on energy input in all cases

Table 4.3. Percentage of fibres showing mode of separation of the outer layer.

	SEPARATION OF OUTER LAYER	
PULP ID	PEELING	OUT/IN
A-TMP1	25	2
A-TMP2	22	3
A-TMP3	22	5
A-TMP4	19	4
A-CTMP1	10	3
A-CTMP2	11	8
A-CTMP3	17	4
A-CTMP4	13	6
A-CMP1	14	8
A-CMP2	14	5
A-CMP3	9	5
A-CMP4	11	8
B-TMP1	14	19
B-TMP2	11	20
B-TMP3	21	10
B-TMP4	11	3
B-CTMP1	8	17
B-CTMP2	7	24
B-CTMP3	9	24
B-CTMP4	11	25
B-CMP1	8	24
B-CMP2	8	18
B-CMP3	7	19
B-CMP4	4	13

Table 4.4. Percentage of fibres under categories defined for the exposure of the S_2 layer.

	%S2 EXPOSED				
PULP ID	S2e=0	S2e<50	S2e>50	S2e=100	S2e INDEX
A-TMP1	28	27	17	28	48
A-TMP2	24	19	22	36	57
A-TMP3	26	19	25	30	54
A-TMP4	18	28	13	41	58
A-CTMP1	66	22	8	4	16
A-CTMP2	43	17	16	24	40
A-CTMP3	38	18	6	37	46
A-CTMP4	29	17	17	37	54
A-CMP1	56	15	14	14	29
A-CMP2	33	14	21	32	51
A-CMP3	31	11	11	46	57
A-CMP4	27	17	14	42	57
B-TMP1	40	14	10	35	47
B-TMP2	35	13	9	43	53
B-TMP3	38	7	7	49	56
B-TMP4	24	9	7	60	67
B-CTMP1	78	3	3	16	19
B-CTMP2	59	7	4	30	35
B-CTMP3	61	6	5	28	34
B-CTMP4	55	6	4	35	39
B-CMP1	70	6	2	22	25
B-CMP2	59	4	6	31	37
B-CMP3	62	7	5	26	31
B-CMP4	58	5	6	31	37

Table 4.5. Percentage of fibres showing cell wall damage.

	CELL WALL DAMAGE	
PULP ID	RADIAL FAILURE	DELAMINATION
A-TMP1	17	3
A-TMP2	19	9
A-TMP3	20	3
A-TMP4	19	16
A-CTMP1	3	10
A-CTMP2	5	21
A-CTMP3	5	21
A-CTMP4	9	28
A-CMP1	4	14
A-CMP2	2	22
A-CMP3	2	20
A-CMP4	5	32
B-TMP1	12	3
B-TMP2	11	5
B-TMP3	21	3
B-TMP4	22	4
B-CTMP1	3	3
B-CTMP2	3	14
B-CTMP3	3	15
B-CTMP4	4	7
B-CMP1	3	4
B-CMP2	3	15
B-CMP3	2	9
B-CMP4	3	10

Table 4.6. Percentage of distorted fibres in refiner pulps from chemically-treated wood chips.

PULP ID	ASPEN	BIRCH
CTMP1	69	64
CTMP2	65	56
CTMP3	54	62
CTMP4	64	49
CMP1	78	80
CMP2	90	81
CMP3	89	77
CMP4	84	78

Table 4.7. Percentage of tension wood fibres in aspen by analysis of cross sections of kraft pulp fibres.

BLOCK No.	TOTAL FIBRES COUNTED	NUMBER OF TENSION WOOD FIBRES	PERCENT TENSION WOOD FIBRES
1	319	100	31
2	299	92	31
3	401	123	31
TOTAL	1019	315	31

Table 4.8. Percentage of tension wood fibres in aspen refiner R48 pulp fractions.

PULP ID	G-LAYER				
	INSIDE		ONLY	TOTAL	
A-TMP1	13	*	6	19	*
A-TMP2	13	*	6	19	*
A-TMP3	11	*	5	16	*
A-TMP4	11	*	4	15	*
A-CTMP1	33	NS	1	34	NS
A-CTMP2	29	NS	1	30	NS
A-CTMP3	27	NS	2	29	NS
A-CTMP4	26	NS	4	30	NS
A-CMP1	32	NS	1	33	NS
A-CMP2	32	NS	1	33	NS
A-CMP3	30	NS	1	31	NS
A-CMP4	25	*	3	28	NS

* : significantly different from G-layer content in wood (level of significance α=0.05)

NS: not significantly different

Table 4.9. Aspen and birch refiner pulp characteristics and fibre size classification.

PULP ID	REFINING ENERGY MJ/kg	UNSCREENED FREENESS mL	SCREENED FREENESS mL	PULMAC SHIVES %	R28 %	28/48 %	R48 %	48/100 %	100/150 %	150/200 %	P200 %
A-TMP1	5.5	373	380	0.76	11.3	37.0	48.3	26.3	2.6	2.7	20.1
A-TMP2	7.8	200	220	0.10	6.1	37.4	43.5	26.5	2.3	2.6	25.1
A-TMP3	9.7	142	161	0.04	4.9	36.6	41.5	27.0	2.3	2.5	26.7
A-TMP4	11.3	106	125	0.02	3.8	36.0	39.8	27.2	2.6	2.7	27.7
A-CTMP1	2.5	285	300	0.10	5.2	40.4	45.6	33.9	3.1	2.0	15.4
A-CTMP2	4.3	159	164	0.05	3.3	39.2	42.5	33.5	3.2	2.8	18.0
A-CTMP3	5.9	112	122	0.05	2.3	36.4	38.7	33.5	3.7	3.5	20.6
A-CTMP4	7.2	71	67	0.00	0.6	22.9	23.5	35.1	5.4	5.6	30.4
A-CMP1	6.8	312	298	0.25	17.1	42.5	59.6	27.1	1.9	0.9	10.5
A-CMP2	7.7	220	214	0.10	17.1	39.0	56.1	26.9	2.6	1.2	13.2
A-CMP3	8.7	154	156	0.00	16.0	37.8	53.8	27.9	2.7	1.2	14.4
A-CMP4	9.7	101	98	0.00	12.2	37.4	49.6	28.7	3.1	1.3	17.3
B-TMP1	5.6	514	552	1.35	13.2	36.9	50.1	21.0	2.3	2.4	24.2
B-TMP2	9.9	205	240	0.02	5.0	36.7	41.7	23.6	3.0	3.0	28.7
B-TMP3	12.3	165	180	0.00	4.6	36.3	40.9	22.5	2.5	2.8	31.3
B-TMP4	14.5	105	116	0.02	2.9	32.5	35.4	25.2	3.2	3.5	32.7
B-CTMP1	3.9	460	507	0.24	21.8	43.0	64.8	15.5	2.2	2.1	15.4
B-CTMP2	5.3	225	254	0.05	20.3	40.4	60.7	15.5	2.3	2.0	19.5
B-CTMP3	5.7	165	203	0.01	24.3	38.1	62.4	15.2	2.2	2.3	17.9
B-CTMP4	6.1	107	143	0.01	23.8	36.9	60.7	15.0	2.3	2.7	19.3
B-CMP1	5.3	317	323	0.30	44.6	25.8	59.2	10.7	1.8	1.2	15.9
B-CMP2	6.8	240	206	0.10	42.4	25.2	67.2	11.4	2.1	1.4	17.5
B-CMP3	6.8	160	150	0.05	43.4	24.4	67.8	10.8	1.7	1.3	18.4
B-CMP4	8.4	90	76	0.00	43.5	21.7	65.2	10.3	2.0	1.7	20.8

Table 4.10. Physical properties of pulp handsheets.

PULP ID	BULK cm^3/g	DENSITY g/cm^3	TEAR INDEX $mN.m^2/g$	BURST INDEX $kPa.m^2/g$	TENSILE INDEX N.m/g	TEA * INDEX mJ/g	STRETCH %
A-TMP1	3.15	0.317	2.4	0.5	14	75	1.08
A-TMP2	2.90	0.345	2.5	0.8	17	95	1.12
A-TMP3	2.64	0.379	2.6	0.9	20	95	1.00
A-TMP4	2.52	0.397	3.2	1.1	24	150	1.23
A-CTMP1	1.91	0.524	5.7	2.0	40	450	1.78
A-CTMP2	1.91	0.524	5.6	2.4	40	395	1.59
A-CTMP3	1.98	0.505	5.4	2.4	44	490	1.79
A-CTMP4	1.68	0.595	4.9	2.7	48	635	2.04
A-CMP1	1.87	0.536	6.0	2.2	37	310	1.32
A-CMP2	1.80	0.556	6.0	2.5	44	475	1.64
A-CMP3	1.71	0.583	6.3	2.8	47	505	1.65
A-CMP4	1.66	0.603	6.6	3.1	49	555	1.71
B-TMP1	3.61	0.277	1.7	0.3	8	25	0.74
B-TMP2	3.10	0.323	2.4	0.6	15	100	1.12
B-TMP3	2.92	0.342	2.7	0.8	19	70	0.91
B-TMP4	2.86	0.350	2.7	0.9	20	105	1.05
B-CTMP1	2.61	0.383	6.3	1.7	35	325	1.53
B-CTMP2	2.46	0.406	7.4	2.2	43	415	1.56
B-CTMP3	2.39	0.419	7.0	2.5	46	450	1.64
B-CTMP4	2.25	0.445	7.7	2.8	52	610	1.90
B-CMP1	2.20	0.454	8.3	2.8	45	480	1.64
B-CMP2	2.10	0.475	9.7	3.1	49	515	1.66
B-CMP3	2.03	0.494	8.3	3.5	55	670	1.90
B-CMP4	1.92	0.522	8.3	3.7	59	745	1.93

(*): Tensile Energy Absorption

Table 4.11. Surface and optical properties of pulp handsheets.

PULP ID	POROSITY Sheffield	ROUGHNESS ROUGH Sheffield	ROUGHNESS GLAZED Sheffield	SCATTERING COEFFICIENT m^2/kg	OPACITY %	BRIGHTNESS %
A-TMP1	172	385	332	58	92.4	64
A-TMP2	81	373	253	64	94.2	64
A-TMP3	45	369	211	65	94.7	64
A-TMP4	29	369	168	67	94.9	65
A-CTMP1	33	366	160	37	89.7	50
A-CTMP2	11	358	100	40	89.7	56
A-CTMP3	8	355	122	44	90.3	59
A-CTMP4	5	349	60	45	90.9	59
A-CMP1	14	398	140	35	94.1	41
A-CMP2	6	390	101	36	94.0	42
A-CMP3	3	384	87	35	94.1	41
A-CMP4	2	375	71	35	94.0	41
B-TMP1	400	402	387	49	96.3	51
B-TMP2	130	385	323	61	97.1	55
B-TMP3	92	378	320	62	97.6	54
B-TMP4	63	372	258	68	98.3	55
B-CTMP1	204	398	345	34	92.3	46
B-CTMP2	38	396	255	35	93.5	46
B-CTMP3	20	398	164	34	93.3	45
B-CTMP4	7	396	126	34	93.0	46
B-CMP1	19	416	182	32	96.1	36
B-CMP2	7	408	142	35	96.9	37
B-CMP3	4	411	146	33	96.2	37
B-CMP4	3	402	164	33	96.4	37

Table 4.12. Size of vessel elements from kraft pulps.

SPECIES	NUMBER VE MEASURED	AVERAGE LENGTH µm	STANDARD DEVIATION µm	STANDARD ERROR µm	COEFFICIENT OF VARIATON %
ASPEN	50	625	128	18	20.6
BIRCH	50	904	189	27	20.9

Table 4.13. Aspen vessel element size frequency analysis (N=50).

CLASS	BOUNDARIES µm	KRAFT	TMP1	TMP2	TMP3	TMP4	CTMP1	CTMP2	CTMP3	CTMP4	CMP1	CMP2	CMP3	CMP4
112.5	162.5	0	25	32	34	37	11	8	11	18	6	13	10	13
162.5	212.5	0	14	7	6	8	7	11	11	12	7	7	7	6
212.5	262.5	0	6	5	3	3	6	8	6	8	4	6	6	5
262.5	312.5	0	0	2	4	1	4	9	10	6	4	2	4	5
312.5	362.5	0	1	2	1	0	4	2	2	1	0	1	3	1
362.5	412.5	3	3	1	1	1	6	3	1	1	2	1	1	2
412.5	462.5	4	0	1	1	0	3	0	0	1	5	2	0	2
462.5	512.5	4	1	0	0	0	3	1	1	0	1	2	4	3
512.5	562.5	6	0	0	0	0	3	2	2	2	2	2	1	2
562.5	612.5	4	0	0	0	0	0	2	2	0	7	4	1	2
612.5	662.5	6	0	0	0	0	0	3	0	1	5	2	3	5
662.5	712.5	8	0	0	0	0	1	0	4	0	2	3	2	3
712.5	762.5	9	0	0	0	0	1	0	0	0	3	5	5	0
762.5	812.5	5	0	0	0	0	1	1	0	0	1	0	2	0
812.5	862.5	0	0	0	0	0	0	0	0	0	0	0	1	0
862.5	912.5	1	0	0	0	0	0	0	0	0	0	0	0	0
912.5	962.5	0	0	0	0	0	0	0	0	0	1	0	0	0
962.5	(+)	0	0	0	0	0	0	0	0	0	0	0	0	1

Table 4.14. Birch vessel element size frequency analysis (N=50).

CLASS	BOUNDARIES µm	KRAFT	TMP1	TMP2	TMP3	TMP4	CTMP1	CTMP2	CTMP3	CTMP4	CMP1	CMP2	CMP3	CMP4
112.5	162.5	0	22	29	34	25	8	7	14	19	13	14	16	23
162.5	212.5	0	12	10	4	13	8	7	9	6	10	7	11	4
212.5	262.5	0	8	9	5	6	5	9	7	9	5	8	3	2
262.5	312.5	0	5	1	4	5	5	7	6	2	5	6	4	8
312.5	362.5	0	0	1	1	0	2	1	3	2	0	3	3	4
362.5	412.5	0	0	0	1	0	2	4	2	2	2	1	3	1
412.5	462.5	0	1	0	0	1	1	1	0	1	1	1	1	0
462.5	512.5	0	1	0	0	0	1	1	2	1	1	3	0	0
512.5	562.5	1	0	0	0	0	5	2	2	2	2	1	1	1
562.5	612.5	1	0	0	0	0	2	1	0	0	2	1	0	0
612.5	662.5	5	1	0	0	0	2	1	1	3	1	0	0	0
662.5	712.5	3	0	0	0	0	1	1	1	1	1	0	2	2
712.5	762.5	3	0	0	0	0	1	3	0	1	1	1	2	1
762.5	812.5	4	0	0	1	0	1	0	1	1	0	0	0	0
812.5	862.5	3	0	0	0	0	3	0	2	0	0	0	1	0
862.5	912.5	6	0	0	0	0	0	0	0	0	1	1	0	1
912.5	962.5	5	0	0	0	0	0	1	0	0	0	0	2	0
962.5	1012.5	7	0	0	0	0	0	2	0	0	3	0	0	0
1012.5	1062.5	3	0	0	0	0	2	0	0	0	0	1	0	2
1062.5	1112.5	2	0	0	0	0	1	0	0	0	0	1	0	0
1112.5	1162.5	2	0	0	0	0	0	2	0	0	1	1	0	0
1162.5	1212.5	0	0	0	0	0	0	0	0	0	0	0	1	0
1212.5	1262.5	4	0	0	0	0	0	0	0	0	1	0	0	0
1262.5	(+)	1	0	0	0	0	0	0	0	0	0	0	0	1

Table 4.15. Chi-square tests on vessel element size distribution (α=0.05).

	PULPS TESTED	NUMBER PULPS IN TEST	DEGREES OF FREEDOM	CHI-SQUARE CALCULATED	CHI-SQUARE CRITICAL	RESULT
A)	ASPEN REFINER PULPS	12	55	221.5	73.3	*
	BIRCH REFINER PULPS	12	44	131.7	60.5	*
B)	ASPEN TMPs	4	6	9.5	12.6	NS
	ASPEN CTMPs	4	15	21.1	25.0	NS
	ASPEN CMPs	4	18	12.9	28.9	NS
C)	BIRCH TMPs	4	9	13.0	16.9	NS
	BIRCH CTMPs	4	18	20.4	28.9	NS
	BIRCH CMPs	4	12	13.4	21.0	NS
D)	ASPEN TMPs vs CTMPs	8	28	110.4	41.3	*
	ASPEN TMPs vs CMPs	8	28	136.8	41.3	*
	ASPEN CTMPs vs CMPs	8	42	65.1	58.1	*
E)	BIRCH TMPs vs CTMPs	8	28	108.4	41.3	*
	BIRCH TMPs vs CMPs	8	28	85.9	41.3	*
	BIRCH CTMPs vs CMPs	8	42	46.7	58.1	NS
F)	ASPEN vs BIRCH TMPs	8	21	28.7	32.7	NS
	ASPEN vs BIRCH CTMPs	8	35	53.4	49.8	*
	ASPEN vs BIRCH CMPs	8	35	41.1	49.8	NS

* : Significantly Different
NS: Not Significantly Different

Table 4.16. Survival of whole vessel elements in refiner pulps.

PULP ID	WHOLE VE PER GRAM OF PULP (x1000)	WHOLE VE PER GRAM OF WOOD (x1000)	WHOLE VE SURVIVAL AS % VE IN WOOD
A-KRAFT	860	482	100
A-TMP1	8	7	2
A-TMP2	3	3	1
A-TMP3	0	0	0
A-TMP4	0	0	0
A-CTMP1	202	186	39
A-CTMP2	167	153	32
A-CTMP3	142	131	27
A-CTMP4	59	54	11
A-CMP1	439	386	80
A-CMP2	389	342	71
A-CMP3	325	286	59
A-CMP4	294	259	54
B-KRAFT	261	136	100
B-TMP1	0	0	0
B-TMP2	0	0	0
B-TMP3	0	0	0
B-TMP4	0	0	0
B-CTMP1	37	34	25
B-CTMP2	36	33	24
B-CTMP3	24	22	16
B-CTMP4	23	21	16
B-CMP1	91	82	60
B-CMP2	73	66	48
B-CMP3	69	62	46
B-CMP4	69	62	46

Table 4.17. Aspen and birch fibre length measurements.

SPECIES	NUMBER OF FIBRES MEASURED	AVERAGE FIBRE LENGTH μm	STANDARD DEVIATION μm	COEFFICIENT OF VARIATION %
ASPEN	169	897	181	20.2
BIRCH	220	1326	266	20.1

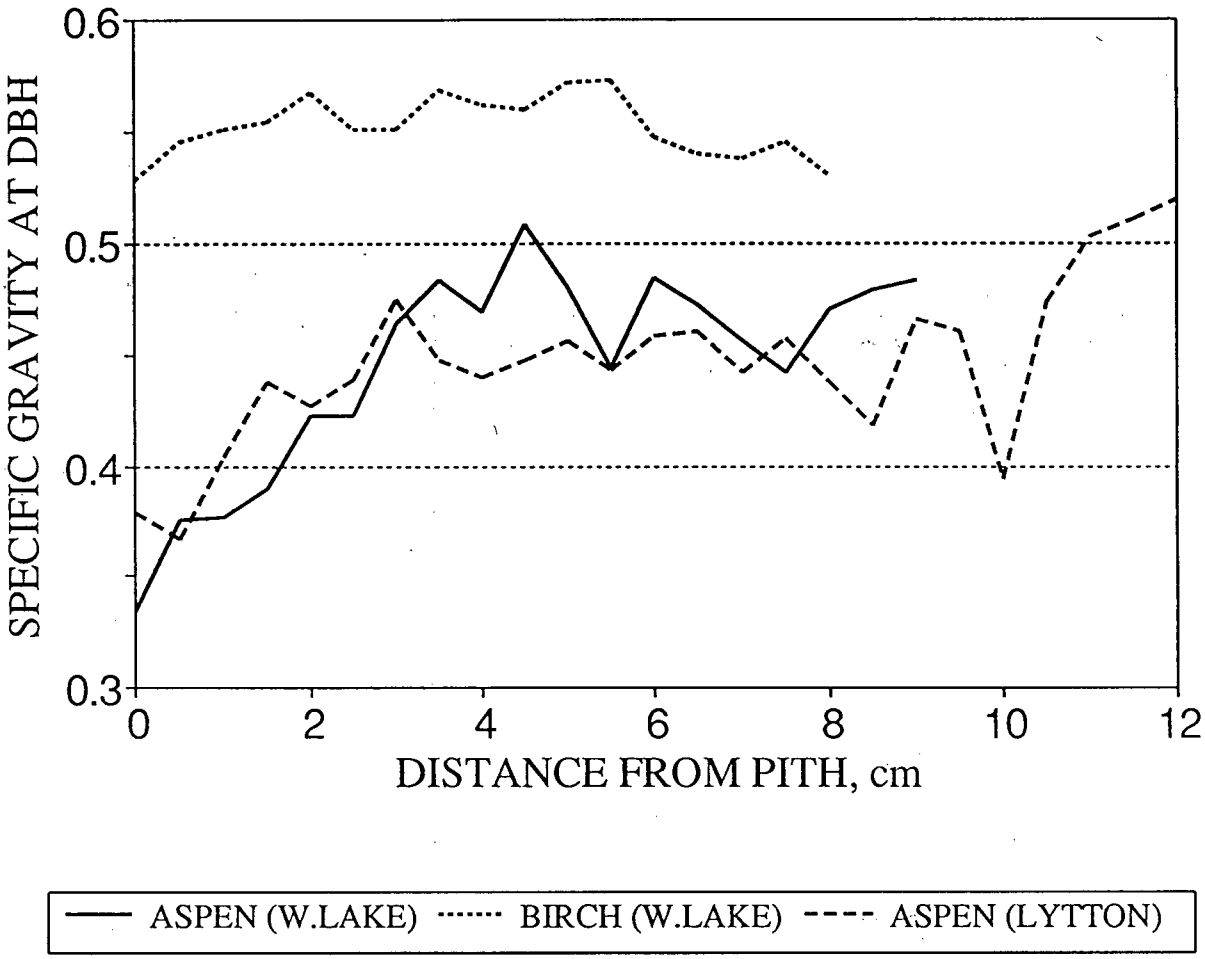


Figure 4.1. Wood specific gravity variation at DBH for the trees used in this study.

V. DISCUSSION

5.1. Analysis of Fibre Cross Sections

This section is divided into topics that will be discussed separately. The discussion of the repeatability of the results obtained by the analysis of fibre cross sections is given in Appendix C.

5.1.1. Retention of Compound Middle Lamella and S_1 Layer

When wood is ruptured in the longitudinal plane, separation can take place in a variety of modes depending on material and testing conditions. Intrawall failure results in the separation within the compound middle lamella, S_1/S_2 boundary and within the S_2 layer. Transwall failure, on the other hand, is responsible for failure across the cell wall, e.g., broken fibres (Koran 1967, 1968). In the present study, however, these types of wood failure do not apply directly since only isolated fibres were analyzed, i.e., fibres that were not part of a shive or fibre bundle. The fibres in the analysis, consequently, had already separated from the wood matrix and undergone some degree of refining. The average surface of fibres initially separated from the wood can thus differ from that of the fibres analyzed in this study. The objective was not to study wood failure, but rather the exposed surface of the fibres produced under different refining processes and after certain degree of refining. Yet, it is recognized that the initial mode of

separation can play an important role in fibre surface quality upon refining.

It should be emphasized that an analysis of fibre cross sections had not been carried out before for hardwood refiner pulps. In fact, published information on the analysis of mechanical pulp fibres in cross section has been based on softwood fibres so far (Kibblewhite 1983). The use of polarizing filters to view the presence or absence of the S_1 layer and, at the same time, to make use of staining techniques to visually assess the presence or absence of the ML, is a new technique and provides a definite advantage in evaluating fibre surface quality. Although polarizing filters had been used to observe the presence of the S_1 layer in wood and wood failure studies, to this author's knowledge it has never been used for quantitative evaluation of S_1 layer retention in isolated fibres from hardwood refiner pulps.

Although the techniques involved in dissolving resin from fibre sections to observe fibre cross sections under the SEM were practical, the SEM observation failed to provide additional information to that obtained from light microscopy. Since staining techniques cannot be applied to SEM samples, it was hoped that changes of texture between fibre wall layers in cross section could be observed under SEM. However, the cell wall layers allowed no differentiation by means of this technique.

Before discussing the results presented in Table 4.1 on ML

and S_1 retention, it is considered important to establish that, after the point of fibre liberation was reached, which occurs at freeness levels of around 700 mL CSF according to Corson (1989), no serious fibre shortening took place in the pulps for the refiner pulping processes under study. Instead, more fines (P200 fraction) were produced as refining energy increased. This is shown in Figure 5.1, which indicates that little change had occurred in the middle fraction (48/100) and the shorter fractions 100/150 and 150/200. The reduction in the R48 fraction of the high freeness pulps, resulted essentially in an increase in the P200 fraction in the lower freeness pulps. This confirms observations by Gavelin (1982a) that one cannot vary the fines fraction without also influencing the fibre characteristics. Although the general shape of these curves were set largely by pretreatment conditions, within any of the six groups of pulp (same process and species, but different freeness levels), it is clear that refining acted upon the fibres mainly by removing surface layers rather than by reducing their length, with the exception of the low freeness aspen CTMP pulp (A-CTMP4), in which some fibre shortening was evident from these curves.

The results presented in Table 4.1. and those of Table 4.2 for Chi-square tests on distribution pattern differences of ML and S_1 retention due to refining, establish the fact that there was quantitative removal of fibre surface layers. As expected, in all pulp groups the percentage of fibres with

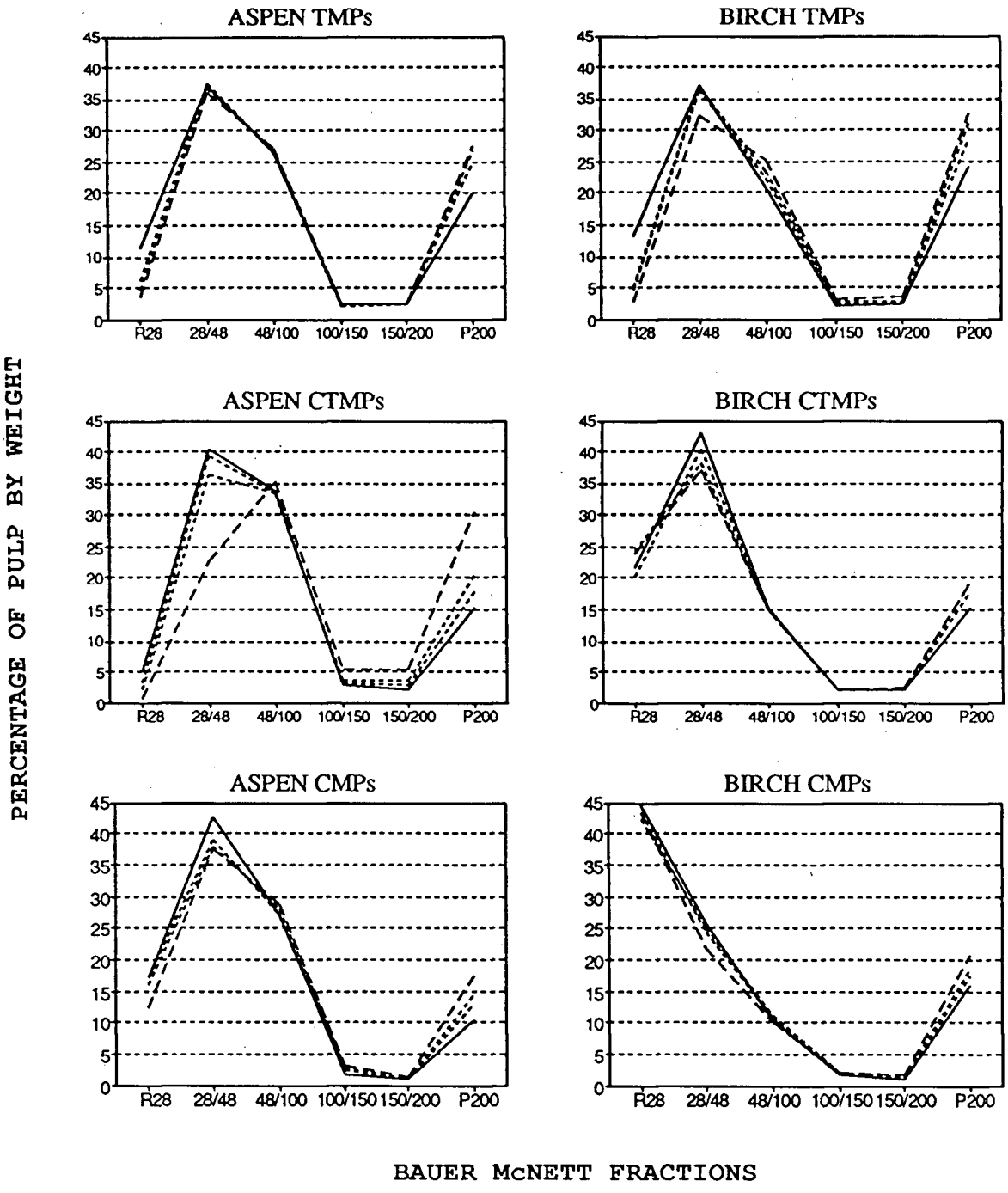


Figure 5.1. Bauer McNett fractionation patterns for all hardwood refiner pulps under study. Solid lines: high freeness pulps; dashed lines: low freeness; dotted lines: intermediate freeness.

total ML retention, ML(r=100), and of MLr index (MLrI), was highest for the pulps with highest freeness values within each group, i.e., for those pulps for which the refining energy applied was also the lowest for the group. In these pulps, the work done on fibre surface development was relatively small compared to that for low freeness pulps. There were, however, important differences between these pulp groups. The trend of ML removal depended on the process and species and was different also with respect to pulp freeness . There were also important differences in the initial retention of ML and in the distribution pattern of ML retention. Figures 5.2 and 5.3 depict the trends followed by the different groups in terms of MLr index and fibres with total retention of ML. Only the general trends are delineated here to simplify the more complex variation within each group of four pulps. The degree of ML retention was generally lower for TMP pulps than for fibres from chemically-treated chips. Presumably, in TMP processing, the lignin present in high concentrations in the ML was not sufficiently softened by the presteaming, preheating or refining conditions to allow fibre separation at the ML zone. This is supported by the lower percentage of R48 fractions found in these pulps compared to CTMP or CMP. Thus, these results indicate that some transwall failure had probably occurred. This agrees with studies by Carlsson and Lagergren (1957) who showed that birch wood failed mainly across the cell wall in tensile testing of heated water-

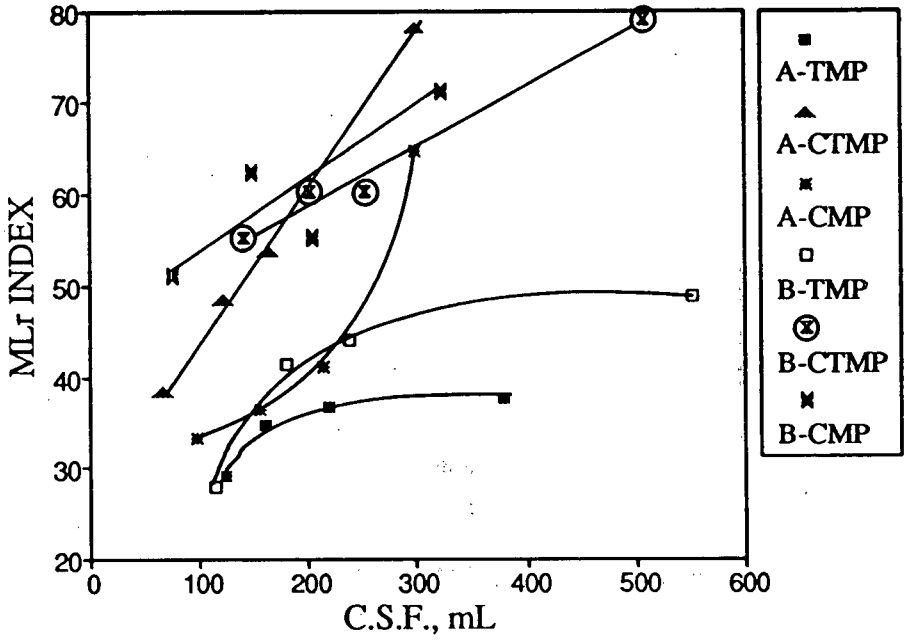


Figure 5.2. Plot of ML retention index against pulp freeness.

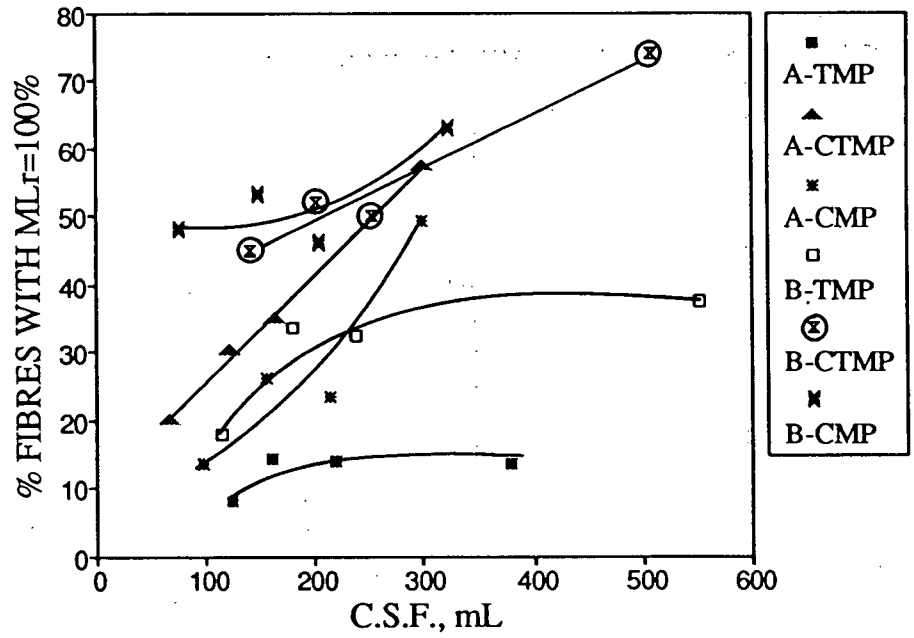


Figure 5.3. Plot of percentage of fibres with total ML retention against pulp freeness.

swollen specimens. It is anticipated that atmospheric refining of untreated chips, an increase in the removal of ML will occur. This also enhances fractionation of the fibres. Studies on *Pinus radiata* (Kibblewhite 1983) confirm this observation when RMP and TMP pulps were compared.

As refining under TMP conditions continued, ML removal proceeded slowly, and little change occurred until the freeness levels were close to 150 mL CSF. Then, a rather abrupt reduction of ML retention was recorded. For birch, the MLr index was reduced from 49% to 28%, whereas in aspen it decreased from 38% to 29%. The exposed surface of TMP fibres appears to be largely set by the initial separation of fibres from the wood matrix, since refining does not easily allow the removal of the ML from the fibre surface unless freeness values of about 100 mL CSF are reached.

The general trends of ML removal for pulps produced from chemically-pretreated chips were quite different from those of TMP pulps. At high levels of pulp freeness, the values for retention of ML were much higher than those for TMP fibres. Fibre separation had occurred largely at the ML and, when freeness values of about 300 mL CSF were reached, the levels of ML retention were still well over 50%. This smooth separation along the ML was accompanied by the expected reduction of shives in these pulps (Table 4.9) compared to TMP pulps.

The chemical pretreatments chosen in this study should cause swelling of the fibre wall, as well as lignin sulphonation

(Giertz 1977). Wood then tends to fail within the ML, and liberated fibres will show high ML retention at high levels of pulp freeness. This was also shown to be the case for birch coarse pulp fibre surfaces produced under high yield sulphite pulping conditions (Iwamida *et al* 1980a). It is interesting to note that ML separation was not the main mode of failure for cold soda pulps. In that case, wood fibres separated mainly between the S_1 and S_2 layer (Wardrop and Dadswell 1958). However, under those conditions little lignin degradation was achieved and the aim was to swell the fibre wall rather than to degrade the ML lignin.

It is shown in this study, in which pretreatment temperatures ranged from 125 ^{o}C to 135 ^{o}C, that ML retention ranged between 65 and 80%, expressed in terms of the MLr index, for birch and aspen CTMP and CMP fibres at freeness levels higher than 300 mL CSF, respectively. The percentage of fibres having total retention of ML was also very high, since it was the major contributor to the MLr index. It is clear that, from the point of fibre liberation, which presumably occurred at the ML, to the point at which further refining rendered freeness values of approximately 300 mL CSF, chemical pretreatment had failed to achieve the same levels of ML removal as those for TMP pulp fibres.

With further refining, however, CTMP and CMP fibres lost their ML much quicker than did TMP fibres (Figures 5.2 and 5.3). Aspen fibres, in turn, showed a quicker response than birch in removing ML, as shown by the slopes of the curves

in these figures. They reached lower levels of MLr at similar freeness.

On the other hand, CTMP and CMP fibres followed a similar trend within each species. For birch, the MLr index seemed to decrease at an equivalent rate following values that were similar for CTMP and CMP. For aspen, the difference between CMP and CTMP was more evident. Although they showed similar trends for ML removal, CMP fibres gave lower values of retention than CTMP at similar pulp drainage. This implies that, for aspen relative to birch, the CMP process is more effective in removing the fibre ML than the CTMP process.

An overall difference between the two species was the relative contribution of fibres with total ML retention to the MLr index. This was much higher for birch than for aspen. The index indicated a value reflecting the MLr of an average fibre, thereby also accounting for fibres with partial retention of ML. From Table 4.1 it is seen that the birch values of ML(r=100) were closer to those of the MLr index than those for the same categories in aspen. This shows that aspen, in general, contained a high percentage of fibres with partial retention of ML compared to birch. This was particularly noticeable for aspen TMP, where fibres with partially retained ML contributed more than twice as much to the MLr index than fibres with ML(r=100). In general, then, aspen refiner pulps contained a large proportion of fibres with "patched" surface patterns of ML retention. Examples of these are given in Figures 5.4 and 5.5.

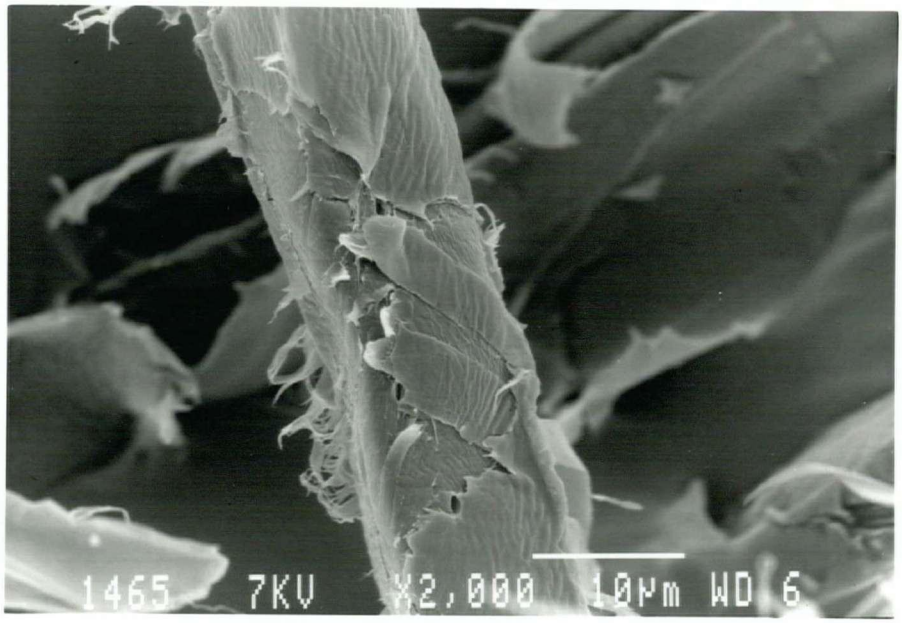


Figure 5.4. SEM photograph of an aspen TMP fibre showing uneven exposure of cell wall layers (Pulp A-TMP4).

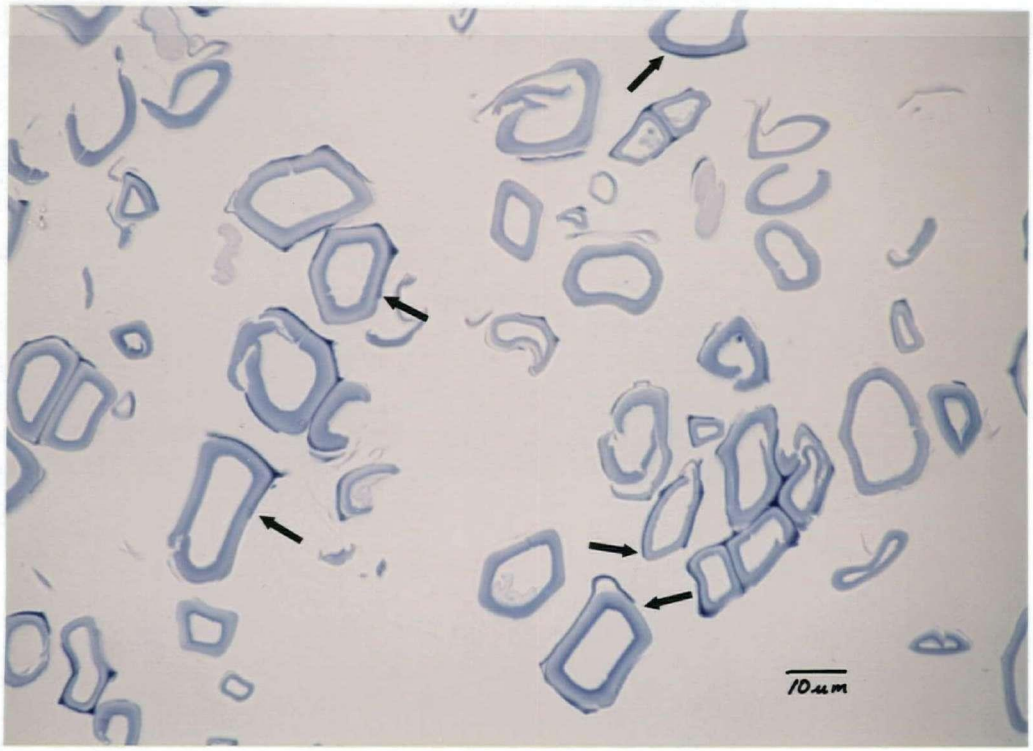


Figure 5.5. Cross section of aspen TMP fibres showing partial retention of ML (pulp A-TMP3).

It is important to note that for a given species, the levels of MLr exhibited by TMP fibres were not exceeded by either of the processes with chemical pretreatment. Aspen CMP and CTMP fibres, however, came closer to the MLr levels of their TMP counterparts at low freeness values than did the corresponding birch pulps. It can be concluded that even the more severe chemical pretreatments of the type used in this study, could not improve ML removal beyond that achieved by TMP processing.

Retention of ML was found to be closely associated with the presence or absence of the S_1 layer. It should be mentioned that it was basically the bulk of the S_1 layer whose presence was recorded in this study. The ultimate exposed layer of the fibre wall would probably be better assessed by observation of the fibre surface under TEM. This technique, however, not only requires preparation of surface replicas, but does not provide information as to what happens to the bulk of the S_1 layer. The retention of the S_1 layer was found to be the main cause for the retention of ML. When ML was removed from the fibre surface, it was largely because the S_1 layer had separated from the S_2 layer. This was found to be the case not only for fibres from chemically-treated chips but also for TMP fibres. Figures 5.6 and 5.7 are examples of separation along or near the fibre S_1/S_2 boundary for birch and aspen TMP fibres.

The differences in S_1 layer retention between processes or species followed the same general pattern as for ML

Figure 5.6. TEM photograph of birch TMP fibre in cross section showing separation of the S_1 layer (pulp B-TMP1).

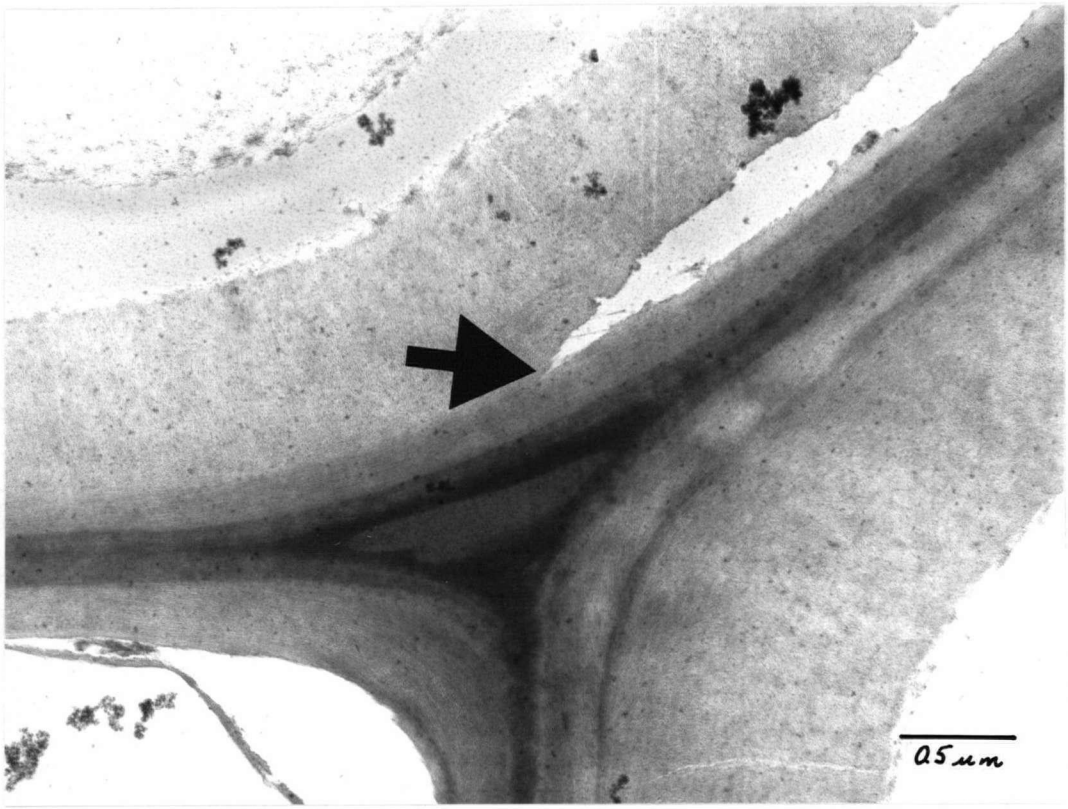


Figure 5.7. TEM photograph of aspen TMP fibre bundle in cross section showing separation of the S_1 layer near the S_1/S_2 boundary (pulp A-TMP1).

retention. There were important differences due to species. For instance, when no chemical pretreatment was applied, there was a larger difference between values of MLr and S_1r for aspen than there was for birch fibres (Figure 5.8). This indicates that for aspen TMP fibres, ML removal did not follow S_1 removal as closely as it did in birch. Thus, aspen fibre surfaces had more areas with exposed S_1 layer. This was in part supported by the fact that many aspen fibres presented partial ML retention. The exposed surfaces ranging from ML to the S_2 layer will have to have a transitional area in which the S_1 is exposed. An example of the S_1 layer being retained and exposed in an aspen TMP fibre is depicted in Figure 5.9.

Basically, the disparity between MLr and S_1r values was small for the fibres of any birch refiner pulp, as illustrated in Figures 5.8, 5.10 and 5.11, indicating that ML removal is a consequence of S_1/S_2 separation. For aspen, these last two figures show that the application of a chemical pretreatment not only reduced the number of fibres with partial retention of ML or S_1 relative to TMP pulps, but also tends to cause a reduction of the difference between MLr and S_1r values. This suggests that for aspen, chemical pretreatments of the type used in this study are responsible for weakening the boundary between the S_1 and S_2 layers, causing increased separation of the S_1 layer. The difference in the severity of the chemical pretreatment appeared to play a comparatively smaller role than the

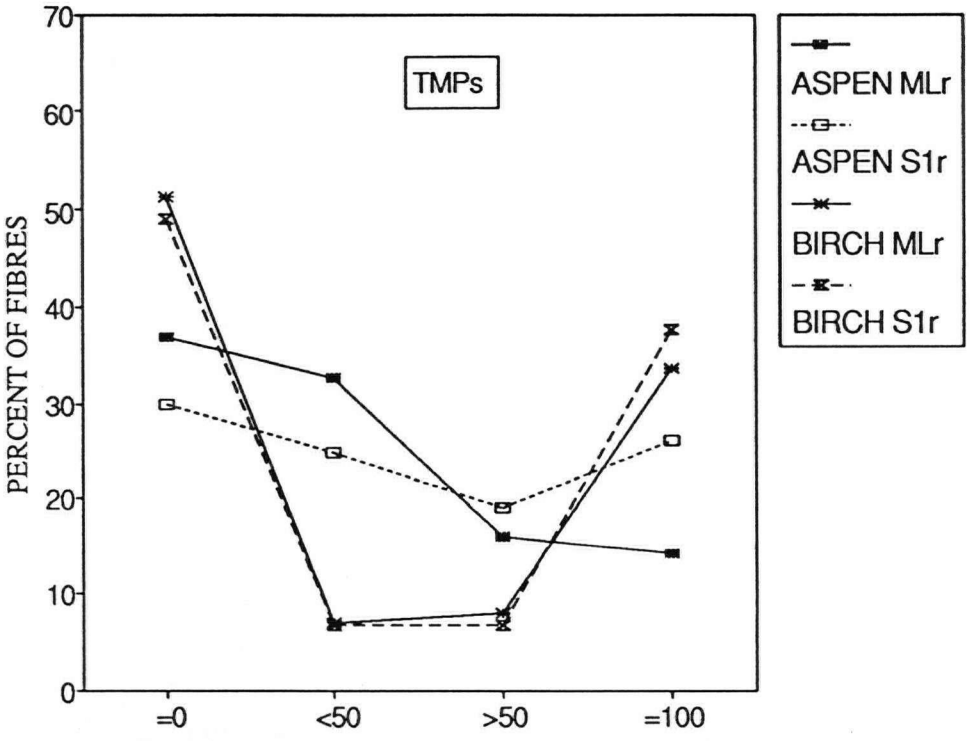


Figure 5.8. Distribution pattern of ML and S_1 layer retention in aspen and birch TMP pulps of similar freeness (pulps A-TMP3 and B-TMP3).

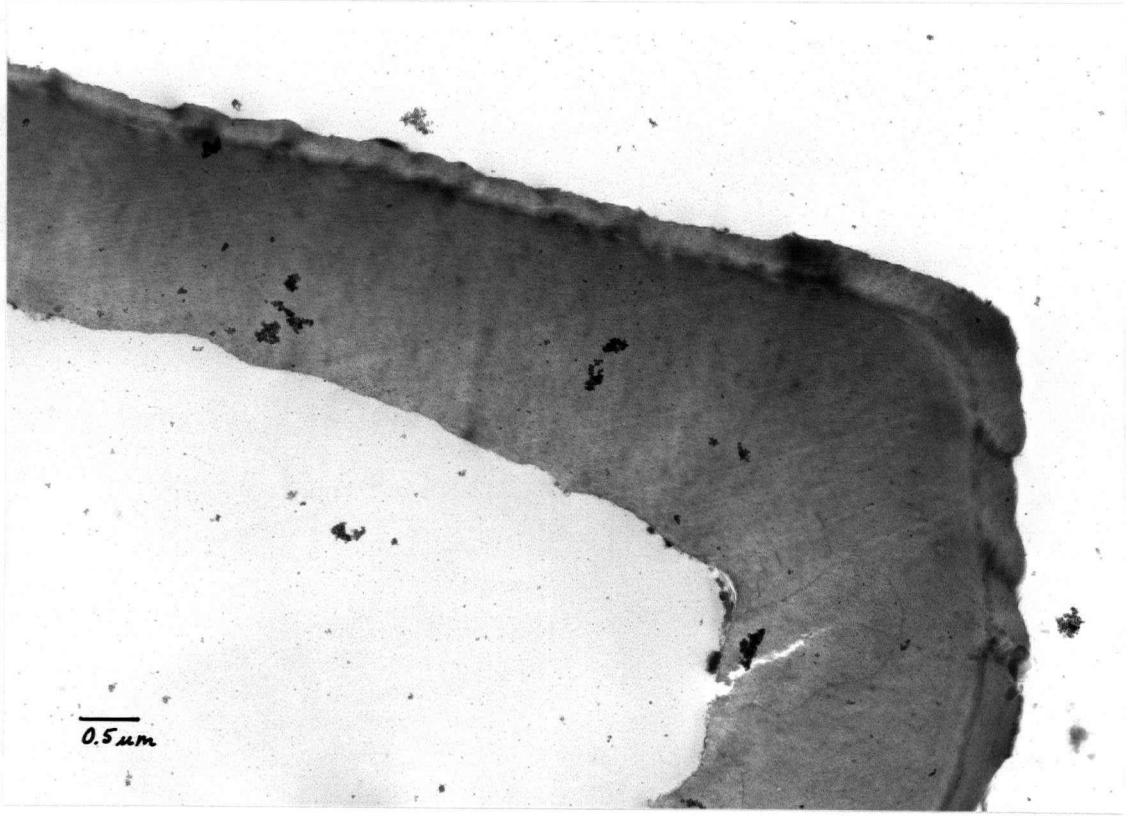


Figure 5.9. TEM photograph of aspen TMP fibre in cross section showing detail of retention and exposure of the S_1 layer (pulp A-TMP1).

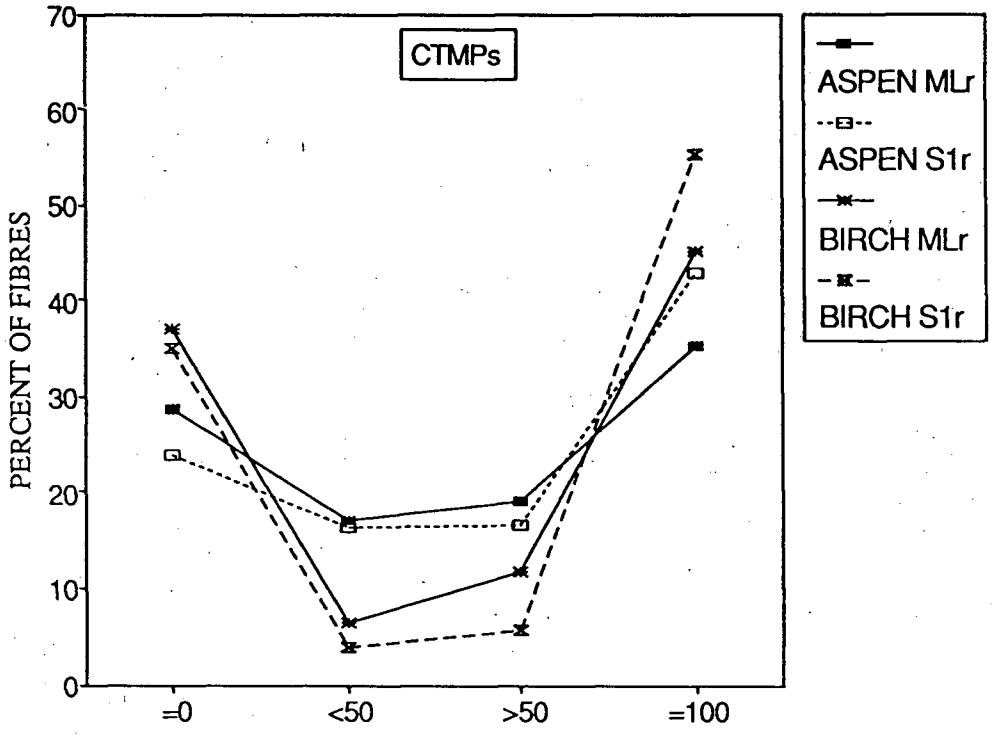


Figure 5.10. Distribution pattern of ML and S_1 retention for aspen and birch CTMP pulps of similar freeness (pulps A-CTMP2 and B-CTMP4).

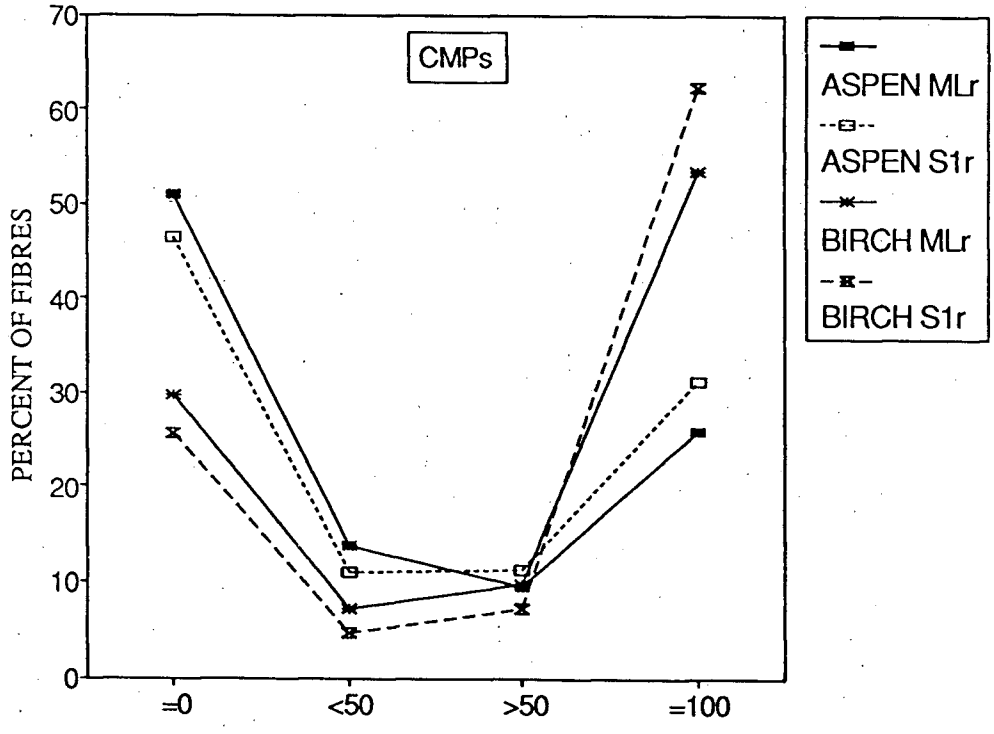


Figure 5.11. Distribution pattern of ML and S_1 retention for aspen and birch CMP pulps of similar freeness (pulps A-CMP3 and B-CMP3).

treatment itself, since the MLr or S_1r patterns were similar for CTMP and CMP fibres for pulps of similar freenesses.

It is evident from Figures 5.8, 5.10 and 5.11, that refining caused a different response in fibre surface quality depending on the processing conditions and on the species. For example, a Chi-square value calculated for the MLr distribution pattern (88.16) for the TMP pulps of Figure 5.8, was significantly larger than the critical value of 7.82. This was also true for the CTMP and CMP curves shown in Figures 5.10 and 5.11, respectively. Thus, it is clear that the distribution pattern of fibre surface quality depends heavily on the type of wood being processed.

For birch refiner pulp fibres, there was evidence of an initial separation between the S_1 and S_2 layers, even when the S_1 layer had not been removed. This visible separation of the S_1 from the S_2 layer was present even at low levels of fibre development (high freeness levels) and was a starting point for further preferential detachment of the S_1 layer along or near the S_1/S_2 boundary. This was referred to as the "out/in" effect presented in Table 4.3. Clearly, birch fibres showed this feature more frequently than did aspen fibres. Birch fibres showed this tendency of producing partial separations between the S_1 and S_2 layers even in unbeaten kraft pulps, as is depicted in Figure 5.12. Presumably, delignification of birch fibres had caused separation between the S_1 and S_2 layers in segments of the

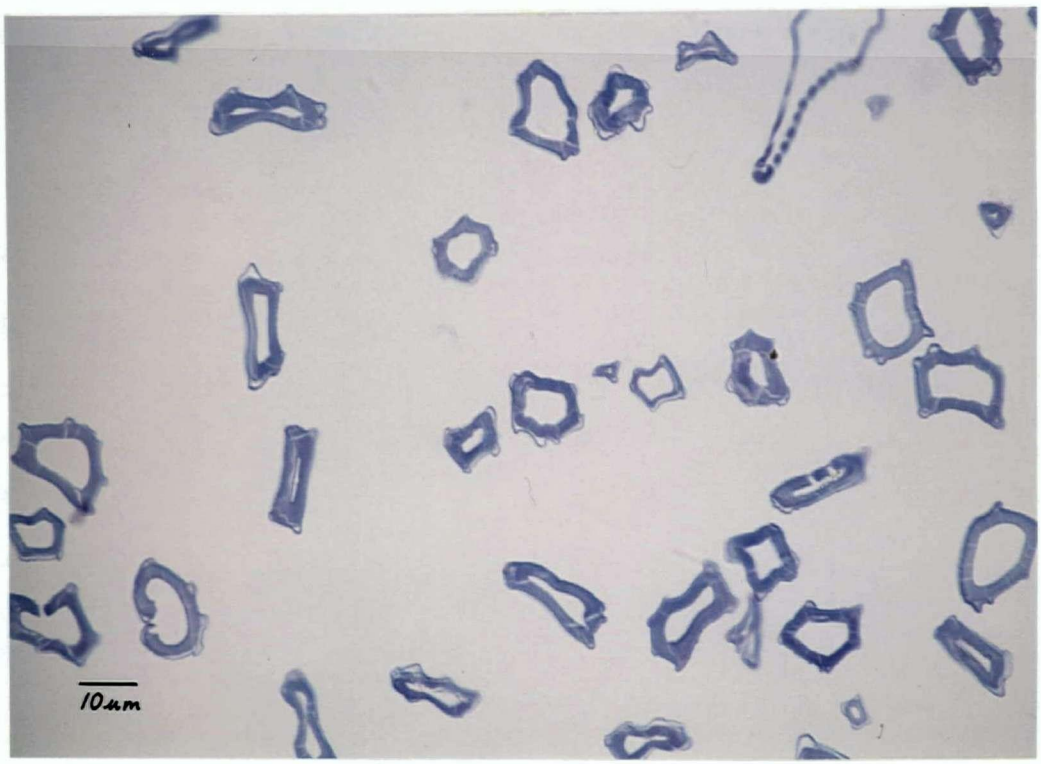


Figure 5.12a. Unbeaten birch kraft pulp fibres in cross section showing gaps between the S_1 and S_2 layers in bright field illumination.

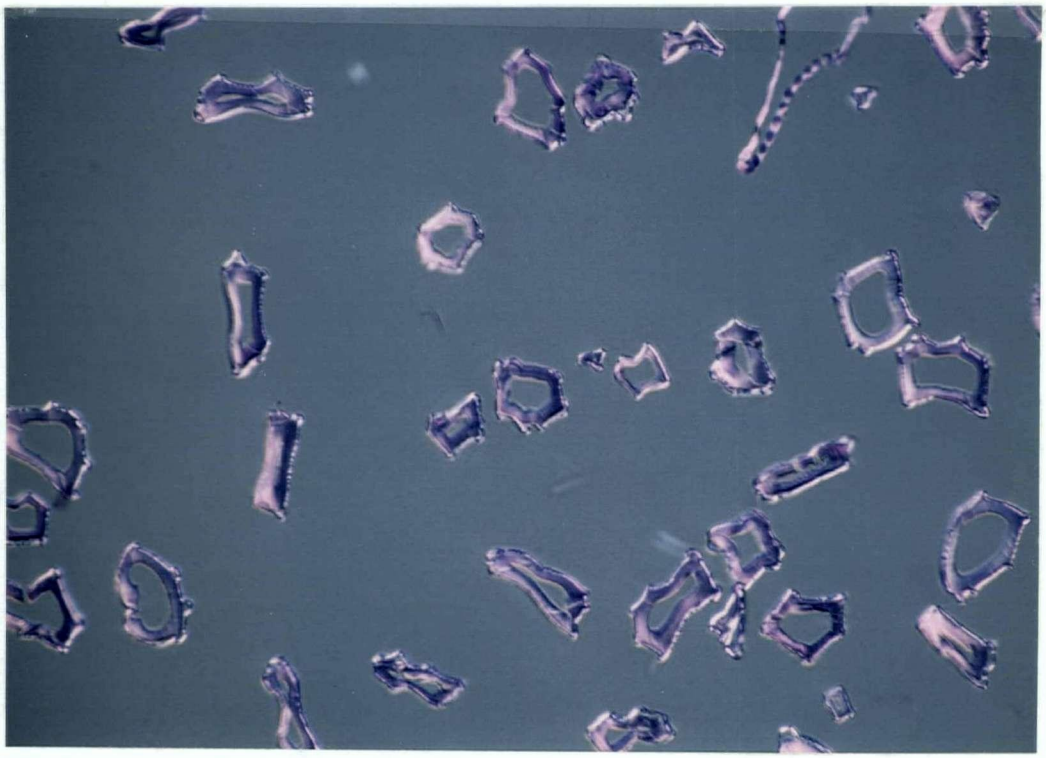


Figure 12b. Same field under partial polarized light.

fibre. By comparison, the unbeaten aspen kraft fibres did not produce these separations (Figure 3.4). Thus, the chemical composition and bond strength at or near the S_1/S_2 interface of the birch fibres appear to be different than those of the aspen fibres. The separation of S_1 and S_2 in birch fibres is thought to be caused by mechanical and/or chemical actions since they were not observed in fibres on wood cross sections.

At freeness levels higher than 300 mL CSF, about 20% of birch TMP fibres demonstrated this out/in effect. At freeness values closer to 100 mL CSF, this feature appeared in only 3% of the fibres. Therefore, most of the S_1/S_2 partial separations had resulted in total separation of the S_1 layer, leaving the S_2 layer exposed. This marked trend was not observed for birch CTMP nor CMP fibres. Instead, the fibres produced from chemically-treated wood chips showed high levels of out/in effect even at low pulp freeness levels. For these fibres, it is evident that the partial separations did not always result in removal of the S_1 from the cell wall. Rather, the S_1/S_2 gaps became longer, to the point where the S_1 had totally separated from the S_2 layer but remained surrounding the fibres, as is shown in the example in Figure 5.13. An extreme case of an unattached S_1 layer is shown in Figure 5.14. These figures indicate that these birch fibres are, at least in part, covered by a sheath of S_1 and ML layers that had separated from the S_2 layer, but were not removed during the mechanical

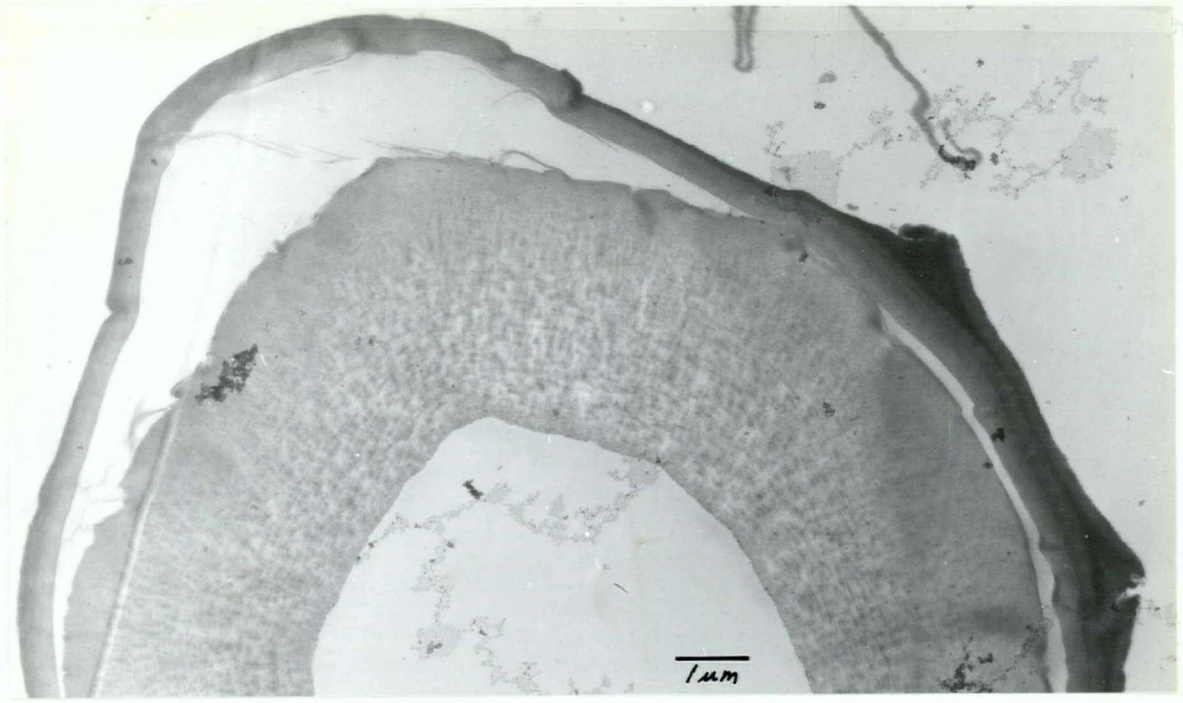


Figure 5.13. TEM photograph showing detail of the gap produced between the S_1 and S_2 layers. The S_1 layer had separated but remained surrounding the fibre (pulp B-CMP4).

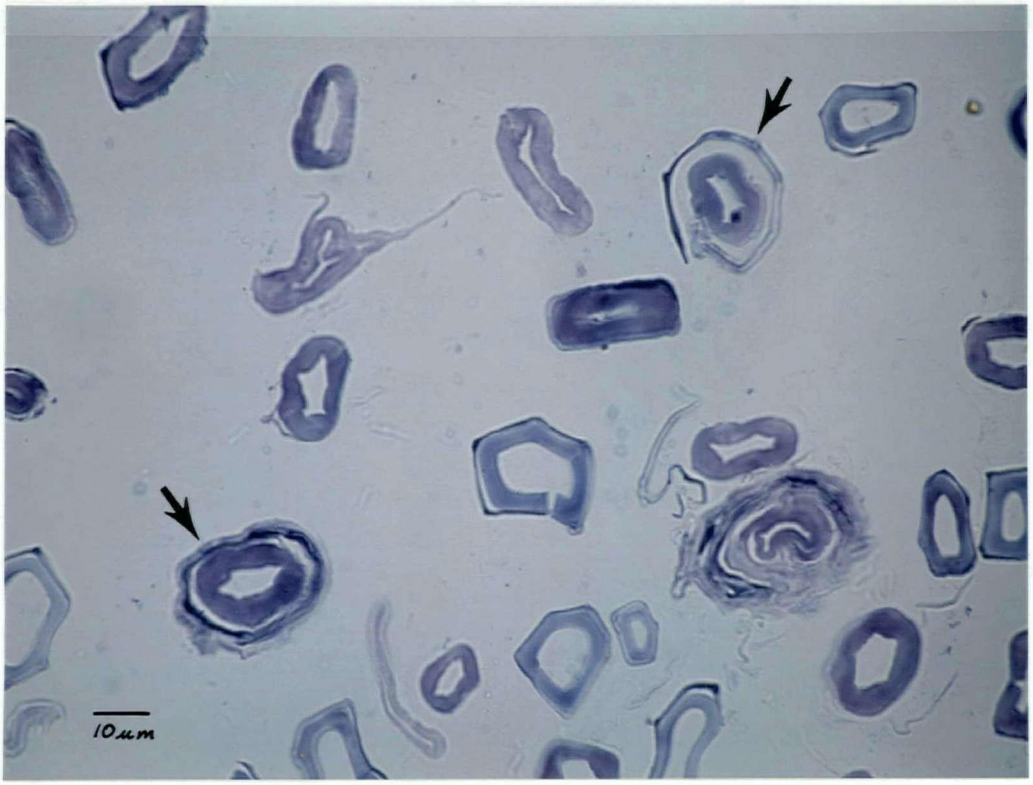


Figure 5.14. Birch CMP pulp fibres in cross section. Note the fibres in which the S_1 layer has completely separated but surrounds the fibre (pulp B-CMP4).

defibration process. The softening of the fibres due to chemical pretreatment seems to be responsible for the increased number of out/in features in birch fibres upon refining, and also for the extended gap between the S_1 and S_2 layers. The S_1 layer has expanded during the process without a memory to retain its original size. Presumably, as fibres go through cycles of compression and decompression stresses in refining (Pearson 1989) as well as longitudinal shear forces (Hoglund *et al* 1976), the relative lack of rigidity causes the fibres to deform momentarily. Layers with high elasticity, such as the high hemicellulose-containing S_1 (Meier 1962), elongate instead of breaking away from the fibre surface in rigid failure, as is more likely the case with TMP fibres. The increased removal of ML and S_1 layers in TMP fibres compared to CTMP and CMP fibres supports this interpretation.

Giertz (1977) reported a "rolling sleeve" mechanism observable on the fibre surface to explain the removal of the S_1 layer and the exposure of the S_2 layer for hardwood refiner pulps produced after chemical pretreatment of chips. He indicated that if hardwood chips were chemically pretreated, the wood was softened in such a way that the primary wall and the S_1 layer were peeled off in the same manner as had occurred with spruce TMP pulp fibres. This was also reported as "skinnings" by Law *et al* (1985). The suggested mechanism for removal of spruce TMP surface layers consisted of the primary wall being rolled back along the

fibre as a sleeve and, simultaneously, the helix of the S_1 layer cracking and fibrils and lamellae being peeled off. This rolling sleeve mechanism was based on observations of pulp slides under the light microscope and not on fibre cross sections. In the present study based on aspen and birch refiner pulps, fibre skinnings were not noted with any frequency in aspen CTMP or CMP pulps. It should also be stated that the number of fibres presenting the out/in effect was relatively small for aspen refiner pulps, the highest being 8% of the fibres analyzed in a sample. In birch CTMP and CMP pulps, however, fibre skinnings were frequent. Figure 5.15 shows examples of these.

The mechanism of rolling sleeve observed in this study was not the one suggested by Giertz (1977). It appears as if the skinnings are due to separation and rolling of the S_1 layer rather than to the primary wall. Figure 5.16 shows a case in which the S_1 layer had been rolled back indicating that the fibre was sectioned at the point at which the skinning appeared. The rolling back seems to be caused by the initial separation of the S_1 layer and not the primary wall external to it. This view is supported by the tendency of hardwoods to fail at the S_1/S_2 boundary for alkali soaked wood (Wardrop *et al* 1961, Carlsson and Lagergren 1957). It should be noted, however, that the mechanism proposed by Giertz was based on birch bisulfite chemimechanical pulp, and not for the alkaline pretreatment conditions used in this investigation. Thus, it is possible that these different

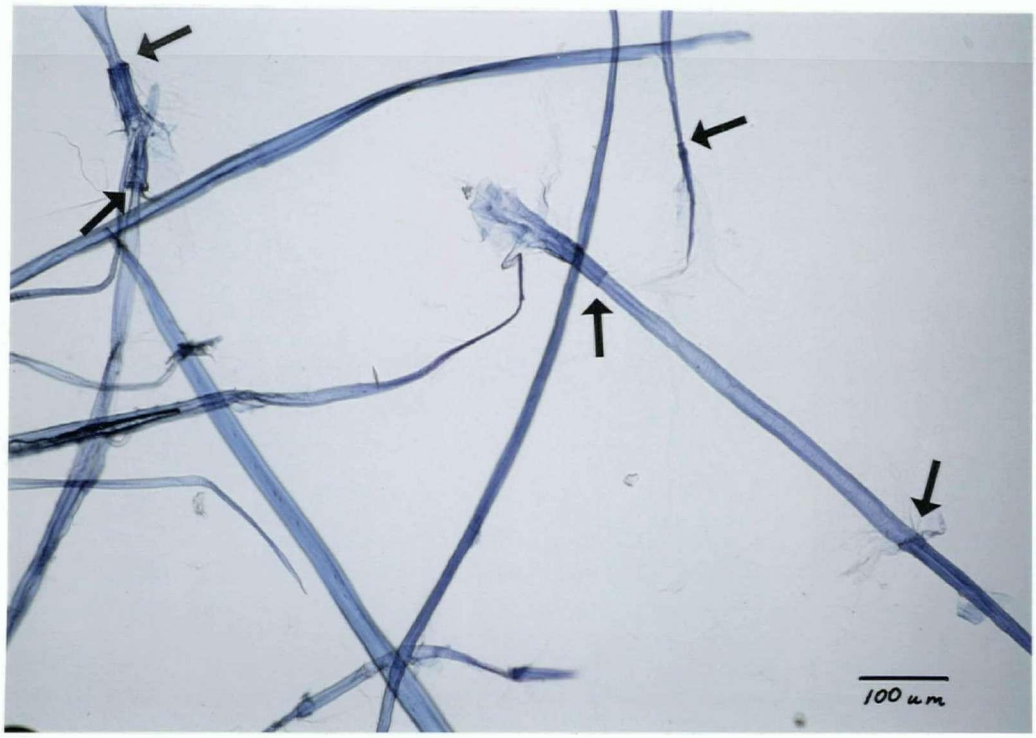


Figure 5.15. Birch CMP fibres showing "skinning" or "sleeve rolling" (pulp B-CMP3).

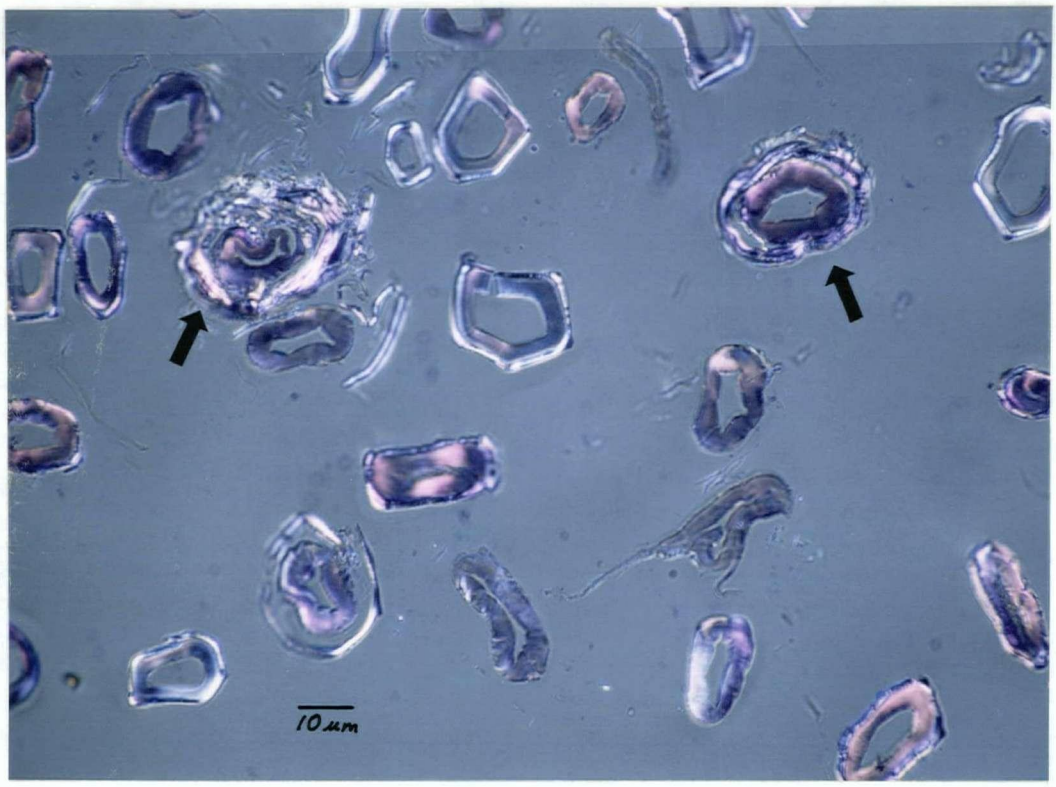


Figure 5.16. Birch CMP fibres sectioned at the point where skinnings occurred along the S_1 layer. Photograph taken under partial polarized light (pulp B-CMP4).

conditions might be responsible for causing different skinning mechanisms to be effective.

The mechanism proposed by Giertz for spruce TMP was attributed to a probable weak transition between the S_1 and S_2 layers. It was indicated that hardwoods might need chemical pretreatment to undergo fibre skinning and expose the S_2 layer. However, in this study birch fibres evidently presented a weak S_1/S_2 boundary, even when no chemical pretreatment was applied. It is not then a matter that skinning and exposure of the S_2 layer is prevented in hardwood TMP due to a suggested stronger S_1/S_2 transition (Giertz 1977). The fact is that, although skinnings did not occur in hardwood TMP fibres, the ML and S_1 layers were removed from the fibre surface in greater proportions than from fibres in CTMP or CMP pulps, probably because of weak S_1/S_2 bonds.

In addition to the out/in effect recorded as a mode of separation of the outer layer, fibres were also classified as either having or not having the outer layer peeled away (Table 4.3). This peeling of the ML also occurred largely on or near the S_1/S_2 boundary. The major differences found in this category were between TMP fibres and either CTMP or CMP fibres. TMP pulps not only showed a higher number of fibres presenting obvious ML peeling --mainly along the S_1 layer-- but also the length of the peeled portion attached to the fibres appeared somewhat longer. This substantiates the observation that for TMP fibres, peeling along the S_1/S_2

boundary was more effective than for CTMP or CMP fibres.

An implication of the mechanism proposed by Giertz (1977) is that chemical pretreatment would cause increased fibrillation of the fibre. This was not observed to be the case for the conditions used in this investigation, as TMP pulp fibres from aspen and birch appeared to be more fibrillated than those from CMP or CTMP pulps. Figures 5.17 and 5.18 compare the extent of fibrillation in aspen TMP and CTMP pulps under phase contrast illumination. Since there was a large difference in the degree of fibre ML and S_1 layer retention between these pulps (over 30%), it is expected that fibrillation of the fibre wall into long filaments originates largely from the S_2 layer. SEM photomicrographs of R48 pulp fibres in Figures 5.19 and 5.20 show that, at lower freeness values, the main difference in the appearance of TMP and CTMP fibres is due to stiffness and not to fibrillation.

Marton *et al* (1979) suggested that the thickness of the S_1 layer was responsible for the poor response of hardwoods to TMP pulping. They reported thicknesses of 0.12 *um* and 0.21 *um* for species of *Populus* and *Betula*, respectively. In the present investigation, it was shown that it was not the S_1 layer thickness that restricted access to the S_2 layer. In fact, birch TMP fibres with thick S_1 layers presented less retention of this layer than did aspen because of the initial weak bond at the S_1/S_2 boundary. It seems that it is the strength of this bond that controls the separation of

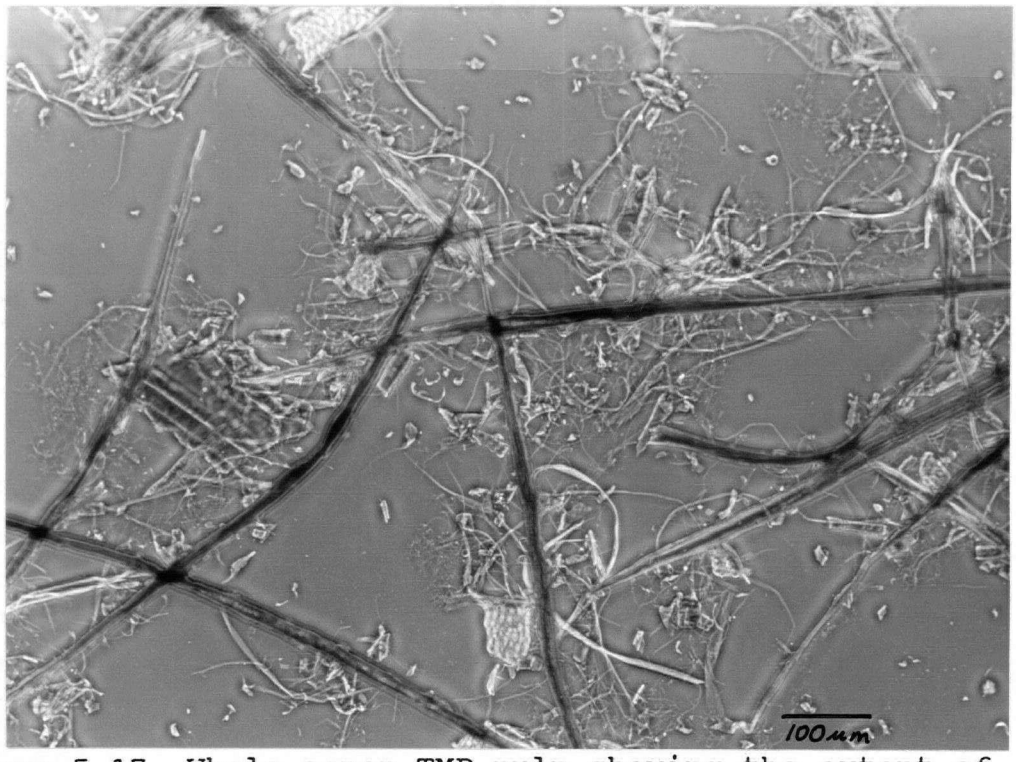


Figure 5.17. Whole aspen TMP pulp showing the extent of fibrillation under phase contrast illumination (pulp A-TMP1).

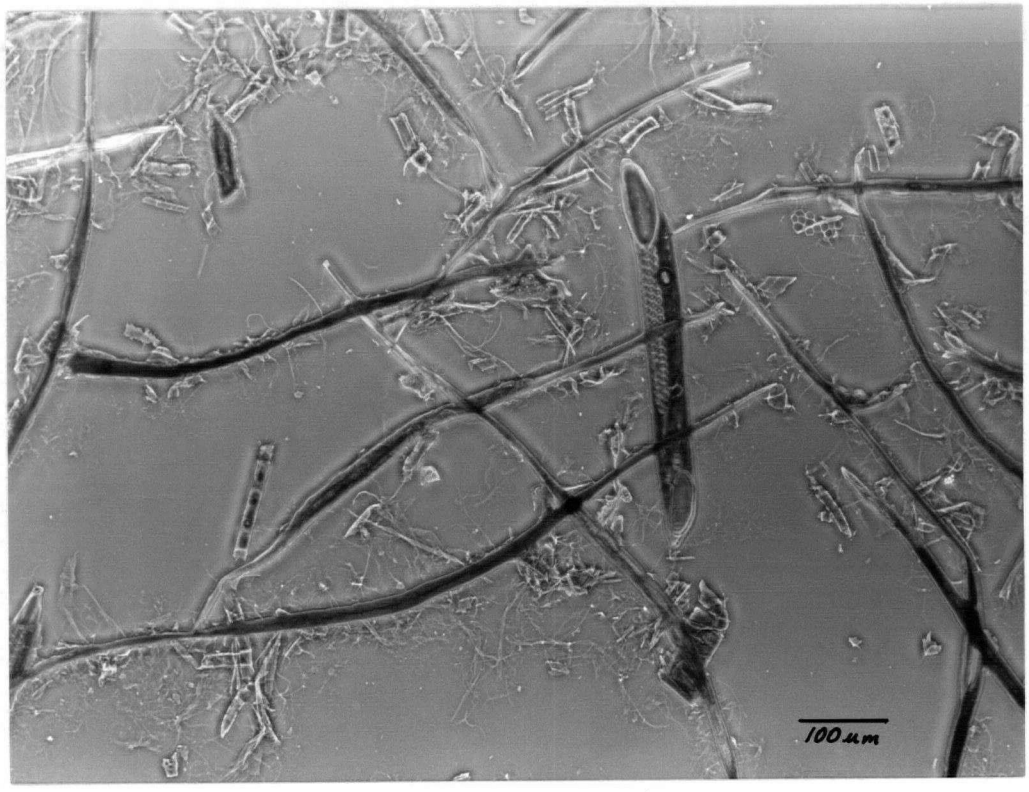


Figure 5.18. Whole aspen CTMP pulp. Note lesser fibrillation compared to the TMP pulp (pulp A-CTMP1).

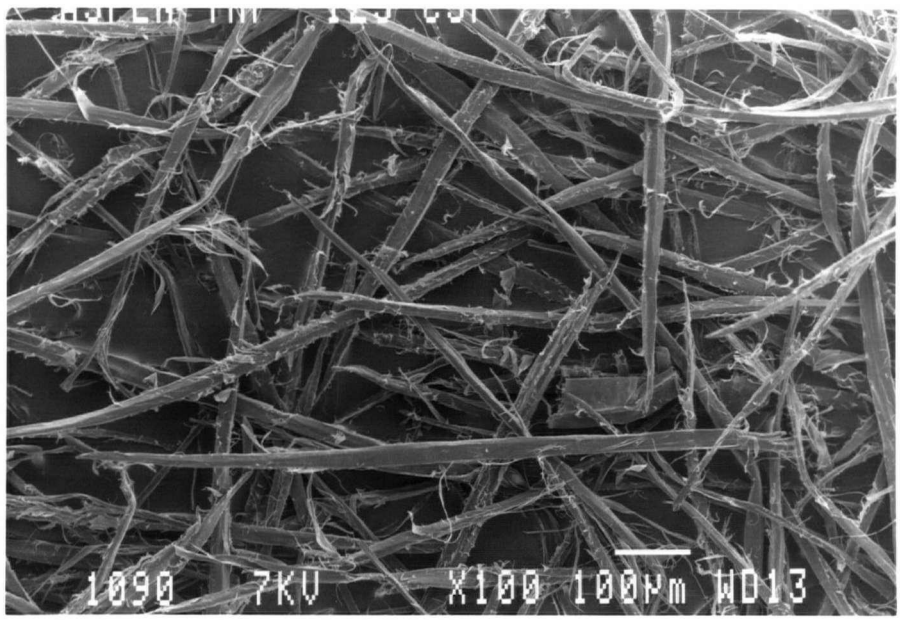


Figure 5.19. SEM photograph of freeze-dried aspen TMP fibres (R48 fraction). The fibres are straight, stiff and fibrillated (pulp A-TMP4).

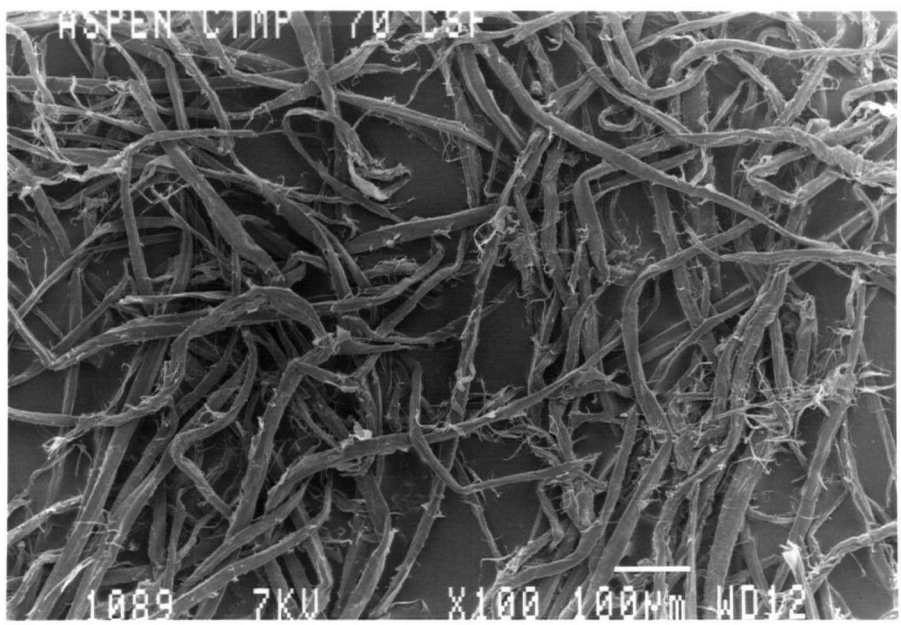


Figure 5.20. Aspen CTMP R48 fraction. Fibres appeared to be more flexible than in TMP (pulp A-CTMP4).

the S_1 layer and, consequently, the exposure of the S_2 layer.

5.1.2. Exposure and Delamination of the S2 Layer

The exposure of the S_2 layer, of course, depends on the retention of the S_1 layer. S_2 layer exposure data are in fact inversely related to those of S_1r, as more retention of the S_1 layer allows less exposure of the S_2 and *vice versa*. The results for S_2e are given in Table 4.4 and are also shown, in part, in Figures 5.21 and 5.22, in which plots are presented of S_2e index and $S_2(e=100)$ against pulp freeness, respectively. As expected from the low retention values of ML and the S_1 layer, TMP pulp fibres had higher S_2e indices than CTMP or CMP pulps made from the same species. Similarly, more fibres had total exposure of the S_2 layer, $S_2(e=100)$, in the TMP pulps. This finding shows that chemical treatments, like the ones used in this study, are not required to cause increased exposure of the S_2 layer.

Birch TMP fibres showed the most S_2 exposure, particularly at freeness levels near 100 mL CSF, even though aspen showed a lower retention of ML. This can be explained as follows. When the ML was removed in aspen TMP fibres, the higher retention of S_1 layers resulted in fibre surfaces with lower S_2 exposure than for birch TMP. Birch TMP fibres had the highest S_2e index and the highest percentage of fibres with total S_2 exposure. The relative contribution of $S_2(e=100)$ to the S_2e index was much higher for birch pulps, relative to

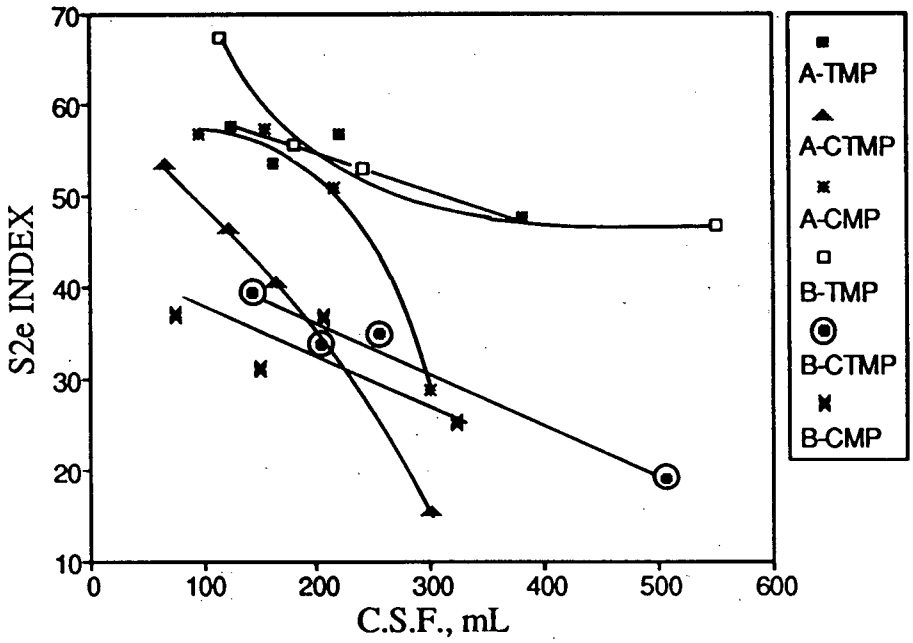


Figure 5.21. Plot of S_2 exposure index against pulp freeness.

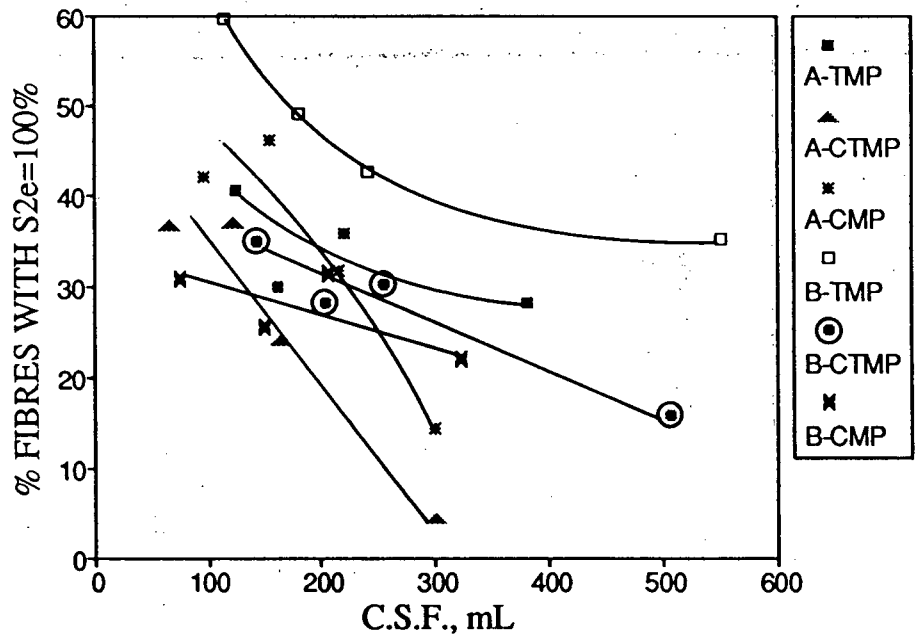


Figure 5.22. Plot of percentage of fibres with total S_2 layer exposure against pulp freeness.

aspen, which carried high proportion of fibres with partial S_2 exposure. The large proportion of fibres in birch TMP pulps featuring a partial separation between the S_1 and the S_2 layers (out/in effect) seems to be responsible for the correspondingly large number of fibres with total S_2 exposure.

The general trends of S_2e are the same as those for S_1r, except that they are inverted. Since, as has been shown earlier, retention of the S_1 layer largely followed the behavior of MLr, the S_2e followed the inverted pattern of MLr, i.e., less retention of ML resulting in more exposure of the S_2 layer. Figures 5.21 and 5.22 show that chemical pretreatments were more effective in exposing S_2 for aspen than for birch, as aspen curves displayed steeper slopes.

Delamination of the S_2 layer was recorded (Table 4.5 and Figures 5.23 and 5.24) to gain insights into the damage and possible fibrillation originating from this layer. Concentric delamination is known to occur in beaten chemical pulp fibres (Wardrop 1963). In fact, the lamellar structure of the S_2 layer in the wood fibre was demonstrated earlier (Ruel *et al* 1978, 1979). Thus, some degree of delamination can be expected, at least for the fibres subjected to chemical treatment prior to refining, because of the expected accessibility within cellulose lamellae and softening of the S_2 layer. In the present study, hardwood refiner pulp fibres did show some degree of delamination. In general, more fibres were delaminated in CTMP and CMP

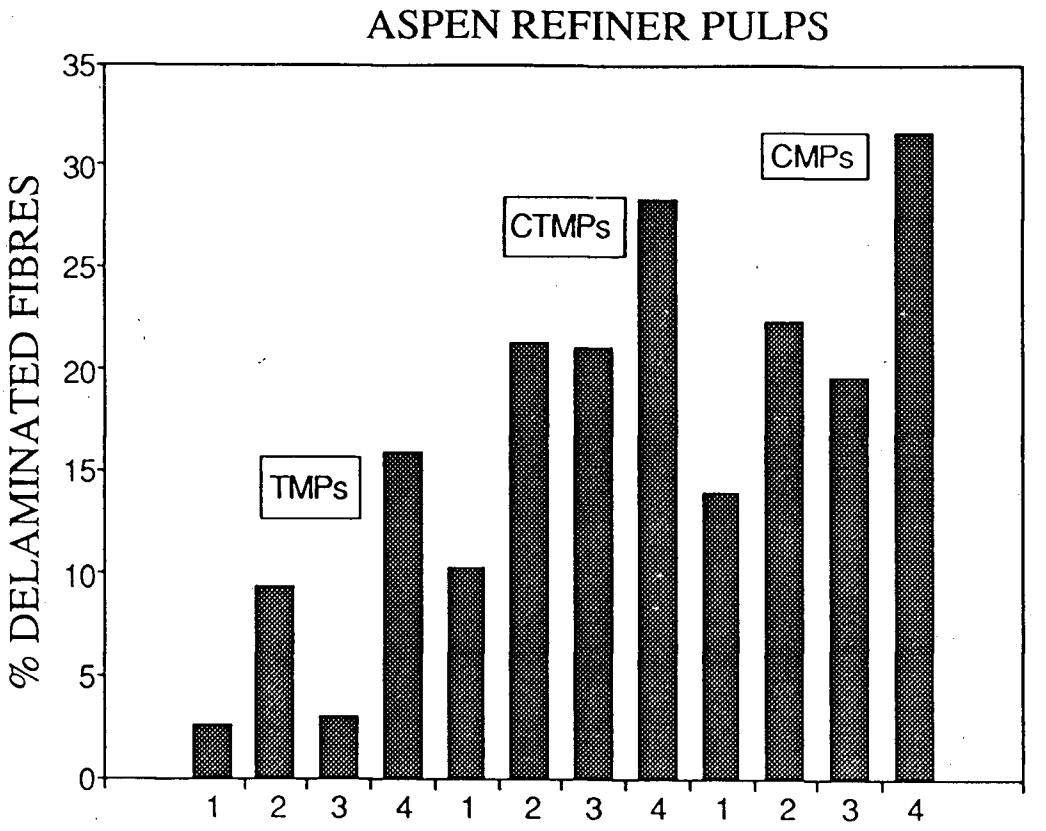


Figure 5.23. Percentage of fibres showing delamination in aspen refiner pulps.

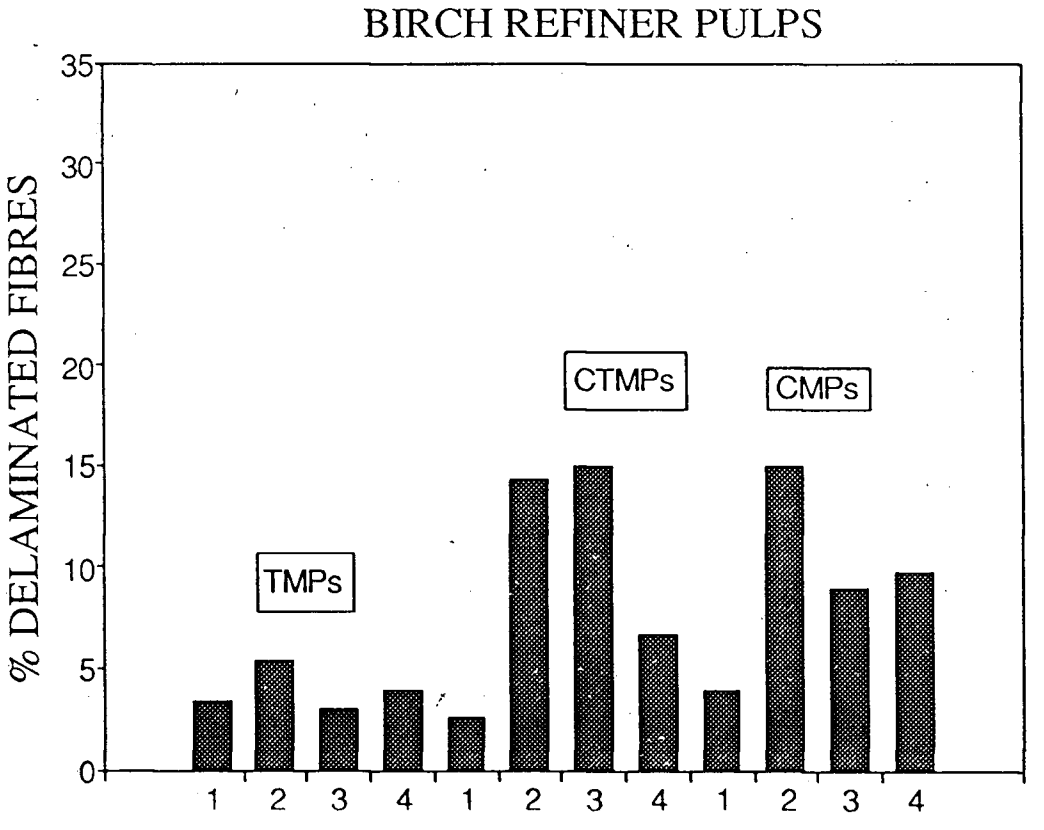


Figure 5.24. Percentage of fibres showing delamination in birch refiner pulps.

than in TMP pulps. As expected, pulps from chemically-treated chips presented more fibres showing delaminations, even though TMP fibres had more exposure of the S_2 layer. Chemical pretreatment combined with refining caused more extensive concentric delaminations than mechanical treatments with limited fibre softening, as in the case of TMP pulps. Therefore, S_2 delamination appears to be related to chemical softening of the cell wall.

Although this feature was recorded to point out the degree of separation within the S_2 layer, as seen under the light microscope, it could be analyzed in conjunction with other recorded features. Table 5.1 indicates that, for most pulps, the majority of the fibres which showed delamination had total exposure of the S_2 layer. This was particularly true for pulps at low freeness levels. The implication is that once the S_2 is exposed, external delamination takes place with the possible production of fines from this layer. However, and although not quantitatively measured, the degree and type of delamination recorded differed somewhat between TMP and the other refiner pulps. In TMP fibres, delamination took place cleanly, as separation occurred within the S_2 layer (Figure 3.6). For CTMP and CMP fibres, delamination appeared to be a gradual peeling of the S_2 layer during mechanical treatment from which fines of a fibrillar nature are produced. An example is shown for birch in Figure 5.25.

Delamination of the S_1 layer was also observed in

Table 5.1. Percentage of fibres with total S_2 layer exposure from among the fibres showing delamination.

PULP ID	% OF S2(e=100) OF DELAMINATED FIBRES
A-TMP1	63
A-TMP2	64
A-TMP3	56
A-TMP4	69
A-CTMP1	7
A-CTMP2	45
A-CTMP3	79
A-CTMP4	66
A-CMP1	14
A-CMP2	54
A-CMP3	64
A-CMP4	64
B-TMP1	30
B-TMP2	50
B-TMP3	33
B-TMP4	67
B-CTMP1	63
B-CTMP2	77
B-CTMP3	96
B-CTMP4	75
B-CMP1	58
B-CMP2	89
B-CMP3	70
B-CMP4	66

Figure 5.25. TEM photograph of a birch CMP fibre in cross section showing delamination of the S_2 layer (pulp B-CMP4).

chemically-treated fibres under TEM. It is known that all layers of the cell wall of a wood fibre exhibit lamellation and that the S_1 layer has a higher porosity than the S_2 (Wardrop 1963). Thus, it is expected that, when the S_1 remained attached to the S_2, some degree of delamination should occur in the S_1 layer in fibres that have undergone chemical and refining treatments. Figures 5.26 and 5.27 show evidence of delamination of the S_1 layer. It is expected that, upon removal, this S_1 delaminated material will produce fines with higher bonding ability than if the S_1 layer were removed intact. Although not quantitatively measured, delamination of the S_1 layer can occur in chemically-treated fibres. The possible breakdown of the S_1 into fibrils after separation and removal deserves further study, as does its role in the formation of fines fractions.

5.1.3. Distorted Fibres

When examining cross sections of fibres from chemically-treated chips, it became evident that the fibres could be classified into two categories: treated and untreated fibres. The appearance of the untreated fibres in cross section resembled that of TMP fibres, while the treated fibres were distorted in shape compared to the former, i.e., more rounded and swollen, as was shown in Figure 3.10. It was thus decided to record the proportion of distorted fibres for CTMP and CMP pulps. Although the separation of these types of fibre would be better done with an image

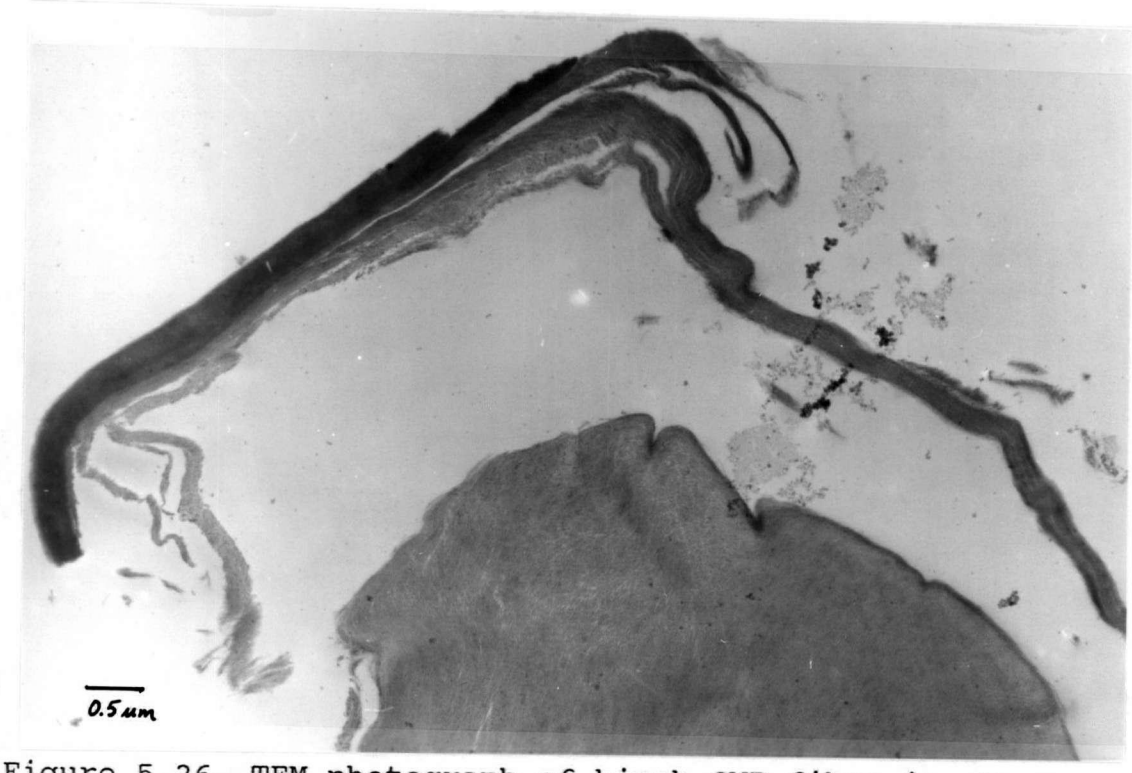


Figure 5.26. TEM photograph of birch CMP fibre in cross section showing detail of delamination of the S_1 layer (pulp B-CMP4).

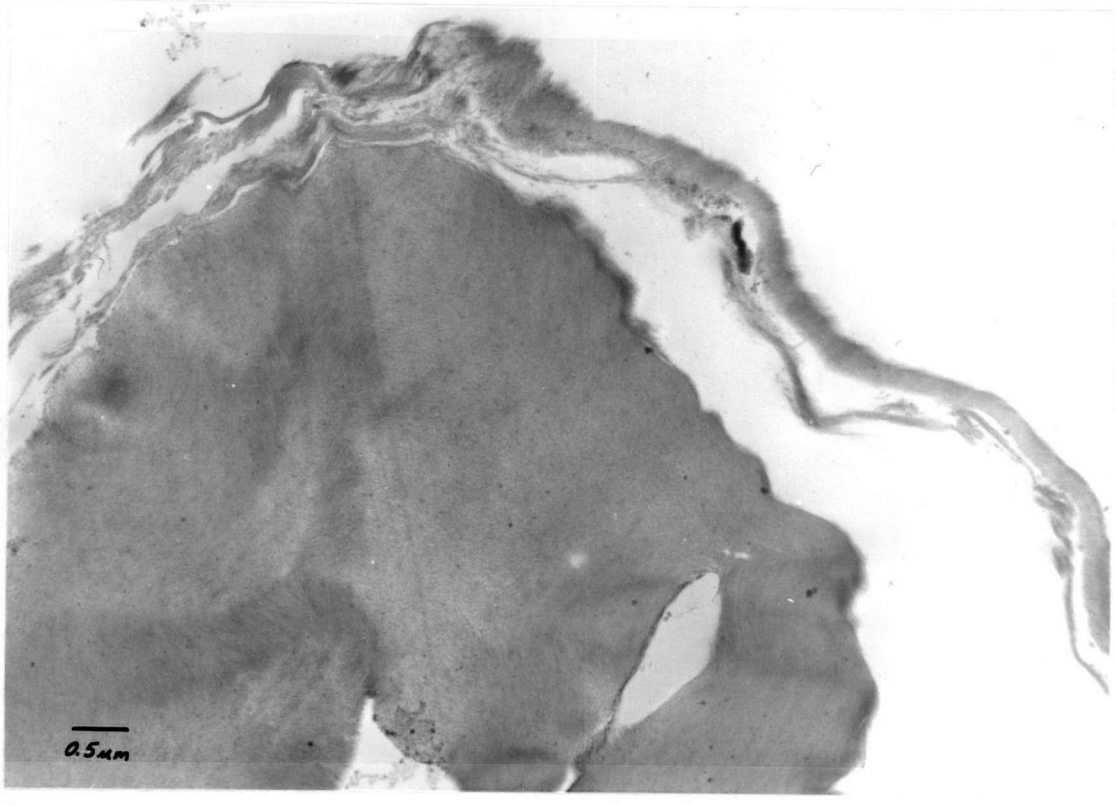


Figure 5.27. Another example of S_1 layer delamination (pulp B-CMP4).

analysis system that could detect differences in staining intensities and shape factors, an estimation of the number of distorted fibres was done under simple light microscopy. Obviously, there were many fibres that could be considered transitional between untreated and treated. These fibres were classified according to their similarity to fibres that were obviously untreated or obviously distorted.

The results (Table 4.6 and Figures 5.28 and 5.29) showed that there was a large proportion of distorted fibres in refiner pulps from chemically-treated chips, approximately from 50-70% for CTMP and from 75-90% for CMP pulps. There seems to be a tendency for aspen to produce more distorted fibres than birch, possibly due to better liquor penetration.

Since liquor penetration is largely initiated through the vessels (Wardrop 1963), the higher values for aspen are consistent with its higher number of VE per unit area of vessels in wood and its thinner fibre walls. On the other hand, the chemical pretreatments were markedly different. CMP treatments involved not only higher concentration of chemicals, but also longer times and higher temperatures than CTMP. The results of the number of distorted fibres showed this difference. Nevertheless, the large difference in chemical treatment conditions provided only a comparatively small difference in the number of distorted fibres between CMP and CTMP. Presumably, this was due to the CTMP screw press in the impregnation system

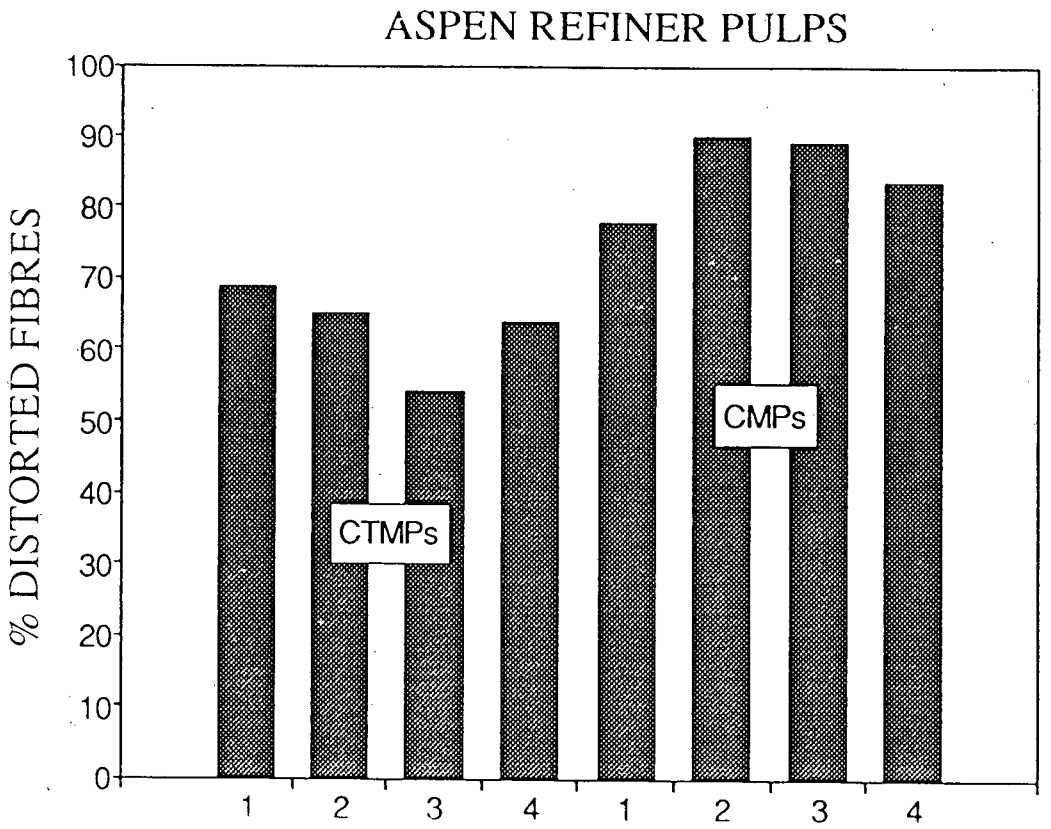


Figure 5.28. Percentage of fibres showing distortion in aspen CTMP and CMP pulps.

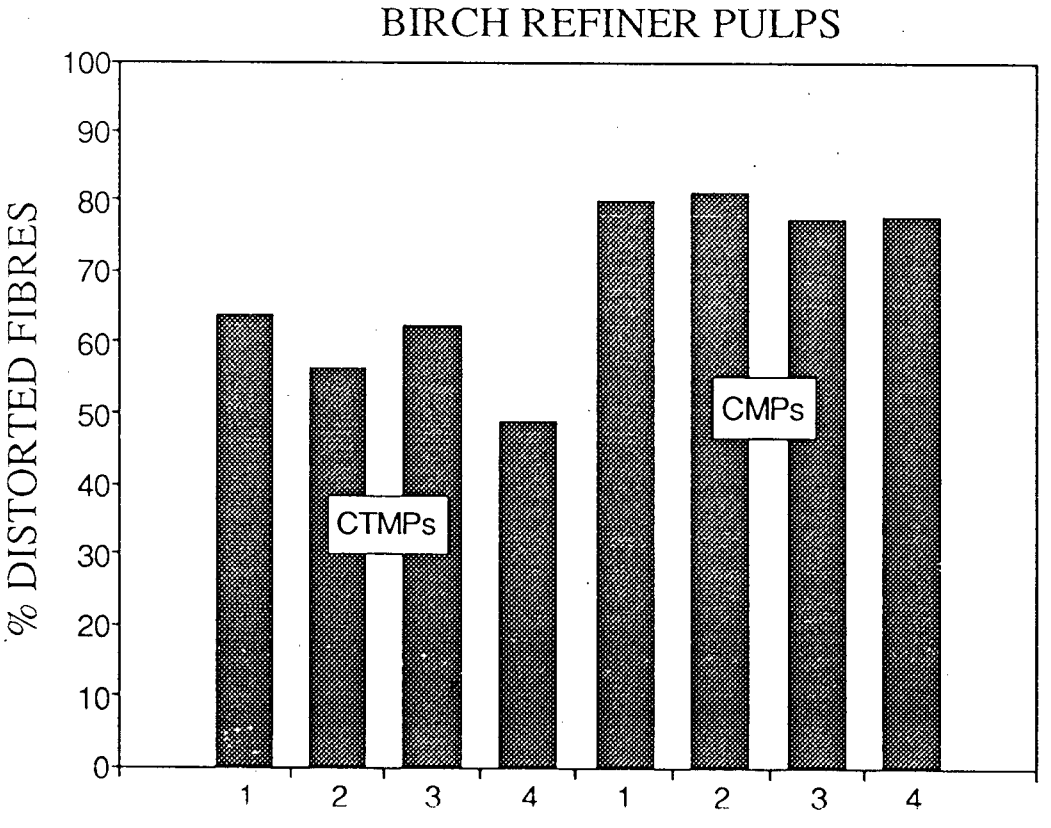


Figure 5.29. Percentage of fibres showing distortion in birch CTMP and CMP pulps.

(compression ratio of 3:1) which caused cracks in the chips through which liquor penetrated.

One evident application of the number of distorted fibres is the investigation of liquor penetration and uniformity of fibre treatment. Results in this study showed that both CTMP and CMP pulps, produced under the conditions used here, are mixtures of treated and untreated fibres, although most fibres were chemically treated particularly in the case of CMP pulps.

It should be mentioned that the degree of fibre distortion between species or processes is not known, only the proportions. The effect of refining on the number of distorted fibres did not follow a definite trend. More precise information, possibly provided with the help of an image analysis system, would be needed to draw conclusions on the effect of refining on fibre shape changes.

5.1.4. Radial Failure (RF)

It has been shown in early studies that the formation of ribbons is highly desirable for the production of a good stone groundwood or refiner mechanical pulp (Forgacs 1963). These ribbons enhance bonding, light scattering and smoothness of mechanical pulps (Mohlin 1982b). Although fibre radial failure may originate in part from initial transwall failure in refining, it could also start from crack development on intact fibres. According to Forgacs (1963), ribbon formation starts with the radial failure

(cracks or splits) of the fibre and the subsequent unravelling of the cell wall along the S_2 layer. Thus, the number of fibres in which radial failure occurs relates to the frequency of ribbons in the pulp. Ribbon formation has been observed not only in mechanical pulps from softwoods but also from hardwoods (Vecchi 1969, Scaramuzzi and Vecchi 1968). This characteristic may appear in greater proportions in the middle fraction rather than in the R48 that was used in this study. However, the 48/100 resembled the R48 fraction in the pulps studied, in terms of fibre shapes. The R48 fraction is expected to provide reliable information on the relative amounts of ribbons produced.

The proportion of fibres that failed radially across the cell wall (Table 4.5 and Figures 5. 30 and 5.31) was clearly higher for TMP pulps and was only minor in most CTMP or CMP fibres, although it approached 10% in A-CTMP4. RF in this pulp might be associated with the fact that this particular pulp showed a reduced R48 fraction implying that fibre cutting may have been prevalent during refining (Figures 3.5 and 5.1.). Otherwise, chemically-treated fibres were softened to the point where refining did not crack the fibre wall to the same extent as in TMP, but little differences in RF were found between CTMP and CMP fibres.

It is anticipated that RMP will produce even higher proportions of radially failed fibres since refining is done under atmospheric conditions and the fibres will be stiffer. Presumably, this would cause more transwall failure as well

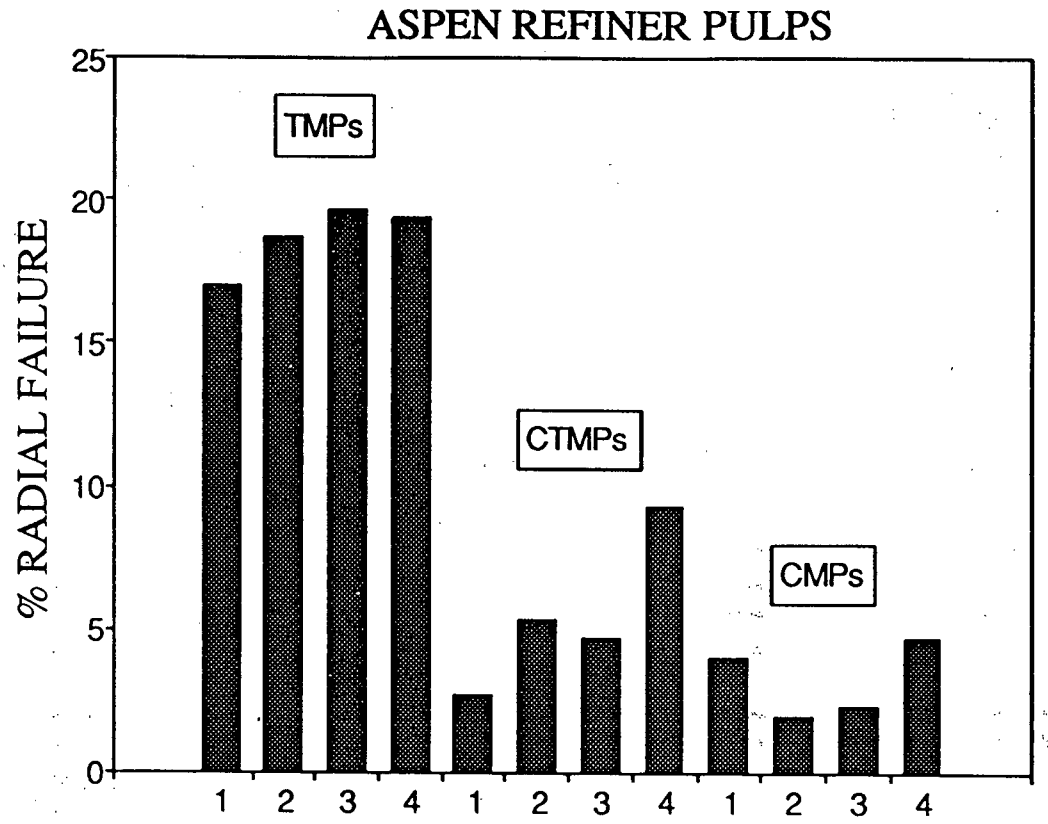


Figure 5.30. Percentage of fibres showing radial failure in aspen refiner pulps.

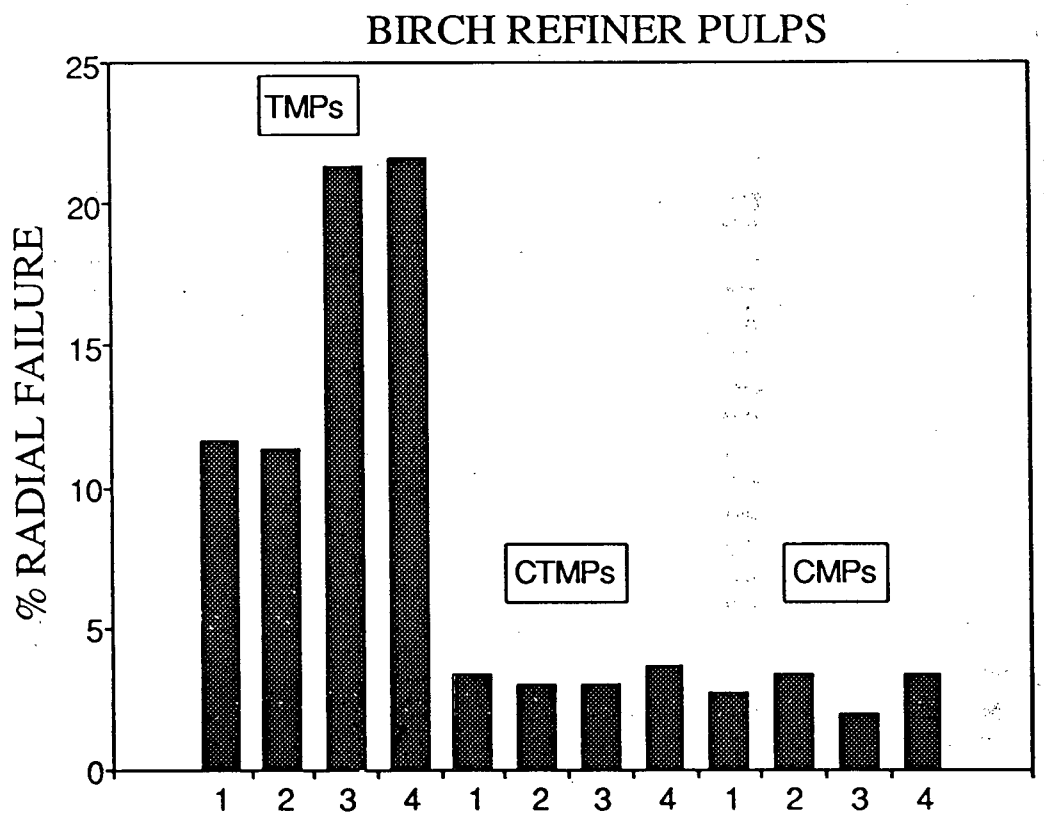


Figure 5.31. Percentage of fibres showing radial failure in birch refiner pulps.

as more cracks in the fibre wall.

Whereas little change in the number of radially-failed fibres due to further refining was detected for aspen TMP pulps, birch TMP pulps showed higher proportions of RF at low freeness. The low number of fibres with RF at high freeness indicates that birch may need more refining than aspen to produce and propagate cracks along the S_2 wall. This may be related to the thicker cell walls of birch fibres.

The results for RF in TMP fibres in this investigation (RF in approximately 20% of the fibres) are similar to those reported for *Pinus radiata* TMP (Kibblewhite 1983). He indicated that 81% of the fibre cross sections were intact, thus leaving 19% as RF.

The increased fibrillation of TMP pulps relative to CTMP or CMP is possibly due not only to fibrillation of the exposed S_2 layer, but to the gradual conversion of some of the ribbons into long fibrils, as is the case for softwood mechanical pulps (Forgacs 1963). To a certain extent, the ribbons present in the pulp will expose the fibre S_3 layer, which should provide a good bonding surface because of its high carbohydrate content (Lange 1959).

The fact that radially-failed fibres appeared in small numbers only in CTMP and CMP pulps, and that these pulps were much stronger than those produced by the TMP process, indicates that increased fibre flexibility due to the chemical pretreatment is more important than potential

benefits which could arise from radial failure.

5.1.5. Breakdown of Tension Wood Fibres

The presence of TW was first noticed while observing the aspen refiner pulp slides. Structures that did not resemble normal wood fibres were identified. The presence of TW was then confirmed by examination of wood cross sections from the two aspen trees chipped. Also a third aspen wood sample was examined from the Peace River region in Alberta. They all contained noticeable amounts of tension wood. The birch samples did not show presence of tension wood.

Examination of wood cross sections could give indication of the extent of G-fibres in the chip furnish. However, such work could not provide accurate estimates of the G-fibre content unless all cells are counted in many slide sections across the diameter of the tree and at different heights. This would have been an enormous task and still it would not have provided accurate values to allow comparisons with values obtained from fibre cross sections in the refiner pulps produced. Accuracy of such estimates would have been low because some parts of the logs were removed before chipping due to staining or decay.

It was, therefore, decided to assess G-fibres from the unbeaten kraft pulp in which the G-layer would be intact inside the fibre. Due to the fibril orientation and high crystallinity, the G-layer gave strong polarization under cross polars when observed in pulp slides. However, it was

not possible to separate G-fibres on this basis for two reasons: 1) the substantial variation in G-layer thickness encountered (Figures 5.32 and 5.33), and 2) because of light polarization due to the S_2 layer.

Based on cross sections of kraft fibres, it was then possible to measure accurately the proportion of G-fibres in the aspen raw material used (Table 4.7 and Figure 3.4). An average of 31% of the total amount of fibres were G-fibres. Thus, this value was taken as being identical to that for the original wood which was chipped and then converted into refiner pulp.

The content of G-fibres in the R48 fraction of the aspen refiner pulps shown in Table 4.8 is illustrated in Figure 5.34. There was significant breakdown of the G-fibres only in TMP pulps as evidenced by their lower retention in the R48 fraction compared to the G-fibre content of the original furnish. Even when free G-layers were included, the total G-layer count was significantly below that of the wood. This indicates that there was a preferential breakdown of G-fibres under TMP processing conditions. Presumably, the thinner walls of the G-fibres (when excluding the G-layer), as reported by Kaeiser and Boyce (1965), promoted the formation of ribbons from the G-fibre walls ("skins"), thereby reducing the weight of the R48 fraction and, at the same time, exposing G-layers, as can be observed in Figures 5.35 and 5.36. This is consistent with earlier observations of abundant exposed G-layers in RMP pulps from *Populus*

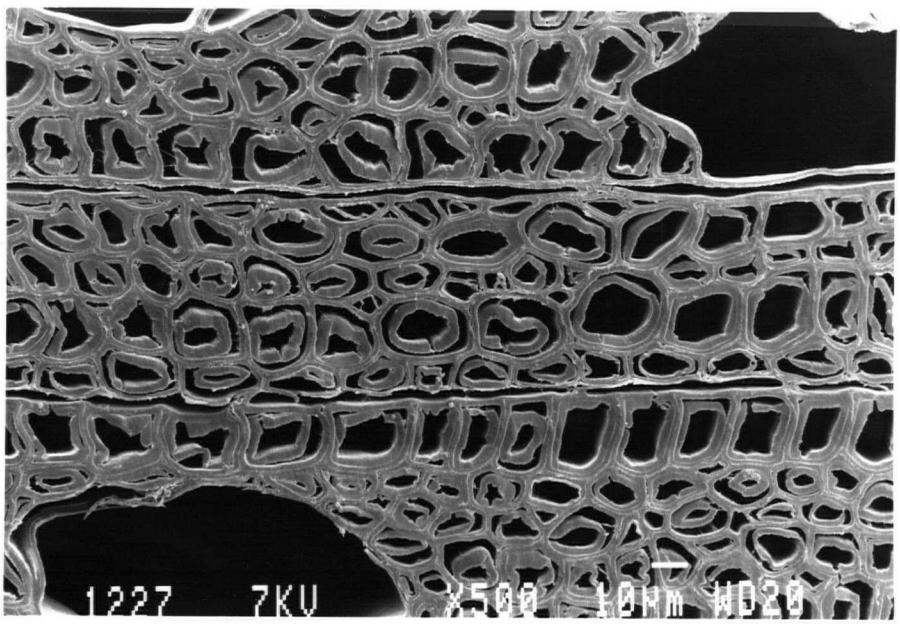


Figure 5.32. SEM photograph of G-fibres in aspen wood cross section showing thin G-layers.

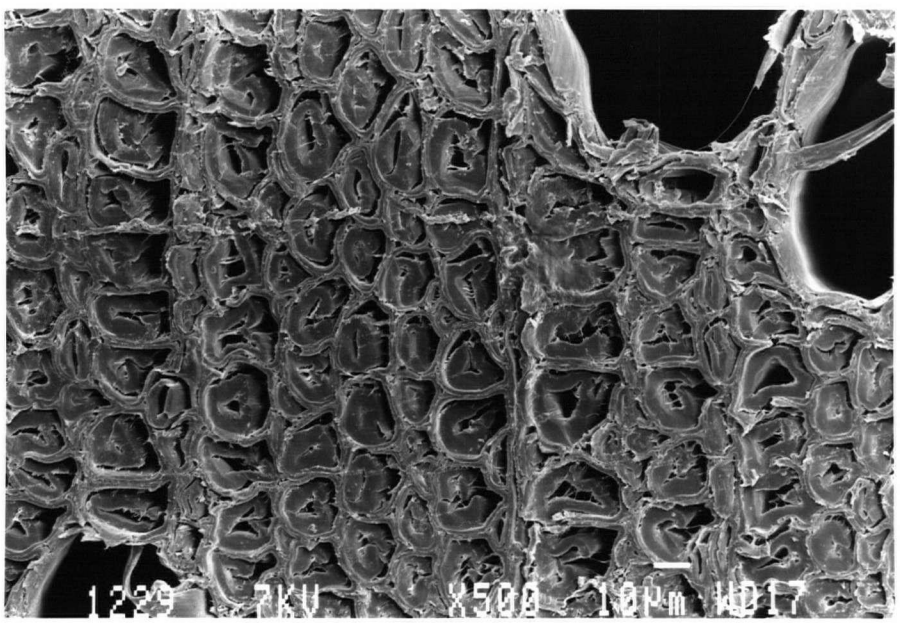


Figure 5.33. SEM photograph of G-fibres in aspen wood cross section showing thick G-layers.

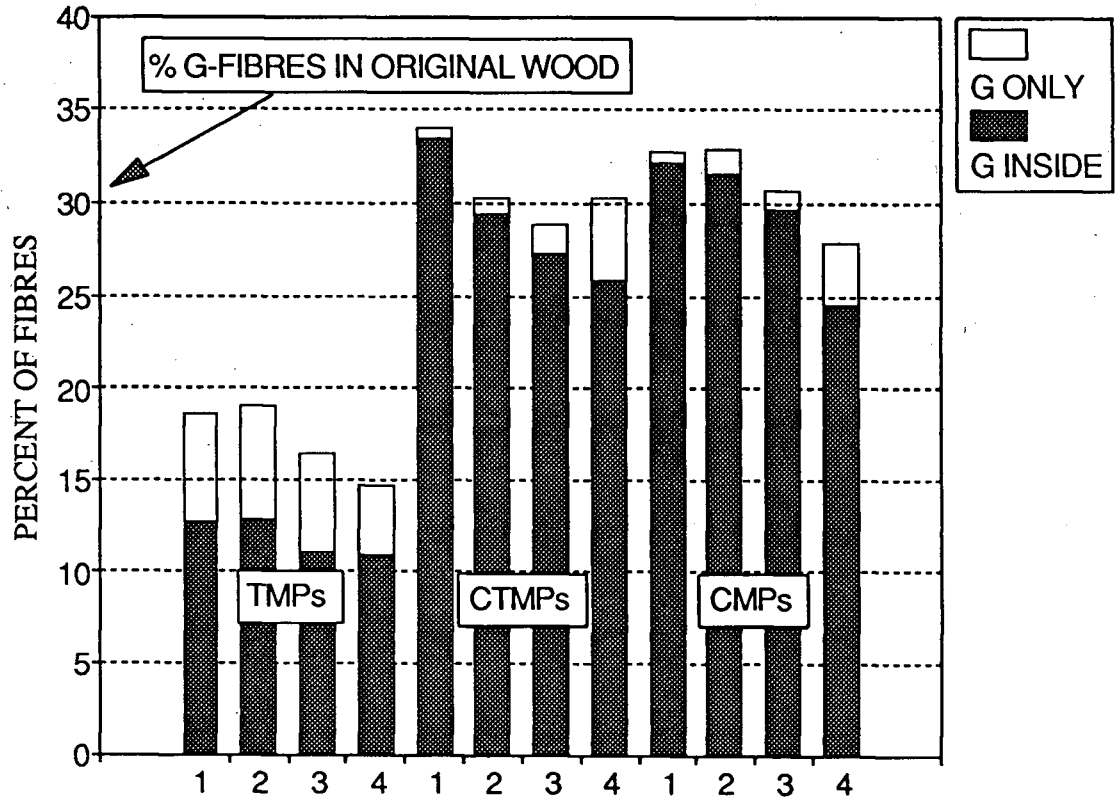


Figure 5.34. Percentage of G-fibres in the R48 fraction of aspen refiner pulps.

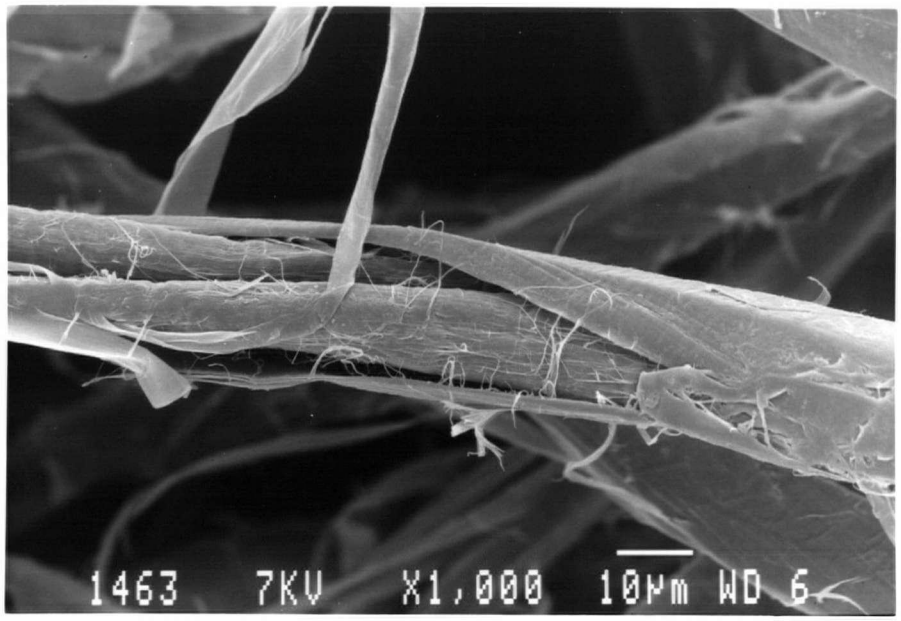


Figure 5.35. SEM photograph of G-fibre showing partial unravelling of the cell wall to expose the G-layer inside (pulp A-TMP4).

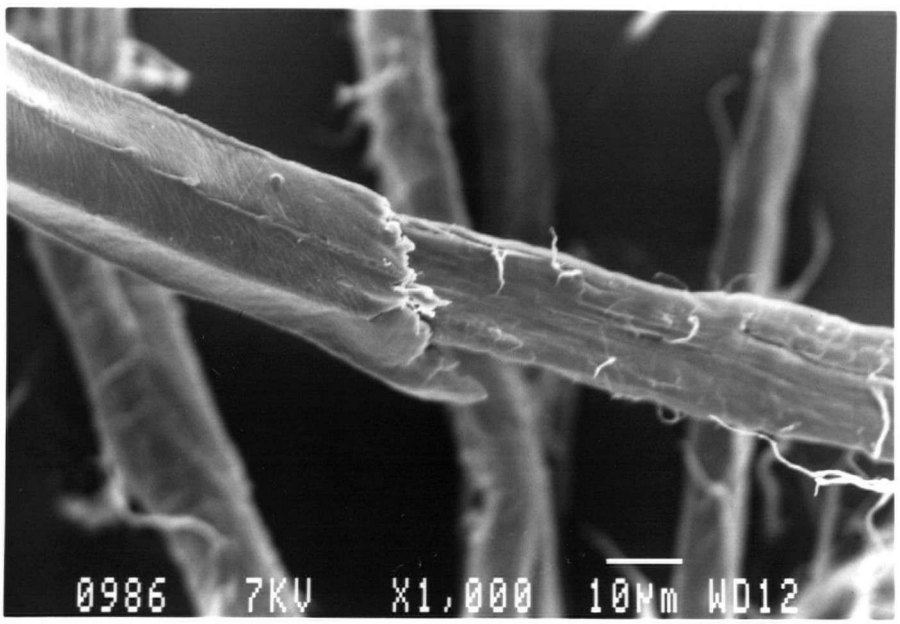


Figure 5.36. SEM photograph in which the fibre has been ripped, leaving an exposed G-layer (pulp A-TMP4).

deltoides (Scaramuzzi and Vecchi 1968). The breakdown of the free G-layers, although not quantitatively measured, can be monitored within the different fractions of the pulp, as seen in Figures 5.37 through 5.40. It can be seen that free G-layers are present in every fraction of the aspen TMP pulps. These photomicrographs show that the integrity of the G-layer is largely maintained. The G-layers do not appear to break down easily, as particularly evident from fibre cross section analyses which shows the presence of free G-layers in the R48 pulp fraction. It seems that the distribution of G-layer material in the different pulp fractions is set at the initial breakdown of the chips into pulp. On the other hand, due to reported weak lateral bonding (Cote and Day 1965, Norberg and Meier 1966), the G-layer would be expected to produce a certain extent of fibrillation during refining. Once the G-layers are initially liberated, the refining action should be able to produce some degree of delamination and fibrillation of this layer. Figures 5.38 and 5.40 support this hypothesis. They show the presence of purple filaments derived from G-layer material. It is expected that the exposed G-layer or any fibrillar material derived from it would contribute to increasing the interfibre bonding in a sheet of paper.

There was no preferential breakdown of G-fibres in CTMP or CMP pulps. The total amount of G-layers in the R48 pulp fraction was not significantly different from that of the parent wood. On the one hand, these two processes preserved

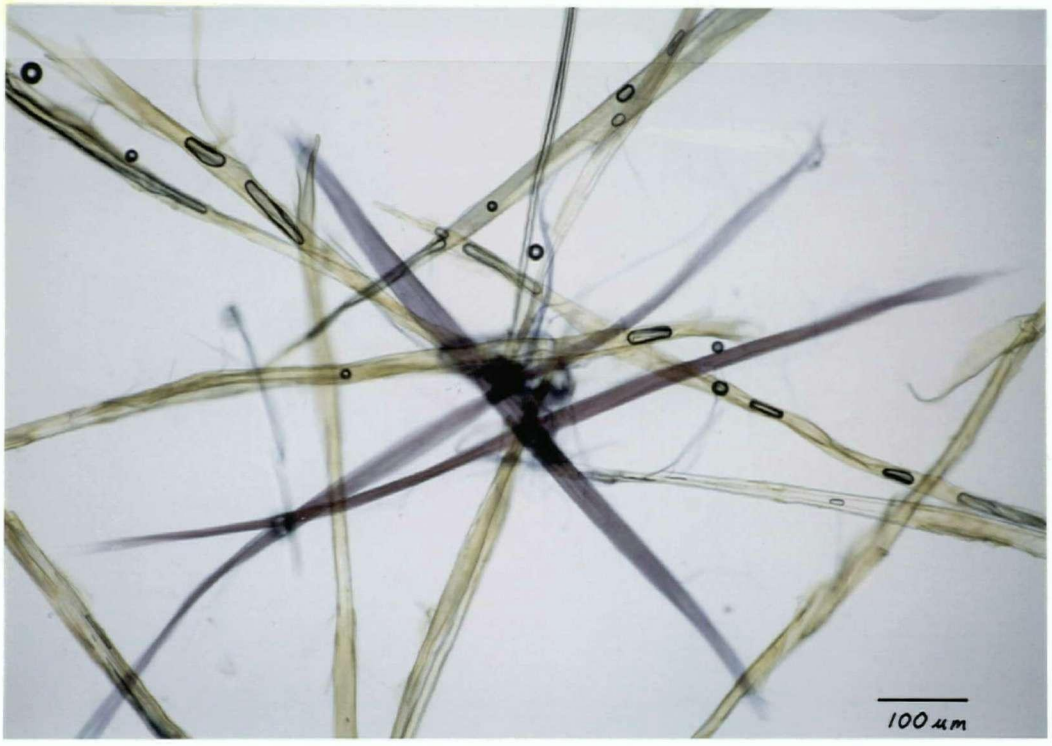


Figure 5.37. Presence of G-layers that were detached from their parent fibres in the R48 fraction of pulp A-TMP3.

Figure 5.38. Isolated G-layer showing fine cellulosic filaments detaching from the surface. Fraction 48/100 fraction of pulp A-TMP3.

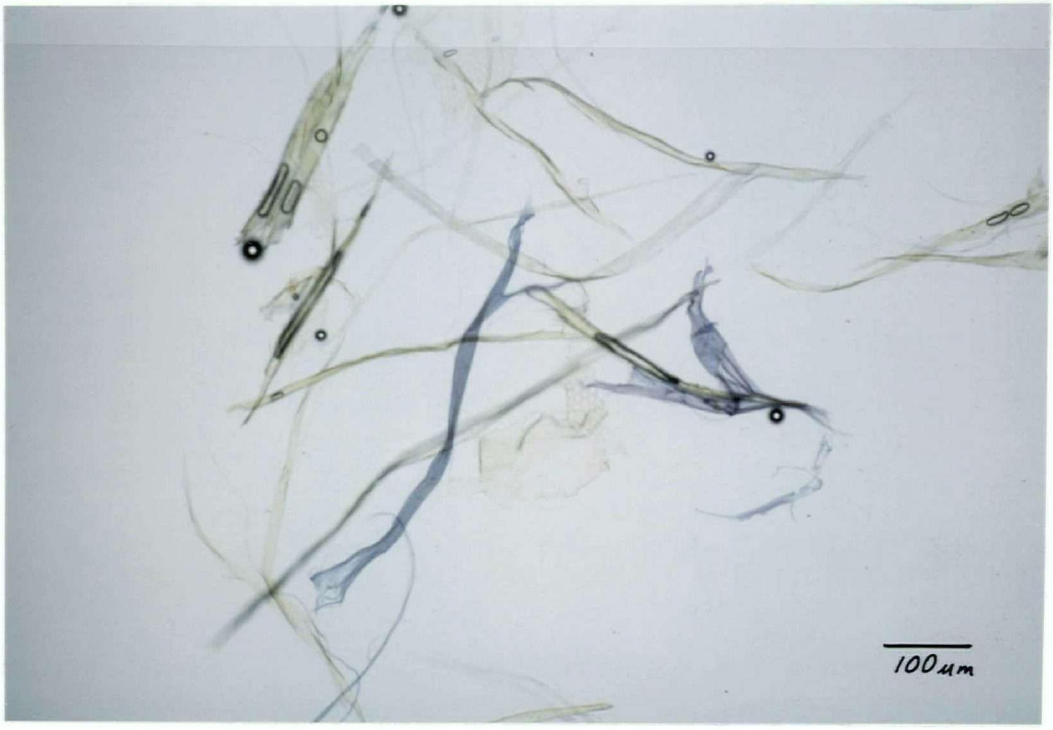


Figure 5.39. G-layer fragments in the 100/150 fraction of pulp A-TMP3.

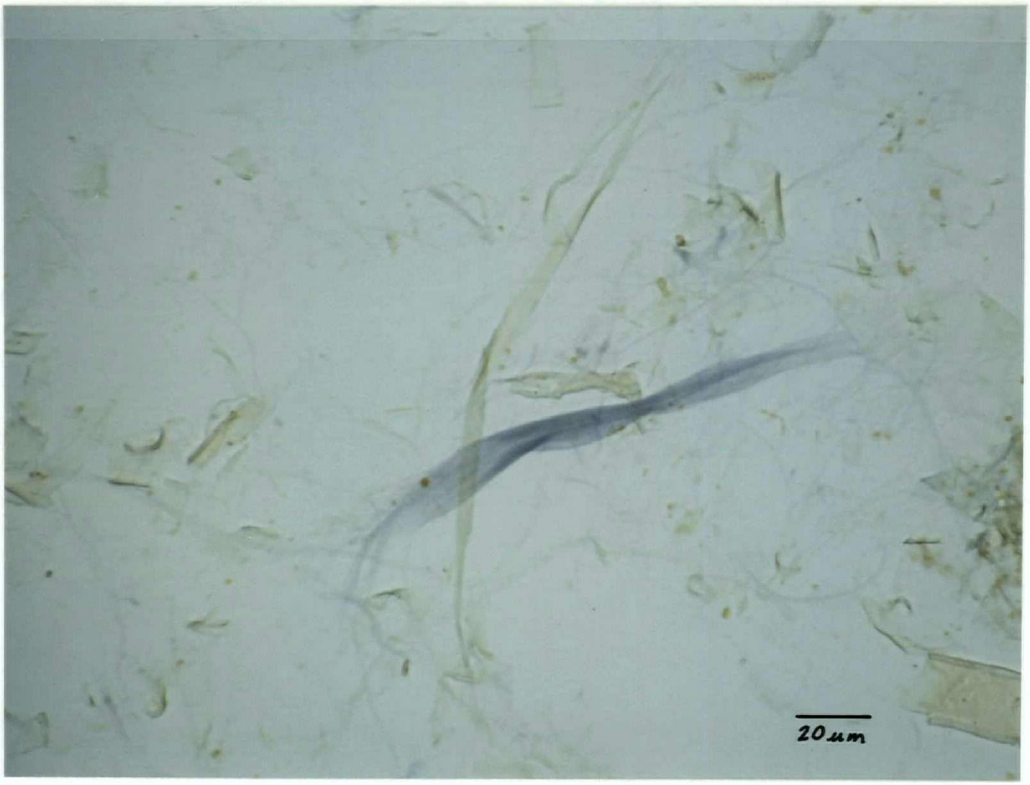


Figure 5.40. G-layer filaments in the P200 fraction of pulp A-TMP3.

fibre length to a much greater extent than did TMP processing, so that the chance of fibres breaking and liberating G-layers was small. On the other hand, only a small portion of the CMP or CTMP fibres failed radially, as measured on fibre cross sections, thereby minimizing the chance for G-layers to be stripped off after the fibres cracked and opened. It is inferred that, for CTMP and CMP pulps, the breakdown of the G-fibres followed a pattern similar to that for the rest of the fibres, at least initially from the R48 into the 48/100 fraction. Nevertheless, at low freeness levels, the presence of some G-fibre breakdown was evident since the value for free G-layers increased. Examples of these are shown in Figure 5.41 and 5.42.

An important consideration regarding the liberation of G-fibres concerns the "skins" that are left behind when this occurs. In TMP pulps these skins are likely to be a source of ribbons, whereas in CTMP these skins appeared as partly rolled and twisted fibres, or as free thin lamellae. Overall, the bonding potential of these skins should be high compared to normal fibres, based on the apparent flexibility and large surface area shown. Figure 5.41 shows examples of these skins.

The literature had indicated some beneficial effects of TW on the strength of pure mechanical pulps, but detrimental effects in chemical pulps. In TMP pulps, G-layers were not only liberated in large numbers, but they also may be

Figure 5.41. G-layer in aspen CTMP R48 fraction. Note the thin-walled skin left behind (pulp A-CTMP4).

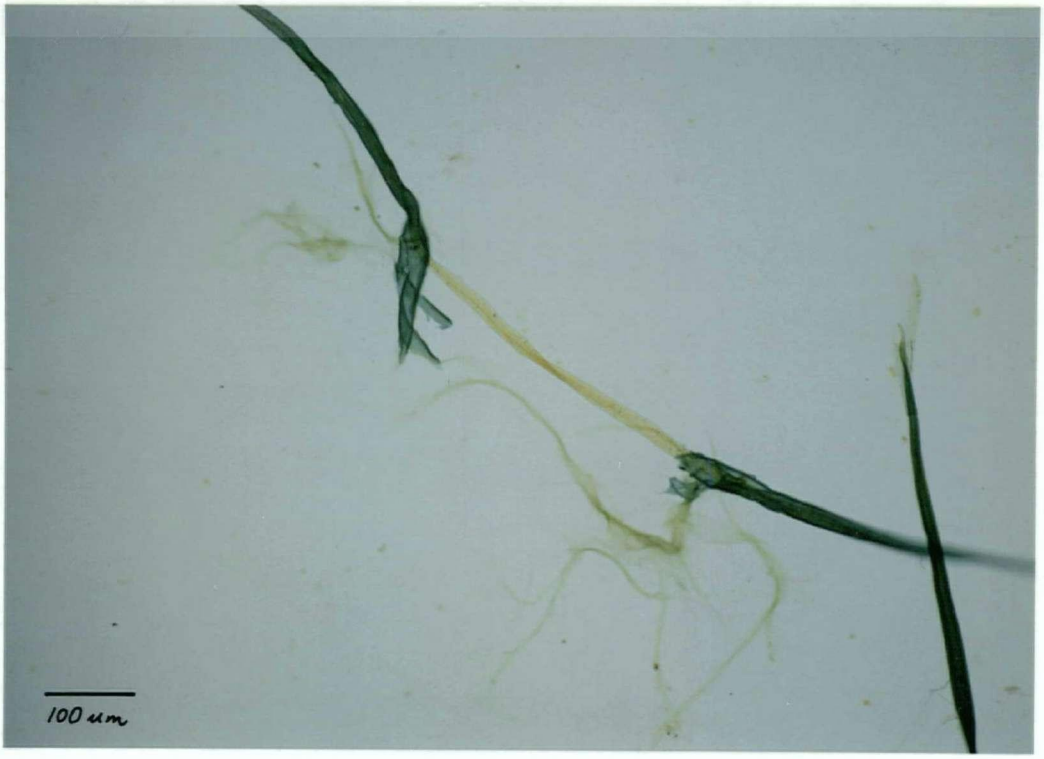


Figure 5.42. G-layer exposed at the centre of a fibre while covered by the fibre walls at the extremes (pulp A-CTMP4).

responsible for part of the formation of ribbons. The benefits from the presence of TW in TMP pulps seem to be clear. On the other hand, the effect of TW is unknown on CTMP and CMP pulp strengths and an investigation designed to study this may be needed. However, from the results obtained in the present study for the chemi-mechanical pulps, it would seem that the low proportion of liberated G-layers would produce fewer collapsed fibres, and thus bulkier sheets with lower strength if compared with pulps from normal wood.

The technique developed for assessing the proportion of tension wood fibres has excellent potential for use in studies in which the proportion of TW (measured in terms of G-fibres) is related to specific chemical components of the wood, wood behavior and pulp properties. It is a relatively simple method that provides accurate assessment of the proportion of G-fibres in a wood sample.

5.2. Breakdown of Vessel Elements

Whole VE are presumably the ones with higher picking tendency during printing. Therefore, it was considered important to assess their survival during refining. The study of the breakdown of VE was done by measuring the size distribution of VE fragments in the pulp and by counting, in each pulp, the number of whole VE that survived refining. Because a minimum size value had to be set in order to positively identify VE fragments, the size frequency

distribution did not provide information on the production of particles smaller than 112.5 um. With this information alone, no conclusion could be drawn on the number of whole VE that were reduced to fragments of different sizes.

The information provided by the counting technique used to assess survival of whole VE, complemented the findings on particle size distribution. For example, if two pulps show no difference in VE size distribution, but do show different numbers of whole VE per gram of wood (or of pulp if there is no yield difference), it can then be concluded that the pulp with fewer whole VE has more fine particles (smaller than the minimum set value) that originated from the breakdown of VE that did not survive the refining process.

The results presented in Tables 4.13 and 4.14 show that chip refining under TMP conditions effectively reduced the size of the VE to fine particles for both species. Figures 5.43 and 5.44 illustrate particle size distribution for different pulps of similar freeness produced under the different refiner pulping processes studied here. It should be mentioned that the frequency polygon was closed at zero in the lower end, because of simplicity and clarity. It is anticipated however, that the frequency values would increase with decreasing particle size.

In TMP pulps, about 90% of the VE fragments measured were smaller than 250 um and about half of them were smaller than 150 um. This is the size of the openings in a 100 mesh screen of a Bauer-McNett Classifier. A large proportion of

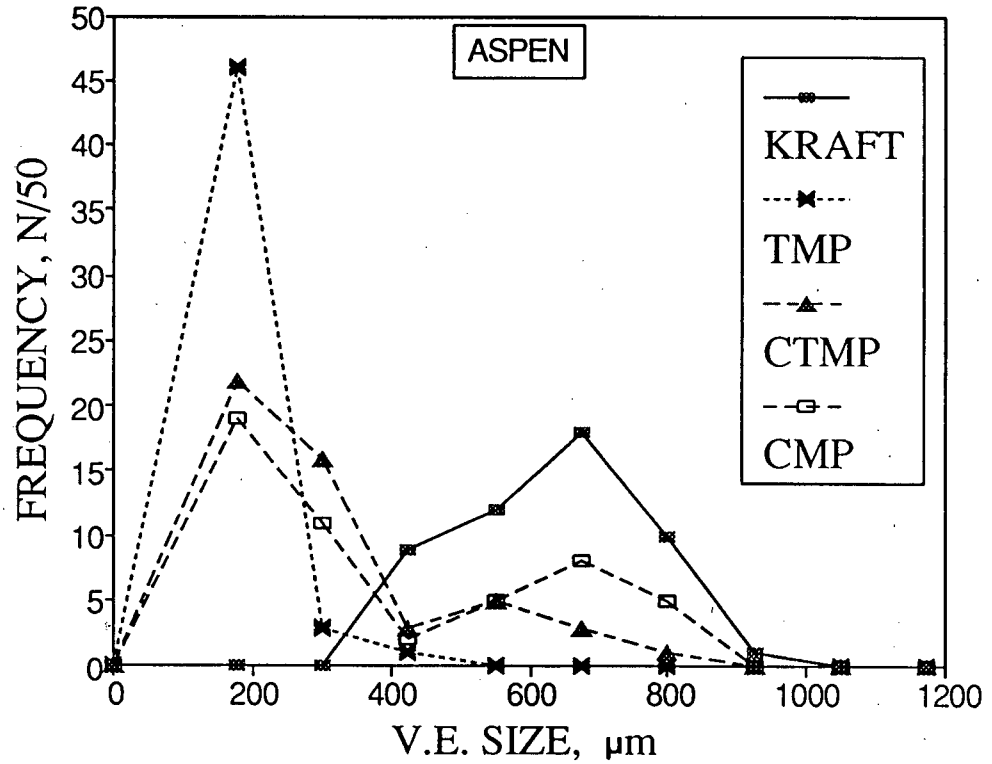


Figure 5.43. Frequency polygons for VE size in aspen refiner pulps of similar freeness.

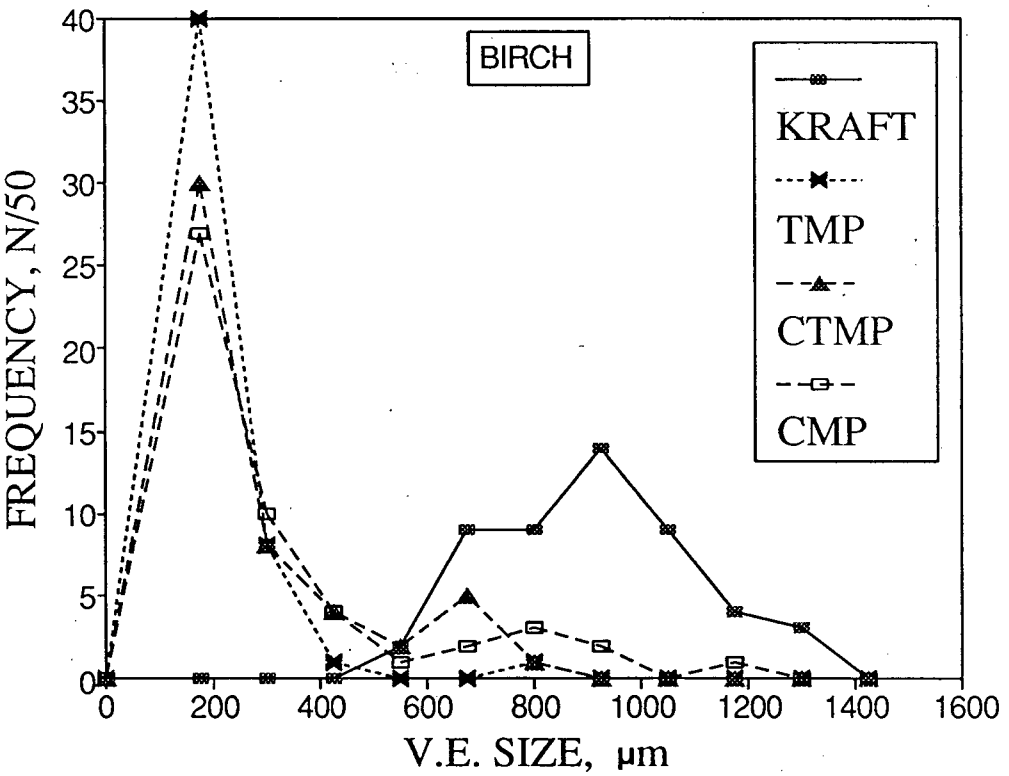


Figure 5.44. Frequency polygons for VE size in birch refiner pulps of similar freeness.

small VE particles was also present in CTMP and CMP pulps as the majority of the fragments accumulated in the smaller size categories. Thus, it is not surprising that VE are said to be reduced to fine material during refining, and found mostly in the 100/200 fraction (Giertz 1977). However, in the P200 fraction (sieve opening of 74 um), VE fragments were also abundant for these pulps. It can be concluded, therefore, that small fragments of VE are present in large numbers in the fine pulp fractions (100/150, 150/200 and P200) regardless of the refiner process used. Their abundance by weight, however, will depend not only on the VE size and frequency in the original wood, but also on their relative survival upon refining, and the amount of fines produced by other wood elements.

Although the experimental ranges of freeness and refining energy values were wide in TMP pulps, no difference in the VE size distribution of these pulps was found due to refining, regardless of species (parts B and C of Table 4.15). Also, the number of whole VE in these pulps was almost non-existent (only high freeness aspen TMP showed a minuscule proportion of whole VE). This information, from Table 4.16, is illustrated in Figures 5.45 and 5.46. It is obvious that refining under TMP conditions was effective in destroying whole VE and converting them into fine particles, even when those pulps were of a high freeness. In fact there were virtually no VE in the R48 fractions of any TMP pulp. It was the initial breakdown of the wood chips that caused

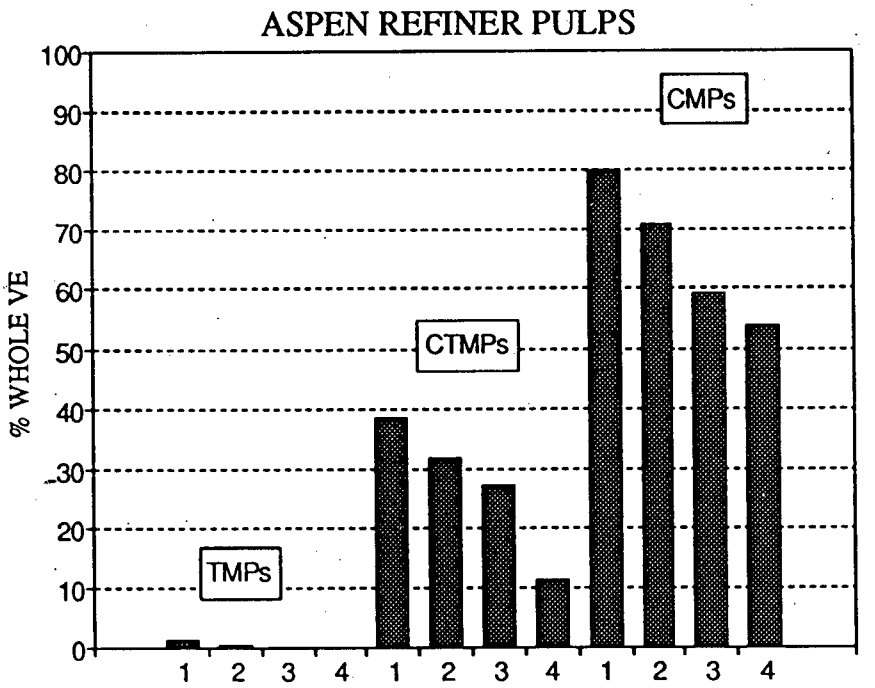


Figure 5.45. Percentage of whole VE that survived during the refining of aspen chips.

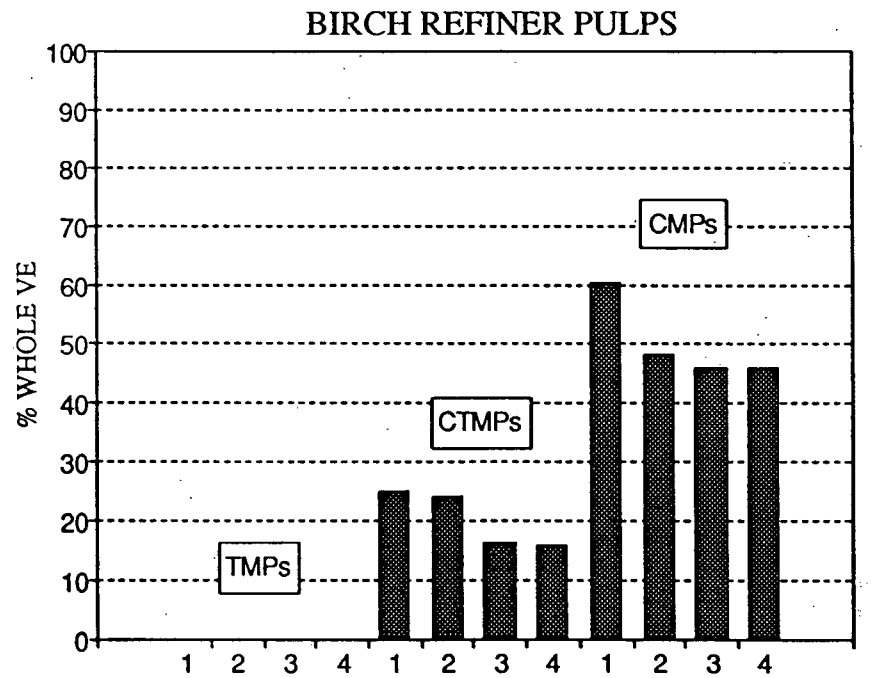


Figure 5.46. Percentage of whole VE that survived during the refining of birch chips.

the VE fragmentation. Further refining may have reduced these fragments into even smaller particles, but only after the wood VE had already been broken down. The fact that almost no overlapping is seen between the TMP and kraft pulp curves (Figures 5.43 and 5.44), confirms this finding.

In spite of the fact that there was a significant difference in the original size of vessel elements (Table 4.12), with average lengths of 625 and 904 um for aspen and birch, respectively, no difference was found between the VE size distribution of their TMP pulps. The VE breakdown under TMP conditions was so effective that even the larger VE of birch were reduced to a size distribution pattern similar to that for the aspen TMP pulps. Moreover, birch VE appeared to break down even more readily than the aspen VE as no whole VE were found in any of the birch TMP pulps. The generally higher tendency of birch VE to break down compared to aspen will be discussed later in this section.

Presumably, the wood defibrated under TMP conditions followed the initial pattern of failure through VE as indicated by Ohsawa and Yoneda (1978) for high shear conditions, i.e., defibration producing transwall failure in cells of low Runkel Ratio (double cell wall thickness over lumen diameter). This condition holds for VE, even after water saturation of wood and temperatures up to 170 ^{o}C. Furthermore, the lignin in the VE cell walls, rich in guaiacyl units (Fergus and Goring 1970, Hardell *et al* 1980), i.e., more cross-linked and less heat sensitive than the

syringyl lignin in the fibre secondary wall, contributes to the stiffness of VE. The low flexibility of VE seems to be responsible for their destruction during refining.

Significantly different results were obtained in terms of VE breakdown when chemically-treated chips were refined. A greater number of large VE fragments could be found in these pulps compared to the TMP pulps. Results in Table 4.15 (parts D and E) showed a large difference between values of Chi-square calculated and Chi-square critical when comparisons were made between VE size distributions of TMP and either CTMP or CMP pulps. This is also shown graphically in Figures 5.43 and 5.44., wherein the curves of VE size distribution of CTMP and CMP pulps overlap with those of kraft pulps of the same species. This overlapping indicates the preservation of some whole VE in these two refiner pulps.

As in the case of TMP, neither CTMP nor CMP VE size distribution patterns showed a dependency on refining energy, when comparisons were made within a species (parts B and C in Table 4.15). Again, the distribution of VE fragment sizes was set at the initial breakdown of wood into pulp for a given process and species, and further refining did not change these patterns.

For a given species, the difference in VE distribution patterns (Chi-square values) was rather small when CTMP and CMP pulps were compared (parts D and E in Table 4.15), although still significant in the case of aspen. The

relatively small differences between refiner pulps from chemically-treated wood, compared to the large difference of these compared against TMP pulps, indicate that wood softening was responsible for the preservation of a larger proportion of whole VE in CTMP and CMP pulps. Yet the distribution of VE size did not vary much when the severity of the treatment increased, since the initial breakdown patterns of VE were similar by the CTMP and CMP processes. The significant difference reported in VE size distribution between aspen CTMP and CMP may be in part due to the cutting effect as indicated for the pulp A-CTMP4 on one hand, and also to the production of slightly more fine particles in CTMP and less elements of large size than in CMP on the other. It is clear, however, that both CTMP and CMP pulps, for both birch and aspen, preserved to some extent the integrity of whole VE. Thereby, these pulps may be more prone to VE picking. Reduction of large VE would require more extensive refining.

It is interesting to note that in both CTMP and CMP curves (Figures 5.43 and 5.44) there were essentially two peaks. The first peak appeared at the lower end, as mentioned earlier, to close the polygon. But the second peak, particularly for aspen, followed the distribution pattern of the whole VE size set by the kraft curve. Breakdown appeared to take place after a number of VE had been initially fragmented, and further fragmentation proceeded from these fragments. On the other hand, a number of VE remained as

whole entities in the pulp. The results in Table 4.16 and Figures 5.45 and 5.46 provide information on the number of VE that did not break down during refining. It is primarily here where the difference between CTMP and CMP pulps (and also between species) becomes apparent. In fact, these figures also show the effect of refining on the reduction of whole VE for a given process.

It is clear that the size distribution alone could not provide complete information on the extent of VE breakdown. Such graphs reveal only the particle size distribution, but suggest nothing about the number of VE that survived refining. Similar VE size distributions, with a disparity in whole VE content, indicates that an increased number of VE are broken down into very fine particles, which are actually smaller than the minimum value measured. Regardless of species, CMP pulping obviously preserved more whole VE than did the CTMP process. The higher chemical application in CMP pulping, combined with higher cooking temperatures and longer cooking times, produced whole VE in amounts that, for high freeness CMP pulps, were closer to those of kraft than to TMP pulps. For CTMP pulps, the amount of whole VE lays somewhere between the TMP and CMP values. This shows that the breakdown of VE, as in the case of fibre breakdown, depended on the severity of the chemical treatment before mechanical defibration. For instance, kraft pulps preserve the length of all fibres and those of VE. Mechanical defibration is only necessary for linerboard-grade pulps.

CMP pulps have larger amounts of long fibre fractions than CTMP which in turn, are larger than those of TMP pulps. The same pattern applies to VE. Obviously, however, VE are less preserved than fibres due to their thinner walls, larger diameters and higher rigidity. Although no information is available on the peeling of surface layers of VE during refining, it is presumed that peeling would be unlikely as most damage will be in the form of fragmentation of the VE. When comparisons of VE size distributions were made between species, no differences were found for TMP or CMP pulps (part F in Table 4.15). Only in CTMP pulps was a significant difference found, which could be in part attributed to the cutting effect in the low freeness aspen CTMP pulps. The most important difference between species, however, was in the number of whole VE that survived refining (Figures 5.45 and 5.46). Birch VE were more readily reduced to fragments than were those from aspen. Although VE cell wall thickness is reported to be higher in birch (1.8-2.3 um) than in aspen (1.3-1.6 um) according to Musha and Goring (1975), the difference in intervessel pitting appears to be the cause of size reduction in birch VE. The fibril angle of VE walls follow the direction of the slit-like apertures of the bordered pits in the vessel wall (Harada 1965). On one hand, birch intervessel pits are minute (2-4 um), with coalescent apertures, i.e., orifices frequently confluent (Panshin and de Zeeuw 1980). These slit-like inner apertures in many cases unite several pits to form spiral grooves that, upon

refining, facilitate the unraveling of the VE (Figure 5.47 and 5.48). On the other hand, aspen has large intervessel pits (8-12 um) and the separation between pit apertures did not lead to unraveling of the VE. Instead, aspen VE were broken down in a more random fashion. This resulted in aspen refiner pulps having a higher proportion of VE survival from the original wood.

Aspen has more VE in wood than does birch (Panshin and de Zeeuw 1980). Also, the percentage of whole VE survival upon refining is higher for aspen. The large difference in VE content between these two species is accentuated during refining. It would be reasonable to expect that aspen refiner pulps would have a higher tendency to vessel picking than birch pulps, not only because of more whole VE, but also because of the substantial difference in fibre length between them. Due to the longer fibres, felting in birch pulps is more extensive, and the fibres would hold VE from being picked during printing. On the other hand, aspen fibres are more collapsible, and the number of fibres per gram of pulp to hold VE during printing is also larger than in birch (Giertz 1977). Thus, it is not easy to clearly anticipate the relative effects of these two species in vessel picking.

The effects of chemical and mechanical treatment are significant in the survival of VE. The more severe the chemical pretreatment, the higher the survival rate of VE. Following this, additional refining causes further breakdown

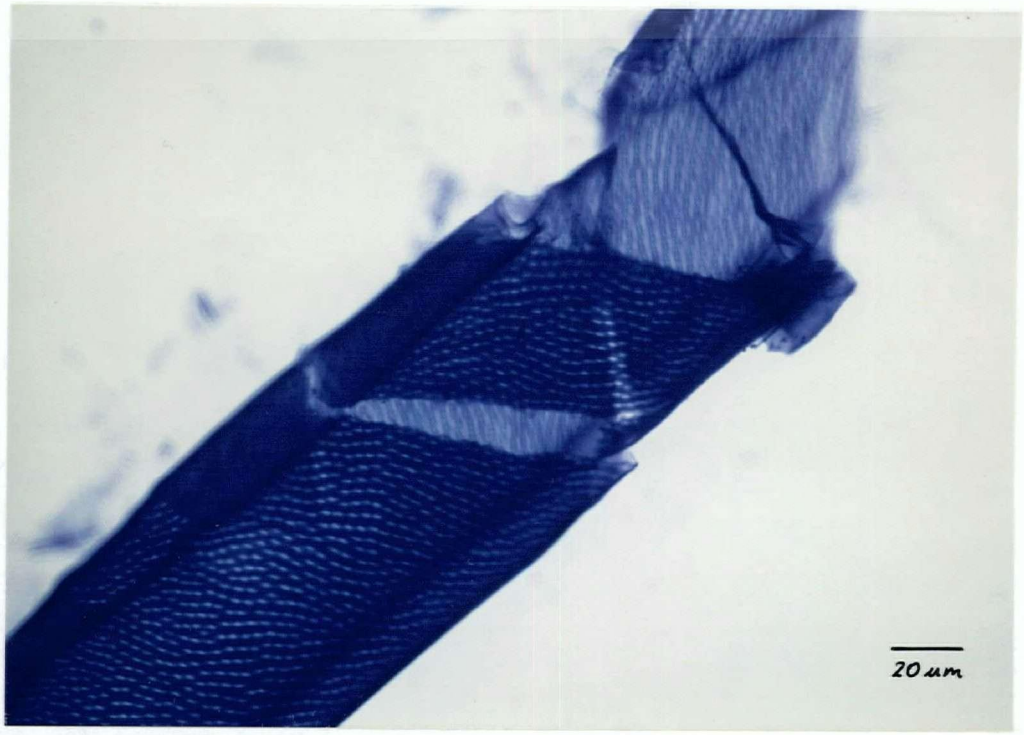


Figure 5.47. Birch VE fragment showing splitting along the line of intervessel pitting (pulp B-CTMP3).

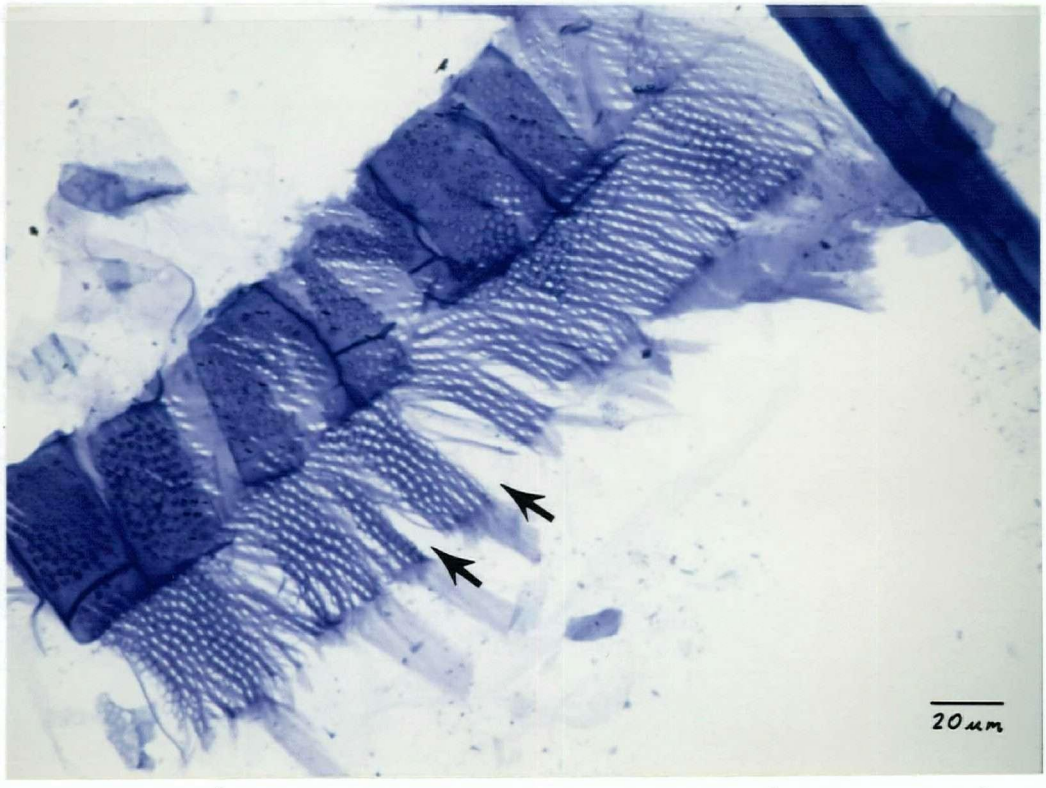


Figure 5.48. Single-walled VE fragment showing separation in the direction of intervessel pitting (pulp B-CTMP3).

of VE. However, as observed before, the starting point was set by the conditions of the chemical pretreatment applied to the chips. Nevertheless, it is difficult again to predict the relative effects of the pretreatment conditions on the VE picking tendency during printing. Although more severe treatment conditions resulted in survival of more whole VE, the parent fibres (and presumably also these VE) are more flexible and conformable, and are thus able to hold VE better.

5.3. Pulp Properties

The properties obtained for the refiner pulps produced in this study are in general agreement with those published in the literature for these species. Nevertheless, several points need to be stressed.

There were considerable differences in the optical properties of the pulps. TMP pulps from both species showed the typical behavior for mechanical pulps, and the light scattering coefficient increased with increased refining. Thus, higher density values for TMP sheets were accompanied by larger unbonded areas per unit gram. In CTMP and CMP pulps the effect was less pronounced, so much so that only aspen CTMP showed an increase in light scattering coefficient upon refining due, possibly, to fibre size reduction in these pulps. The other pulps produced from chemically-treated chips showed essentially no change, indicating their chemimechanical nature.

Brightness values were much higher for TMP pulps. The original high brightness of aspen wood provided higher brightness values than those found for birch pulps. The alkaline chip pretreatments in CTMP and CMP pulp manufacturing resulted, as expected, in lower brightness values for both pulps. The darkening effect of the alkali in the treatment was pronounced and the simultaneous addition of sulphite applied did little to reduce the brightness loss. Printing opacity, on the other hand, was high in TMP pulps due to the high scattering coefficient, and also high in CMP pulps due to their low brightness which, in turn, was caused by high light absorption coefficient values.

The size distribution of pulp fractions was essentially set at the initial breakdown of chips into pulp. Further refining only changed this marginally. Thus, in order to modify this distribution in favor of a larger long-fibre fraction, chemical treatment of the chips is required. This treatment also resulted in more flexible fibres as evidenced by the high sheet density values observed. Thereby, the pulp strength properties improved dramatically from TMP to CTMP or CMP pulps. Consequently, the degree of interfibre bonding can be attributable both to chemical pretreatment as well as to the degree of refining, with certain limitations. This is illustrated in Figure 5.49. It shows the strong influence of sheet density on tensile strength. As shown in this figure, the level of sheet density was again set by the type of treatment. Further refining increases sheet density. TMP

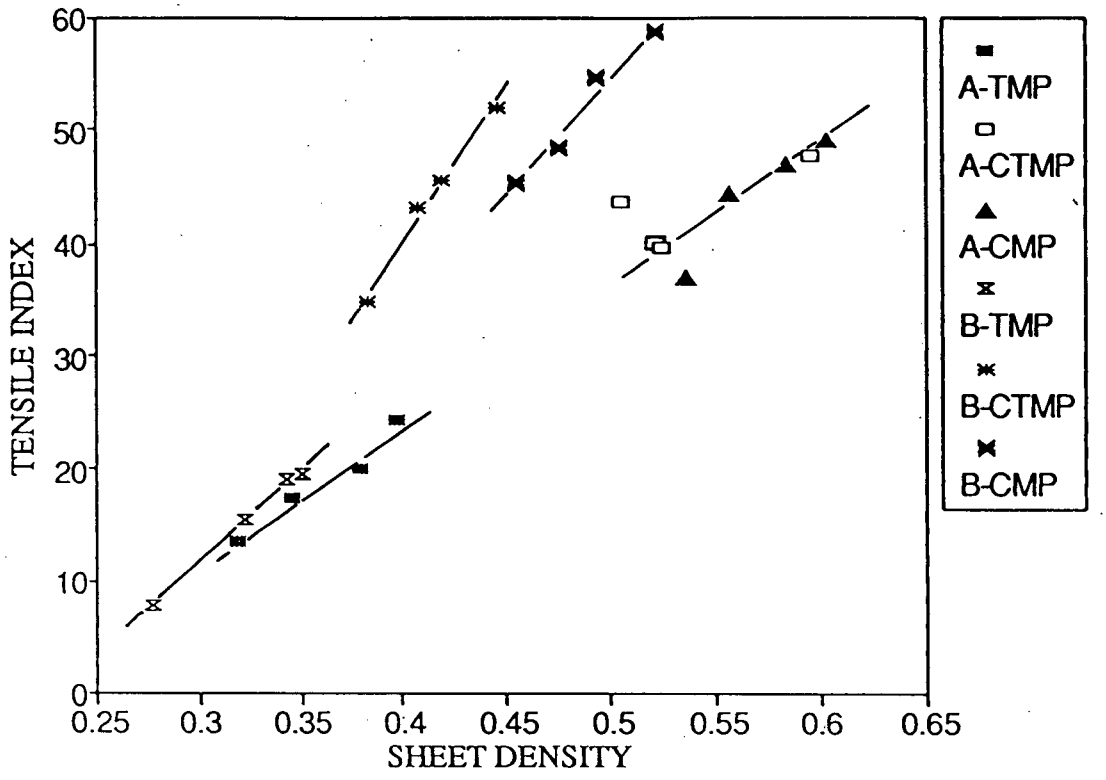


Figure 5.49. Relationship between tensile strength and sheet density for all hardwood refiner pulps studied.

pulps had very low sheet densities for both species. Aspen and birch TMP pulps also had similar Bauer-McNett fractionation patterns (Figure 5.1), so that effects due to fibre length appear to be unimportant. Rather, the levels of sheet density and, consequently, of tensile strength were slightly higher for aspen TMP pulps. This is presumably due to the lower Runkel Ratio of aspen fibres which may render

them more collapsible.

There was a drastic change in sheet density when pulps were produced from chemically-treated chips. In aspen, CTMP and CMP pulps followed the same line initiated by the TMP pulps. However, there seemed to be little difference between aspen CTMP and CMP in terms of either sheet density or tensile strength.

The response to chemical pretreatment was more evident in birch. Although sheet density did not attain levels comparable to aspen, the shift to higher tensile strength values was substantial. The influence of a much larger long-fibre fraction is suspected as the cause for this change, but it could also be due to improved fines quality. The difference between birch CTMP and CMP pulps shown in Figure 5.49 could also be attributed to a larger R48 fraction in CMP pulps. However, it is also seen that the CMP pulps had higher sheet densities than those of the CTMP pulps at equivalent levels of tensile strength . On the other hand, CTMP pulps required lower freeness, i.e., probably a higher production of fines, to achieve the same tensile strength of CMP pulps. Thus, it can be seen that a certain level of interfibre bonding can be obtained by either applying severe chemical pretreatment followed by refining to a high freeness level (as in CMP), or by using milder treatments followed by refining to a lower freeness level (as in CTMP). The difference in pulp tensile strength of birch, relative

to aspen at similar sheet density levels, for pulps from chemically-treated chips, may also be due to superior levels of the long-fibre fraction in birch. It is clear, however, that high wood density was not a limitation in this case, since birch, with much higher wood density levels (0.55 compared to 0.45 for aspen), rendered CTMP and CMP pulps of higher strength than aspen.

5.4. Significance of Findings

Despite the fact that the S_2 layer was found to be more exposed in TMP pulps than in their CTMP or CMP counterparts, TMP pulp strengths were much lower. The removal of the S_1 layer was not sufficient to produce fibre flexibility levels (as indicated by sheet density values) comparable to those obtained by the application of chemical treatments to the chips prior to refining. Thus, the fibres with their S_2 surfaces exposed are not capable of taking advantage of their higher bonding capacity unless the area of contact between them is increased. Figure 5.50 shows a plot of tensile index against the level of S_2 exposure for the various pulps. It is evident that interfibre bonding did not follow the measure of quality of fibre surface. In fact, birch TMP pulps, with fibres of the highest S_2e index, produced the weakest pulps. Thus, TMP pulps even with their high production of ribbons of good bonding ability and, in

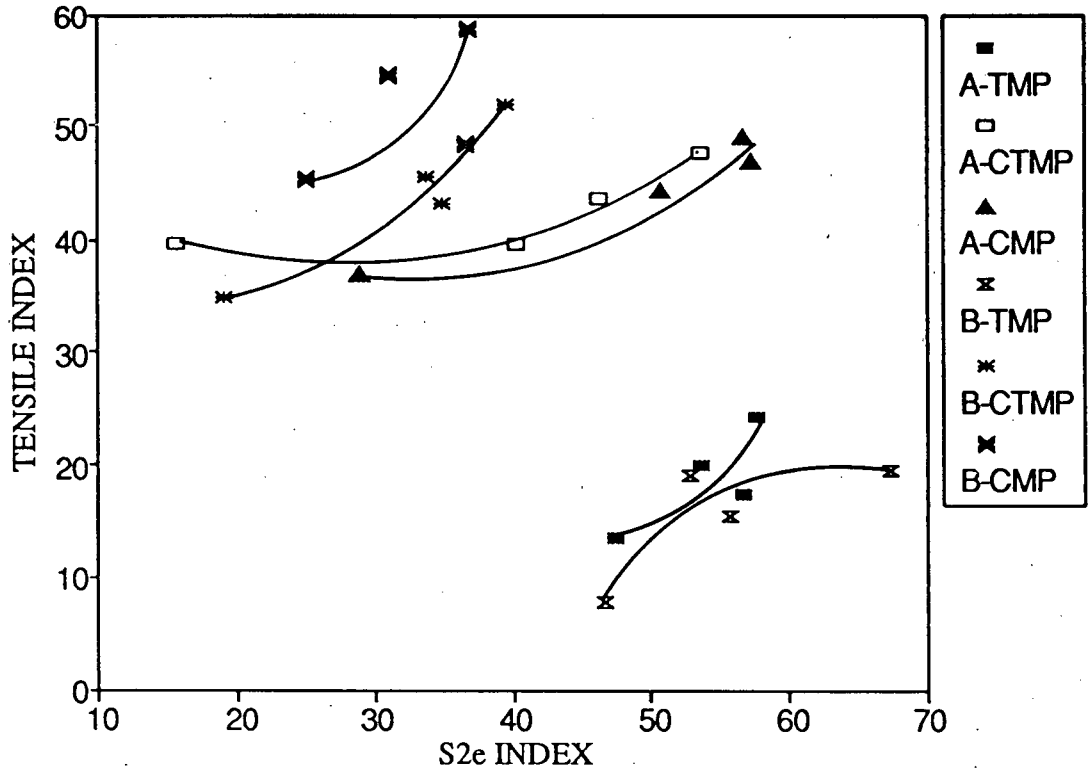


Figure 5.50. Relantionship between tensile index and degree of exposure of the S_2 layer for all hardwood refiner pulps studied.

the case of aspen, with the presence of liberated highly cellulosic material from G-layers, could not compete in strength with CTMP or CMP pulps. Figures 5.51 to 5.54 show the difference in surface appearance of handsheets made from aspen TMP and CTMP pulps. Even at low freeness levels the relative fibre rigidity can be appreciable in TMP pulps. Despite the large amount of TMP fines produced, it seems as if their bonding role in acting as bridges between fibres is limited in sharp contrast to the behavior in CTMP

Figure 5.51. SEM photograph of a handsheet surface for pulp A-TMP1.

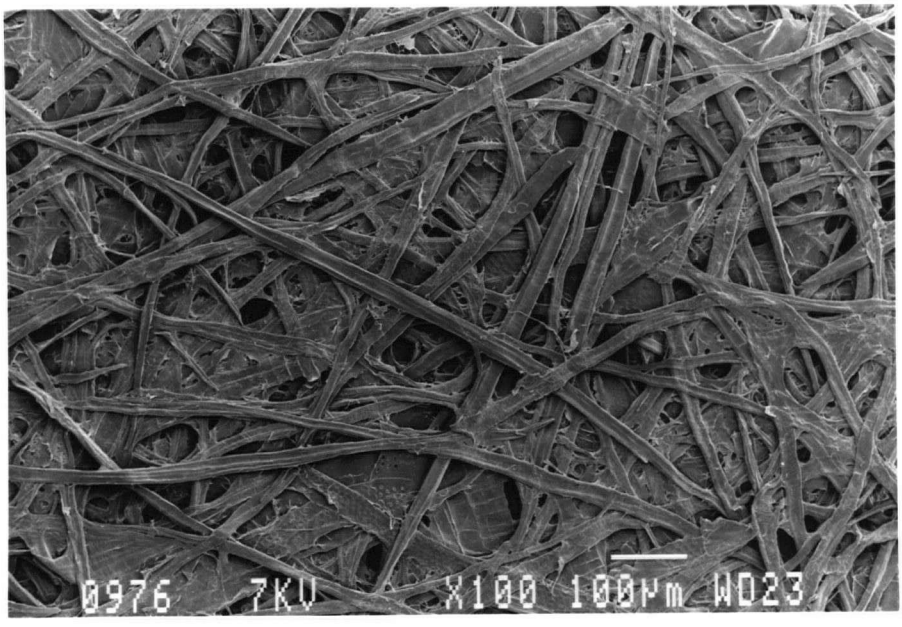


Figure 5.52. SEM photograph of a handsheet surface for pulp A-CTMP1.

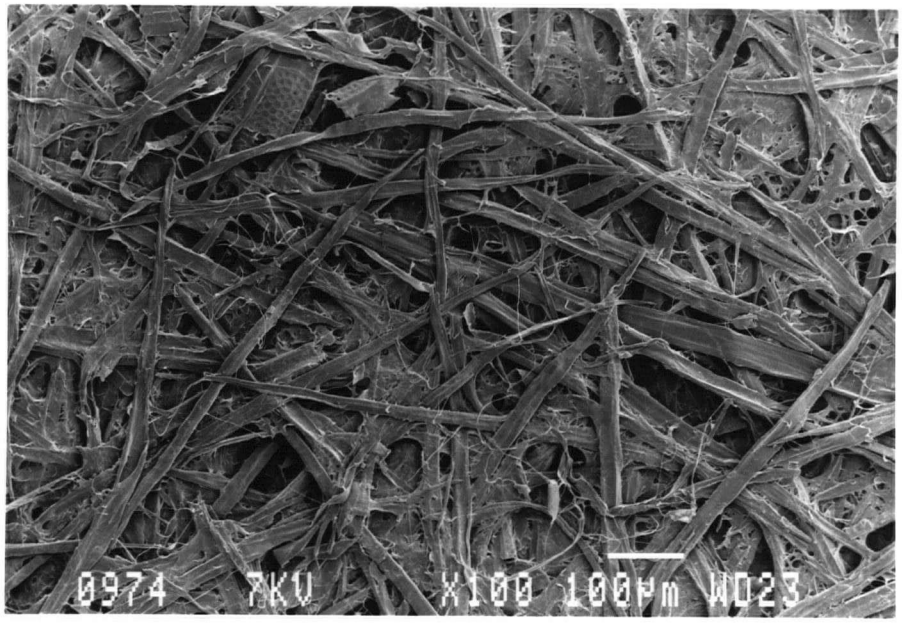


Figure 5.53. SEM photograph of a handsheet surface for pulp A-TMP4.

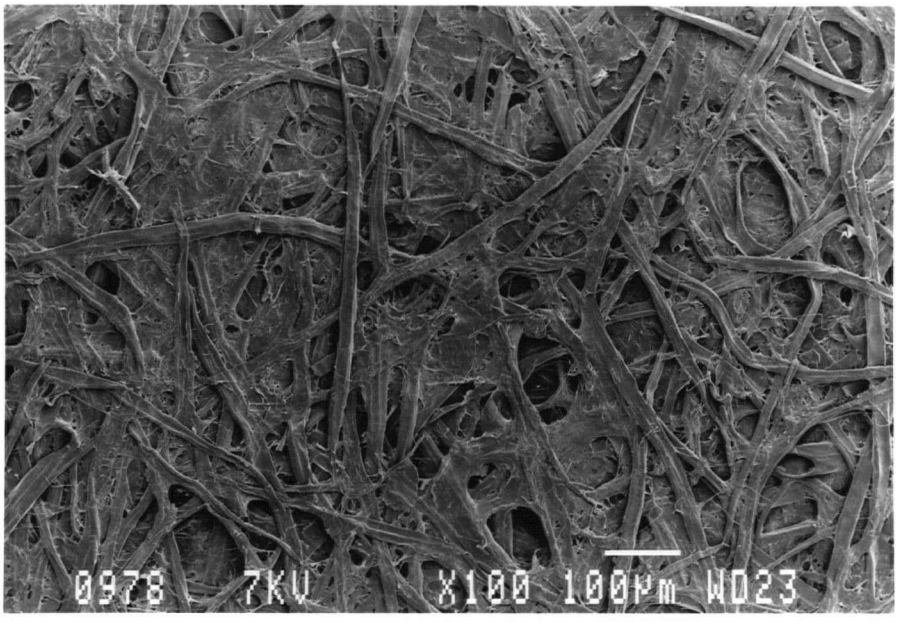


Figure 5.54. SEM photograph of a handsheet surface for pulp A-CTMP4.

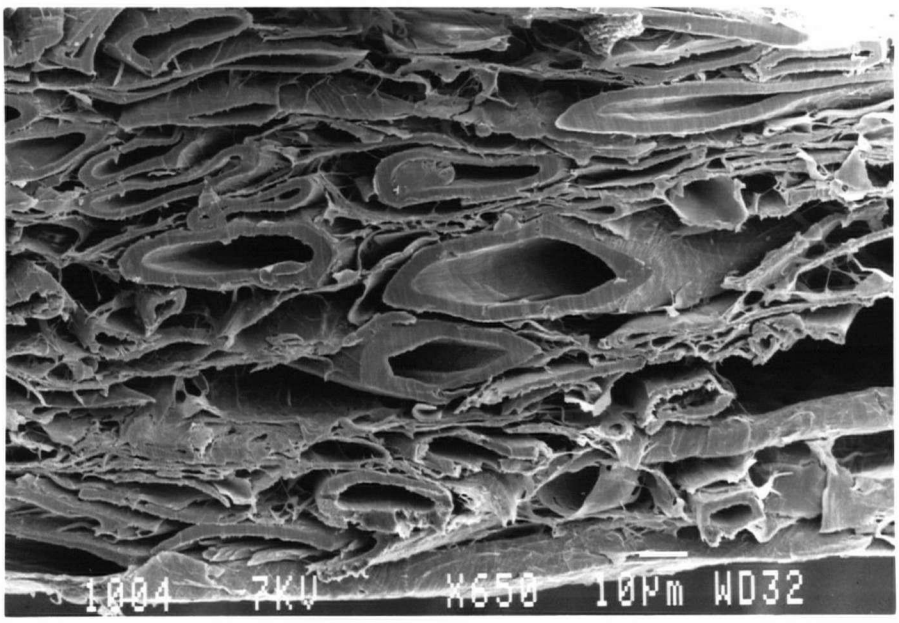


Figure 5.55. SEM photograph of handsheet in cross section of pulp A-TMP4.

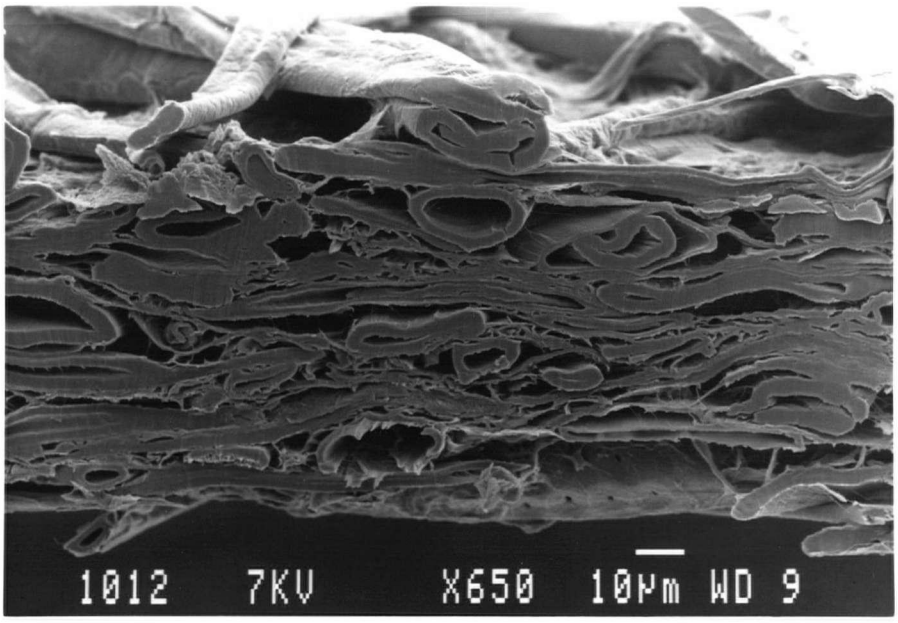


Figure 5.56. SEM photograph of handsheet in cross section of pulp A-CTMP4. Note the difference in fibre collapsibility and bulk compared to TMP.

pulps. This is also illustrated in cross sections of handsheets (Figures 5.55 and 5.56). These photographs show a lack of consolidation in the sheet from TMP fibres as opposed to the more flexible and well collapsed fibres in CTMP sheets. It is concluded that fibre flexibility, as measured by sheet density, takes precedence over any benefit provided by fibre surfaces of high bonding potentials.

The importance of the fines fraction of mechanical pulps has been widely recognized in the literature. It has been said that the main cause for the low strength of hardwood mechanical pulp is due to the poor quality of fines produced (almost completely lacking in fibrils), and that chemical pretreatment improved the fines quality (Giertz 1977, 1981) so that paper of acceptable strength could be produced. However, from the experience obtained in this study, it was found that fines from TMP, as well as from CTMP and CMP, contained not only ray cells and VE fragments, but also abundant fibrillar material and lamellae. In fact, TMP fines appeared to have longer fibrils than those from either CTMP or CMP, as illustrated in Figures 5.57 to 5.60. Although it is not clear what is the influence of the shape and size of the material in the fines fraction on pulp strength, the matter of fines quality deserves further attention. Thus, based on the results of removal of fibre surface layers and on the breakdown of VE, an attempt was made to obtain information about fines quality (P100 fraction) according to the origin of the material. The results are presented in

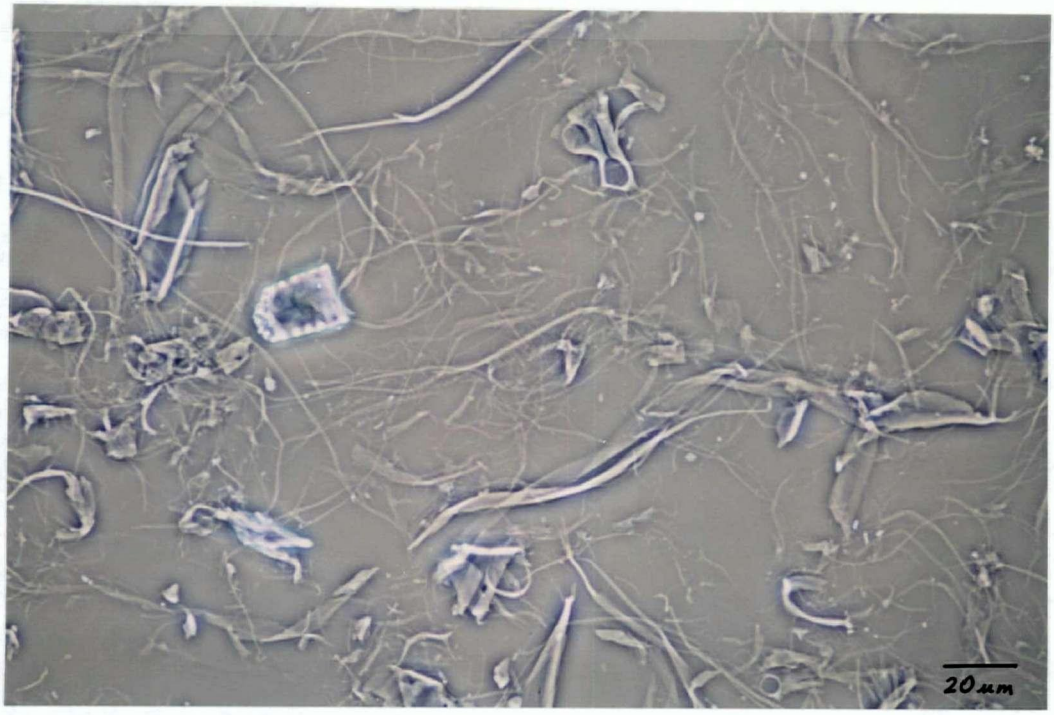


Figure 5.57. P200 fraction of aspen TMP pulp under phase contrast illumination (pulp A-TMP3).

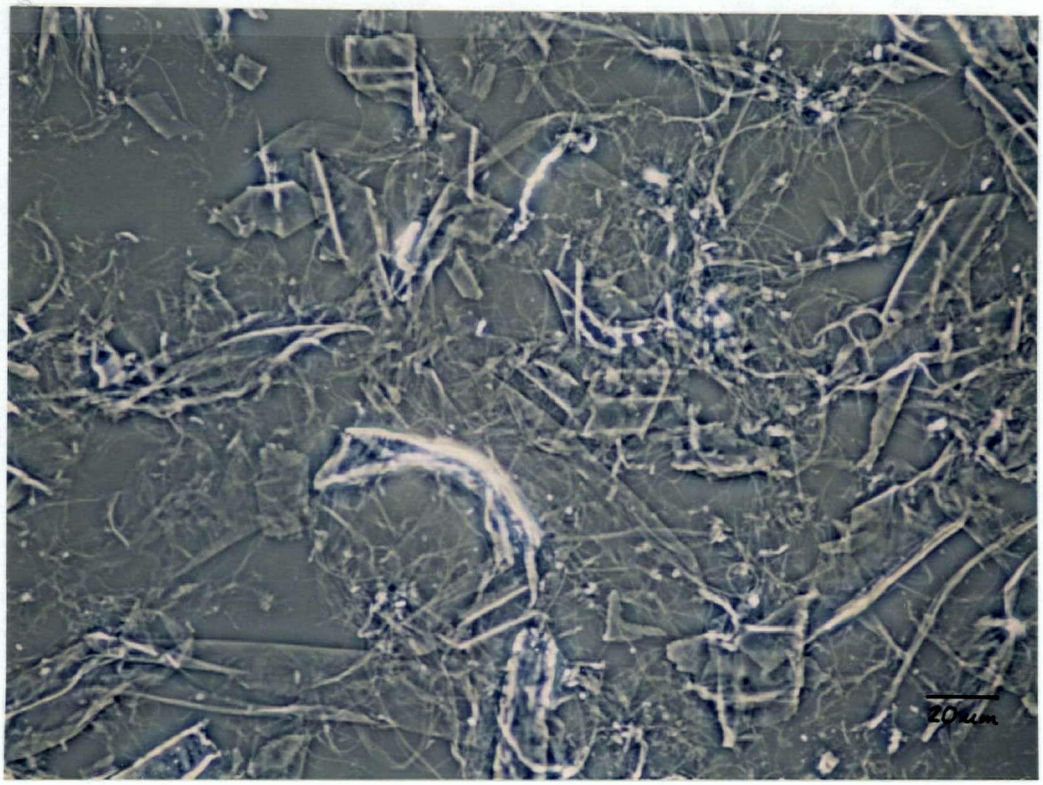


Figure 5.58. P200 fraction of aspen CMP pulp of similar freeness than that in Fig. 5.57 (pulp A-CMP3).

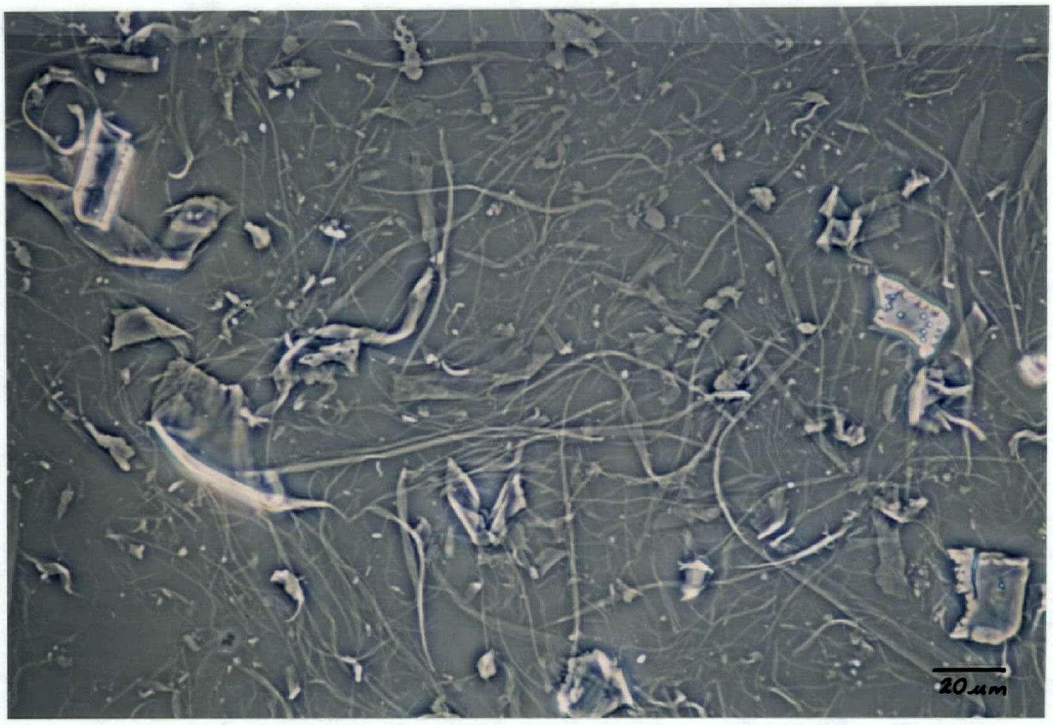


Figure 5.59. P200 fraction of birch TMP pulp under phase contrast illumination (pulp B-TMP3).

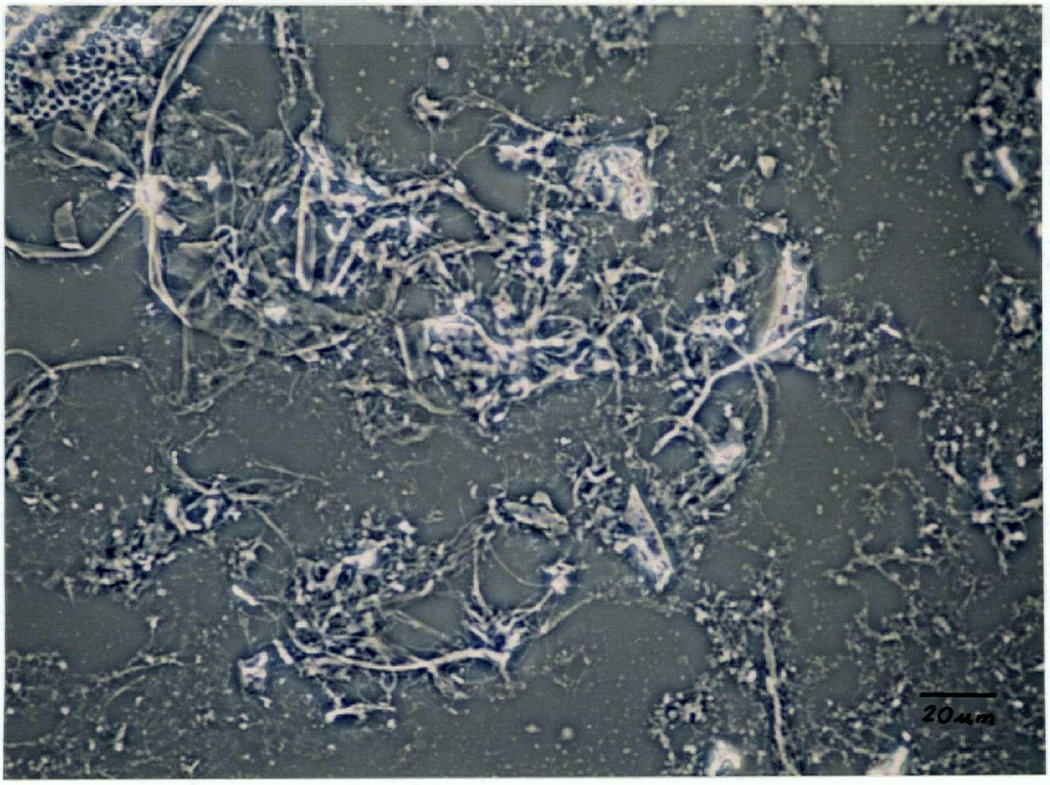


Figure 5.60. P200 fraction of birch CMP pulp of similar freeness than that in Fig. 5.59 (pulp B-CMP3).

Figures 5.61 and 5.62, and the assumptions and calculation procedures involved are listed in Appendix E.

The large amount of material originating from the fibre S_2 layer in pulp A-CTMP4 was mostly due to the cutting effect mentioned earlier for this pulp and is evident from the comparatively large values of the 100/150 and 150/200 fractions. Otherwise TMP fines contained, as expected, large amounts of VE and ray cells, as well as considerable amounts of material from the S_2 layer of the fibre wall. Excluding aspen CTMP pulps, the composition of the fines fraction does not seem to differ much between processes for the same species. If anything, the proportion of fines derived from the S_2 layer, to those from VE and ray cells, appears to be larger for the TMP pulps, particularly for birch refiner pulps. If there is a difference in the bonding capacity of the fines fraction due to the process used in producing the refiner pulp, it may be due more to the flexibility of these particles than to the chemical composition or length of the particles involved.

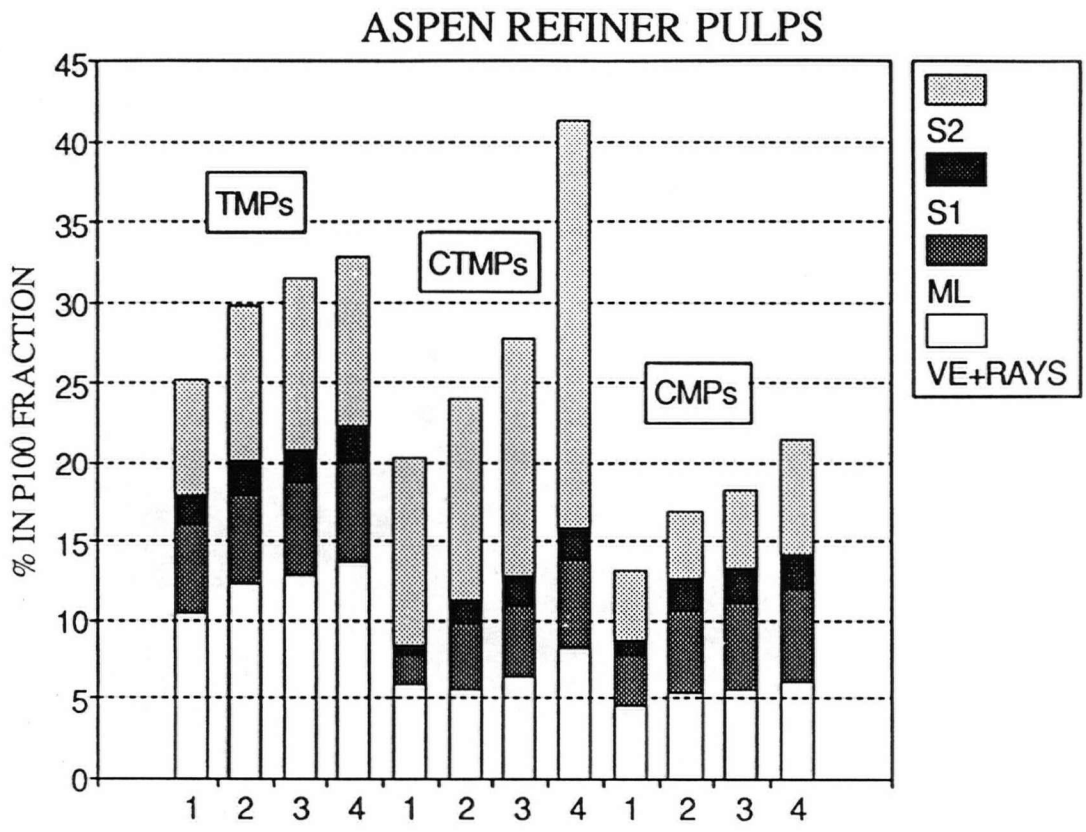


Figure 5.61. Estimated composition of the P100 fraction for aspen refiner pulps according to the origin of the fine material.

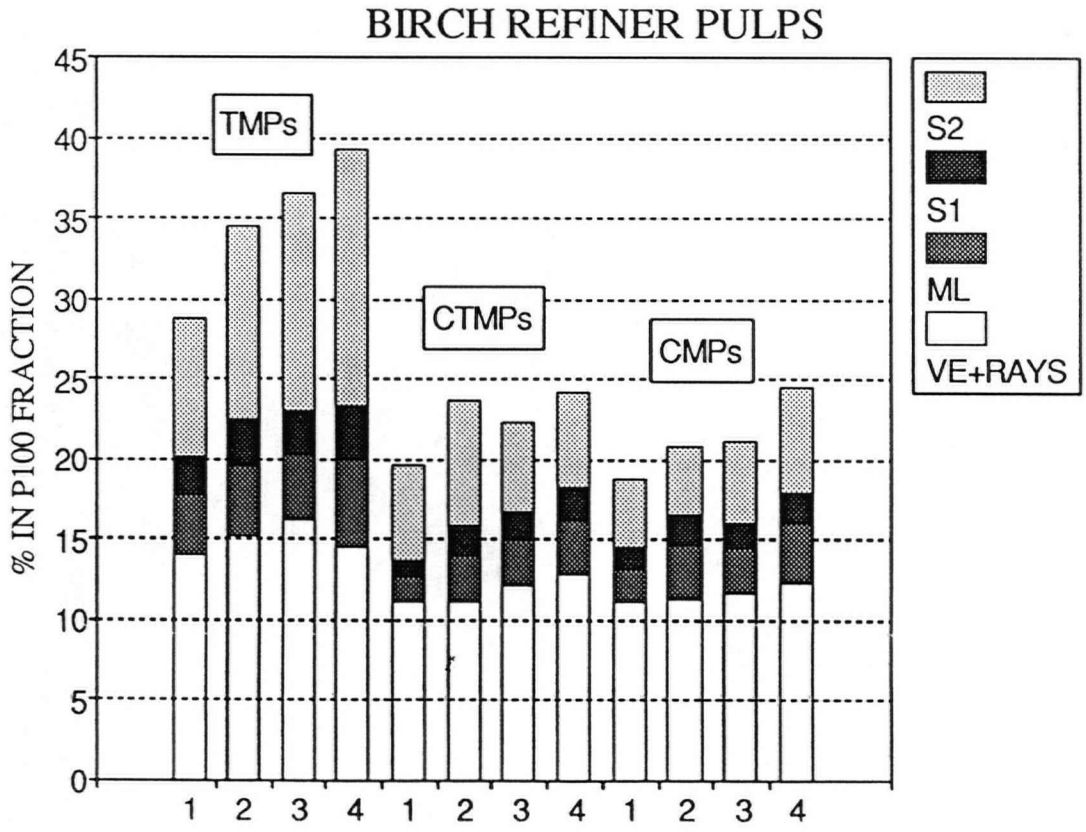


Figure 5.62. Estimated composition of the P100 fraction for birch refiner pulps according to the origin of the fine material.

VI. SUMMARY

Fundamental aspects of the breakdown of hardwood elements into mechanical pulps were studied in detail. The most important aspect of this study was the assessment of the surface quality of hardwood refiner pulp fibres. The investigation was based on the defiberization of trembling aspen (*Populus tremuloides* Michx.) and white birch (*Betula papyrifera* Marsh.) chips. These were refined under TMP, CTMP and CMP conditions, to a broad range of pulp freeness levels (generally between 100 and 300 mL CSF).

For each pulp, thin cross sections of fibres from the R48 fraction were prepared. Three-hundred fibres were analyzed for each pulp. The fibre cross sections were stained to provide differentiation of the compound middle lamella and the secondary wall. In this manner, and by using polarized light microscopy to assess the presence of the S_1 layer, the surface quality of fibres was evaluated.

In general, fibres produced by TMP processing showed high proportions of S_2 layer exposure when compared to those produced under CTMP or CMP conditions, particularly for birch pulps. It was shown that the chemical pretreatments used did not improve fibre surface quality in terms of S_2 layer exposure or extent of fibrillation. In fact, fibres from chemically-treated chips presented more retention of the compound middle lamella.

When chemical pretreatment was applied to the chips, the

species response varied considerably. Birch CTMP and CMP fibres had high middle lamella retention (MLr indices over 70% at pulp freeness levels over 300 mL CSF), and these values decreased slowly with further refining. Moreover the weak S_1/S_2 boundary, along with the high retention of ML, often produced birch fibres covered by a sheath of S_1/ML that had separated from the S_2 layer, but was not removed. This sheath was sometimes rolled back, exposing the S_2 layer. Aspen responded differently to the application of chemical pretreatment. Although the retention of ML in CTMP and CMP aspen fibres was high for pulps of high freeness, increased refining produced a quick response. ML retention decreased faster than it did for birch pulp fibres. Consequently, at freeness levels of about 100 mL CSF, aspen CTMP and CMP pulps showed levels of S_2 exposure, comparable to those obtained for aspen TMP fibres.

Other important fundamental differences were found between TMP fibres and those resulting from chemically-treated chip processing. Radially-failed fibres were frequent in TMP but not in CTMP nor CMP pulps. Formation of ribbon-like particles, therefore, was common only in TMP pulps. These pulps also showed preferential breakdown of the G-fibres present in aspen chips. Not only were the G-fibres broken down into smaller fractions, but in many cases the G-layers were denuded from the cell wall, exposing their highly cellulosic surface. This was not the case in the G-fibres from chemically-treated chips, for which the G-layer

generally remained inside the fibres.

Other categories discussed in the analysis of fibre cross-sections included delamination of the S_2 layer and proportion of fibres distorted due to chemical impregnation. Delamination tends to appear more frequently in chemically-treated fibres, and more often in aspen than in birch. Distortion of the fibre cross-sectional shape is more common when the chip chemical pretreatment is more severe.

Pulp properties were measured. It was found that pulp strength did not relate to the degree of exposure of the S_2 layer, but rather closely followed fibre flexibility (as measured by sheet density). Thus, despite the superior bonding potential of TMP fibres due to their large exposure of the S_2 layer, the fibres remained stiff thereby producing sheets of low density and strength.

The breakdown of vessel elements (VE) was studied by comparing VE size frequency distributions and the proportion of whole VE that survived refining. While TMP processing reduced VE into small fragments, wood softening due to chemical pretreatment was responsible for the survival of high proportions of whole VE in CTMP and CMP pulps. Further refining, however, reduced the amount of whole VE in these pulps. Considerable difference was observed between the relative survival of VE in aspen and birch. Birch VE were destroyed more easily, since failure in the VE walls occurred along the line of the fine intervessel pitting.

The breakdown of VE and the removal of the surface layers of

the fibres allowed the estimation of the origin of the fines in the P100 fraction of a Bauer McNett classifier. Although TMP fines contained larger amounts of VE compared to CTMP or CMP pulps, the amount of material derived from the fibres' S_2 layer was also larger. Furthermore, TMP fines contained longer filaments. For papermaking purposes, however, the quality of the fines may not be judged only on their composition .

VII. CONCLUSIONS

The following are considered the major conclusions of this investigation:

1. TMP processing of birch and aspen wood chips produced fibres with higher exposure of the S_2 layer than normally obtained with refiner pulps from chemically-treated wood chips. For aspen CTMP or CMP pulps, additional refining to freeness values of about 100 mL CSF had to be applied to obtain levels of S_2 exposure equivalent to those of their TMP counterparts. Birch showed a slower response than aspen in the removal of ML and S_1 layers upon the refining of fibres produced from chemically-treated chips. Thus, it can be concluded that there was an effect due to species and processing conditions on the surface characteristics of the fibres.
2. Improvements in pulp sheet density due to the removal of the S_1 layer of the fibres were small compared to the effects resulting from chemical pretreatment. Fibres with high values of ML and S_1 layer retention, for example high freeness CTMP and CMP pulps, were much more flexible (as measured by sheet density) than low freeness TMP fibres with high exposure of the S_2 layer. Tensile strength development followed sheet density levels rather than fibre surface quality. Fibre surface quality did not have an important role in the strength development of the

pulps. Thus, it is not the lack of S_2 exposure that gives hardwood TMP low strength properties, but rather a lack of fibre flexibility.

3. Chemical pretreatments used in this study did not improve the fibre surface in terms of S_2 exposure. On the contrary, chemically-treated fibres had generally more ML retention at equivalent freeness levels. Furthermore, chemical pretreatments used in this study did not improve fibrillation of the resulting fibres.
4. Birch fibres showed gaps between the S_1 and S_2 layers (out/in effect), implying a weak bond between these layers. In TMP pulp fibres, this resulted in facile separation along or near the S_1/S_2 boundary, giving fibres with the S_2 layer largely exposed. In fibres made from chemically-treated chips, this initial gap between S_1 and S_2 layers appeared to be responsible for the total separation of the S_1 layer and for provoking the skinning effect. Thus, the thickness of the S_1 layer, known to be larger in birch than in aspen, was not the limiting factor to achieve separation of this layer from the fibre surface.
5. TMP processing of wood chips was effective in destroying VE. Not only did it produce pulps with virtually no whole VE, but most VE fragments produced were very small indeed. On the other hand, chemical pretreatments preserved whole VE in CTMP pulps and even more so in CMP. At freeness levels close to 300 mL CSF, aspen CMP

preserved about 80% of the VE originally present in the wood, while less than 60% whole VE survived for birch. In general, birch VE were more readily destroyed because of their preferential breakdown along the intervessel pitting arrangement in their walls. There was an effect of species as well as of pulping conditions in the breakdown pattern of VE.

Other important findings of this study are summarized below:

6. The presence of tension wood (presence of G-fibres) accounted for approximately 31% of the aspen fibres. The method developed to assess the proportion of G-fibres in wood or pulp provided accurate results. TMP processing significantly reduced the amount of G-fibres in the R48 fraction of the pulp. Also, TMP pulps contained abundant liberated G-layers that were stripped from their parent shells. Material derived from the G-layers was present in all TMP fibre fractions. CTMP and CMP pulps did not generally show preferential breakdown of the G-fibres.
7. Radial failure of fibres occurred in 20% of the TMP pulp fibres, while it was generally below 5% in CTMP and CMP pulps.
8. Distortion and delamination of fibres were observed as a result of chemical treatment of the chips. However, current techniques of recording such changes are not enough for describing and maximizing the effects of

refining. A measure of the extent of these changes needs to be developed to facilitate the recording of shape change and extent of delamination.

9. The techniques used in the study of cross-sectional characteristics of fibres allowed the detailed analysis of large numbers of fibres under the standard light microscope. On the other hand, TEM provided more details on the cross sections but, due to the intensity of sample preparation involved, only a limited number of fibres could be observed.

10. The fines fraction of TMP pulps contained abundant fibrillar material in addition to many VE fragments. The effect of the fines fraction on the strength of the resulting pulp is not clear.

VIII. SUGGESTIONS FOR FURTHER RESEARCH

Based on the work done in this study, the following suggestions for further research can be made:

a. One obvious topic of study is the influence of S_2 exposure on pulp strength. This can be achieved by testing handsheets of fibres of known but different degrees of S_2 exposure. Only the long-fibre fraction should be used to form the sheets to eliminate the effect of the other fractions. Also the sheets should be produced at the same density, since sheet density has a strong effect on pulp strength.

b. The effect of surface quality on fibre flexibility could be studied within a given pulp. For this, flexibility could be measured on individual fibres which then could be analyzed for surface quality.

c. Even with the S_2 layer largely exposed and fibrillated, and with the production of ribbon-like particles in TMP pulps, these pulps were weak compared to the corresponding CTMP or CMP pulps. In order to take advantage of the potentially beneficial characteristics of TMP fibres, the effect of an interstage treatment that focusses on swelling the fibres should be studied. A high degree of fibre swelling which may translate into flexibility should be achieved which, upon drying, could provide strong pulps. If the rigidity of fibres is the

main reason for the high exposure of the S_2 layer in TMP pulps, perhaps other medium and high density hardwood species with rigid fibres will produce high S_2 layer exposure. These pulps may then be improved with the proper interstage or post-chemical treatment.

d. The effect of tension wood pulp quality should be studied for different proportions of G-fibres in the wood furnish. This can be done by analysis of cross sections of chemical pulps produced from the chip material. Since it is known now that in TMP pulps the G-layers can be stripped off and produce cellulosic filaments, the effect of TW should prove to be beneficial. The effect of chemical treatments before the refining should also be assessed. Of particular interest is the use of an interstage process that will swell and possibly enhance fibrillation of the G-layers after their denuding from the parent fibres.

   The proportion of tension wood (measured as G-fibres) should be determined for geographical areas in which hardwoods are utilized for pulping.

e. Little is known about the quality of the fines from hardwood mechanical pulps. The bonding and light scattering ability of these particles should be studied for different refiner pulping processes. This could be done by mixing a fixed percentage of fines with a standard furnish. It should be mentioned that the fines composition may vary with the level of refining. Thus, it

is recommended that the fine material be collected at different levels of refining. It would be interesting to assess the effect of an interstage treatment and of a post-treatment in the quality of TMP fines. Swelling of the fines particles may prove to enhance bonding on the paper sheet. The interaction of fibres and fines of different qualities might also be investigated.

f. The use of an image analysis system could provide information about the extent of distortion of fibres due to chemical treatment and refining in the analysis of fibre cross sections. The actual restriction to fibre swelling due to retention of the S_1 could then be effectively measured. On the other hand, the staining techniques could be modified to maximize the contrast between chemically affected and untreated fibres. Other applications of the quantitative analysis of distorted fibres in cross section, include studies of liquor penetration. The degree of swelling and the number of swollen fibres could be assessed with this method for different pretreatment conditions.

g. The effect of the presence of whole VE in refiner pulps from chemically-treated chips is not known. Tests on surface strength and picking tendency are recommended. If the presence of whole VE proves to be a problem during printing, several options are available for attempting their solution.

h. On a more fundamental study, the reasons for the initial gap found for birch fibres between the S_1 and S_2 layers, can be investigated. These include a possible abrupt change in fibril angle between these wall layers and/or substantial changes in their chemical composition. The apparent weak bond between the S_1 and S_2 layers, however, may only be present in fibres of birch trees grown under certain conditions. A weak S_1/S_2 boundary may also exist in other hardwood species. After learning to take advantage of the exposed S_2 layer, other species could be studied.

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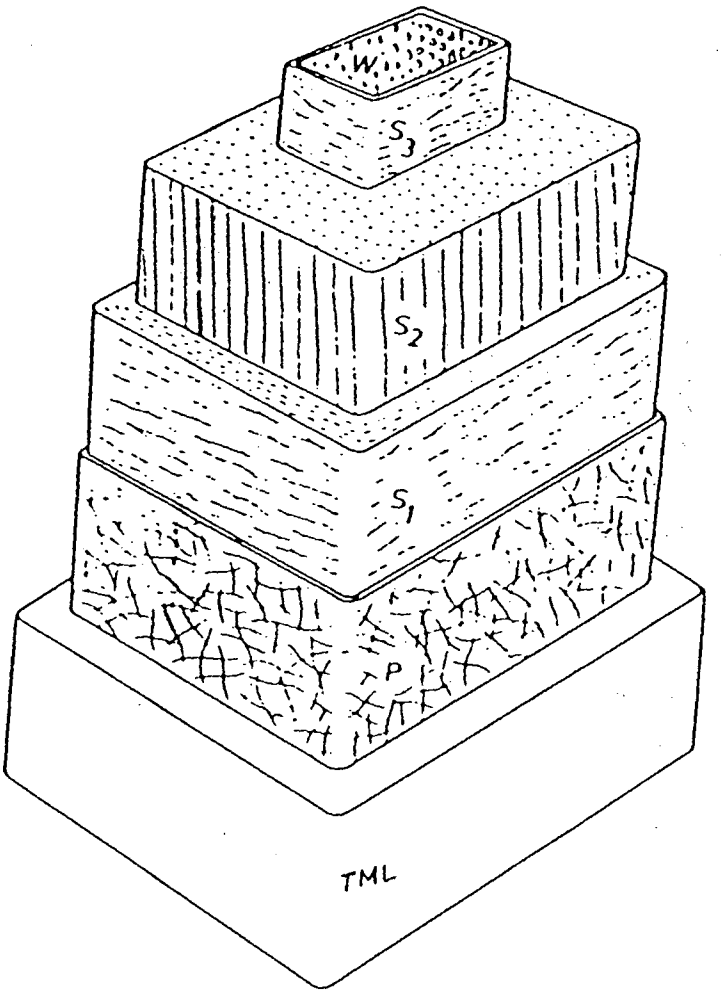
APPENDIX A: Hardwood fibre structure according to Jayme and Azzola (1965).



TML: true middle lamella
P: primary wall
S_2, S_2, and S_3: outer, middle and inner layers of the secondary wall
W: warty layer

APPENDIX B: Chart used for recording categories of fibres in cross section.

ML RETAINED			S_1 RETAINED			ML		S_2 EXPOSED		WALL DAMAGES			G			OTHER
O	<50%	>50%	TOT.	<50%	>50%	TOT.	FLY	IN/OUT	PART.	TOT.	R.F.	DEL.	M.C.	IN	ONLY	"d"	

1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23 24 25

APPENDIX C: Results and discussion on repeatability of the analysis of fibre cross sections.

Results

Table C.1. shows the categories of fibre cross-sectional analysis under which there was no agreement between the two sub-samples (150 fibres each) taken from every pulp, as tested by differences of proportions, at a 95% confidence level. From a total of 328 tests performed, only 30 were found to give significantly different results, i.e. over 90% of the tests performed showed repeatability within the sample, as presented in a summary in Table C.2. The cases in which the differences were found to be significant, did not follow any definite pattern and they appear to be distributed randomly in Table C.1.

Discussion

There is no record in the literature of quantitative microscopical analysis on cross sections of hardwood refiner pulps. Therefore, the statistical evaluation of the techniques involved was pertinent. From the results presented in Tables C.1 and C.2 it was established that over 90% of the tests were repeatable. Although there is no precedent for comparison that would indicate the relative correctness of this work, the results obtained in terms of repeatability were encouraging. It is indicated that when significant differences occurred, the gap between the real difference and the statistically allowable one was always

Table C.1. Repeatability of tests in the analysis of fibre cross sections, based on sub-samples of 150 fibres at a 95% confidence level.

ASPEN REFINER PULPS

FIBRE CROSS SECTIONAL FEATURE	TMP				CTMP				CMP				TOTAL
	1	2	3	4	1	2	3	4	1	2	3	4	
MLr=0%		*											1
MLr<50%													0
MLr>50%							*						1
MLr=100%													0
S1r=0%, S2e=100%													0
S1r<50%, S2e>50%				*									1
S1r>50%, S2e<50%													0
S1r=100%, S2e=0%													0
PEELING OUTER LAYER			*										1
OUT/IN EFFECT					*			*					2
RADIAL FAILURE						*							1
DELAMINATION		*			*								2
DISTORTED FIBRES												*	1
G LAYER INSIDE													0
G LAYER ONLY		*											1
													11

BIRCH REFINER PULPS

FIBRE CROSS SECTIONAL FEATURE	TMP				CTMP				CMP				TOTAL
	1	2	3	4	1	2	3	4	1	2	3	4	
MLr=0%			*					*					2
MLr<50%				*					*	*			3
MLr>50%													0
MLr=100%					*		*						2
S1r=0%, S2e=100%			*					*					2
S1r<50%, S2e>50%	*			*						*	*		4
S1r>50%, S2e<50%						*	*				*		3
S1r=100%, S2e=0%							*						1
PEELING OUTER LAYER					*								1
OUT/IN EFFECT			*										1
RADIAL FAILURE													0
DELAMINATION													0
DISTORTED FIBRES													0
													19

* : Significant differences between subsamples

Table C.2. Summary of repeatable categories on the analysis of fibre cross sections.

FIBRE CROSS SECTIONAL FEATURE	NUMBER OF CATEGORIES (TESTS) ASPEN	BIRCH	TOTAL
MLr	48	48	96
S1r or S2e	48	48	96
PEELING OUTER LAYER	12	12	24
OUT/IN EFFECT	12	12	24
RADIAL FAILURE	12	12	24
DELAMINATION	12	12	24
DISTORTED FIBRES	8	8	16
G LAYER INSIDE	12	-	12
G LAYER ONLY	12	-	12
TOTAL	176	152	328
CATEGORIES:			
Not repeatable	11	19	30
Repeatable	165	133	298
% Repeatable	94	88	91

less than 7 fibres and generally less than 3 fibres for the subsamples of 150 fibres. In all cases, the average for the 300 fibres in each category was taken as the value representing the fibre cross sectional characteristic.

The fact that the cases (less than 10%) in which significant differences occurred, appeared to be randomly spread in Table C.1, suggests that it may be possible to improve the overall repeatability rather than to concentrate on a particular category. Whilst it is difficult to establish if the lack of repeatability in some categories was due to human error or actual variation in the pulp, an examination of the possible causes of error is in order. These include:

- Variation of wood density
- Production of pulp and sampling
- Sample preparation
- Human error

The variation of wood specific gravity was large within a tree, as shown in Appendix D. Even if a variation of this magnitude also applied to such characteristics of the fibres which control their response to refiner pulping (as measured by cross sectional analysis), it is unlikely that this effect would be significant because the wood material was thoroughly mixed before pulping to minimize such variation. It was assumed then that, within a species, every pulp subsample was produced from a representative sample of a

chip mix. Density variation, therefore, does not seem to be a probable cause for lack of repeatability.

Pulp production as a source of error refers to variations in the refining conditions while a pulp is being produced. The experimental pulps were obtained over a period of time in which specific energy was not perfectly constant. Thus, "pockets" of fibres with different characteristics may form within a given pulp. However, the pulp is subsequently mixed and sampled for hot disintegration. This sampling is done from a well mixed pulp by taking small amounts until the required pulp weight for disintegration has been reached. After screening and fractionation, the R48 sample should be quite uniform. Thus, any variation during the pulp production process should have been mitigated or eliminated by subsequent pulp handling.

Methods used for sample preparation may have contributed as sources of error. Preparation was a long and tedious procedure consisting of many different steps of which fibre alignment and staining of cross sections were the most critical ones. Fibre alignment was important to identify properly the presence of the S_1 layer. The technique used rendered satisfactory results, and the alternative technique of aligning fibres one by one was considered impractical for a large number of hardwood fibres. The staining methods also provided satisfactory identification of the ML. Thus, sample preparation does not appear to be a major source of error.

The point at which the fibres were sectioned would have an

impact on their cross-sectional features. The assumption that the cross section of a fibre at any point represents features along the entire length of the fibre, is clearly not entirely correct. However, it is reasonable to assume that the fibres cut at the middle may represent better the features of an entire fibre than of those fibres sectioned near their ends. It may be possible that the majority of the fibres in one subsample were cut near their middle while the section on the second subsample was made near the fibre ends, thus producing a significant difference in some categories when testing repeatability. However, an average section probably contained fibres cut at different places throughout their lengths, so that sectioning does not appear to play a significant role.

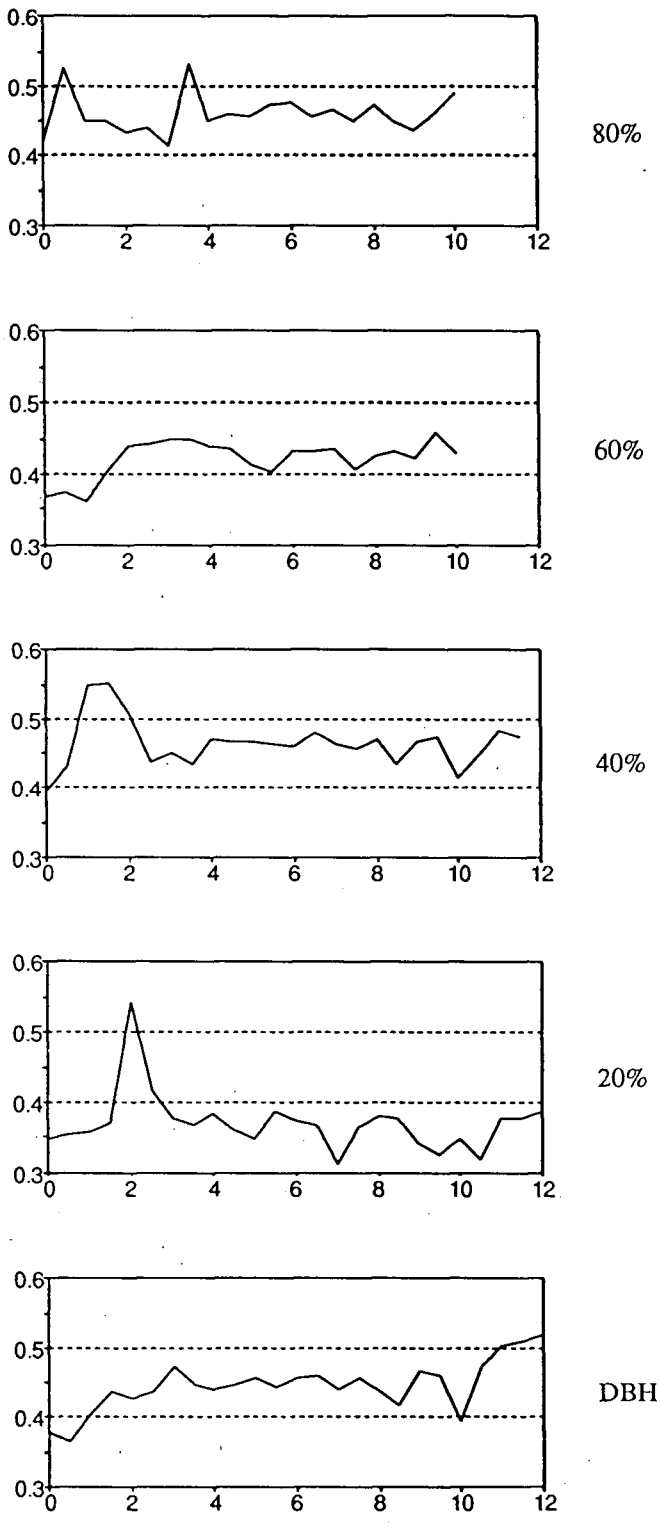
There is the possibility of human error in recording cross sectional features of fibres under the light microscope. Although the samples were analyzed in a random fashion to minimize observer bias, the possibility of error could probably be further reduced if all the fibres were analyzed through a transmission electron microscope. It is felt that the ideal analysis of fibre cross sections would be carried out on ultrathin sections, cut with a diamond knife, for observation under a TEM. This system allows the use of staining techniques combined with very high resolution, which minimizes observer bias. The principal limitation, however, lies in the small number of fibres viewed in one field, as well as in the difficulties encountered and time

required for cutting large amounts of ultrathin sections; in addition, there are high costs involved in using a TEM system. TEM was used in this study to confirm the observations made under the light microscope in terms of the retention of ML and S_1 layer, the exposure of the S_2 layer, and to observe additional details regarding features of the fibres in cross section.

In summary, no clear conclusion could be drawn on the source of lack of repeatability for approximately 10% of the tests on differences of proportions between subsamples. It is possible, however, that there could be significant variation of the fibre cross-sectional characteristics within pulp samples.

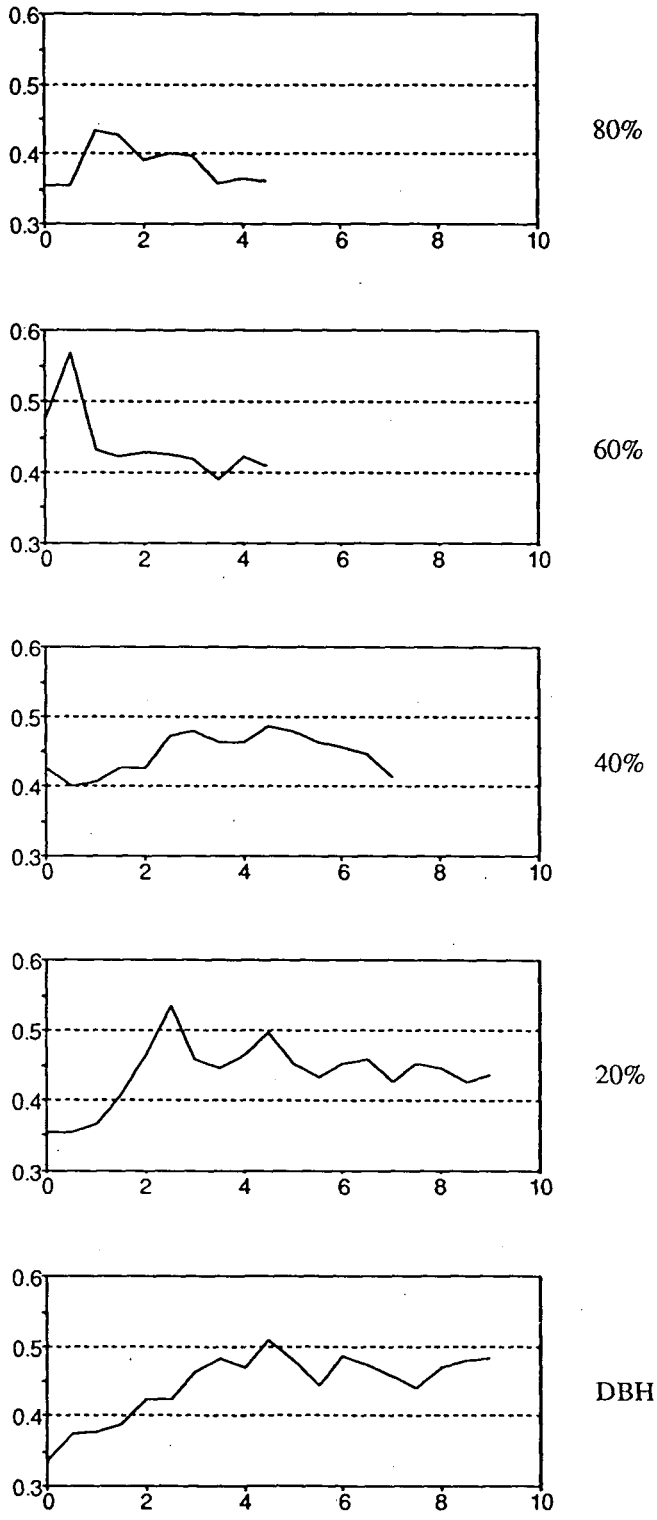
APPENDIX D: Variation of wood specific gravity from pith to bark at different heights (as %) of the tree stem.

SG DISTRIBUTION, ASPEN TREE FROM LYTTON, B.C.



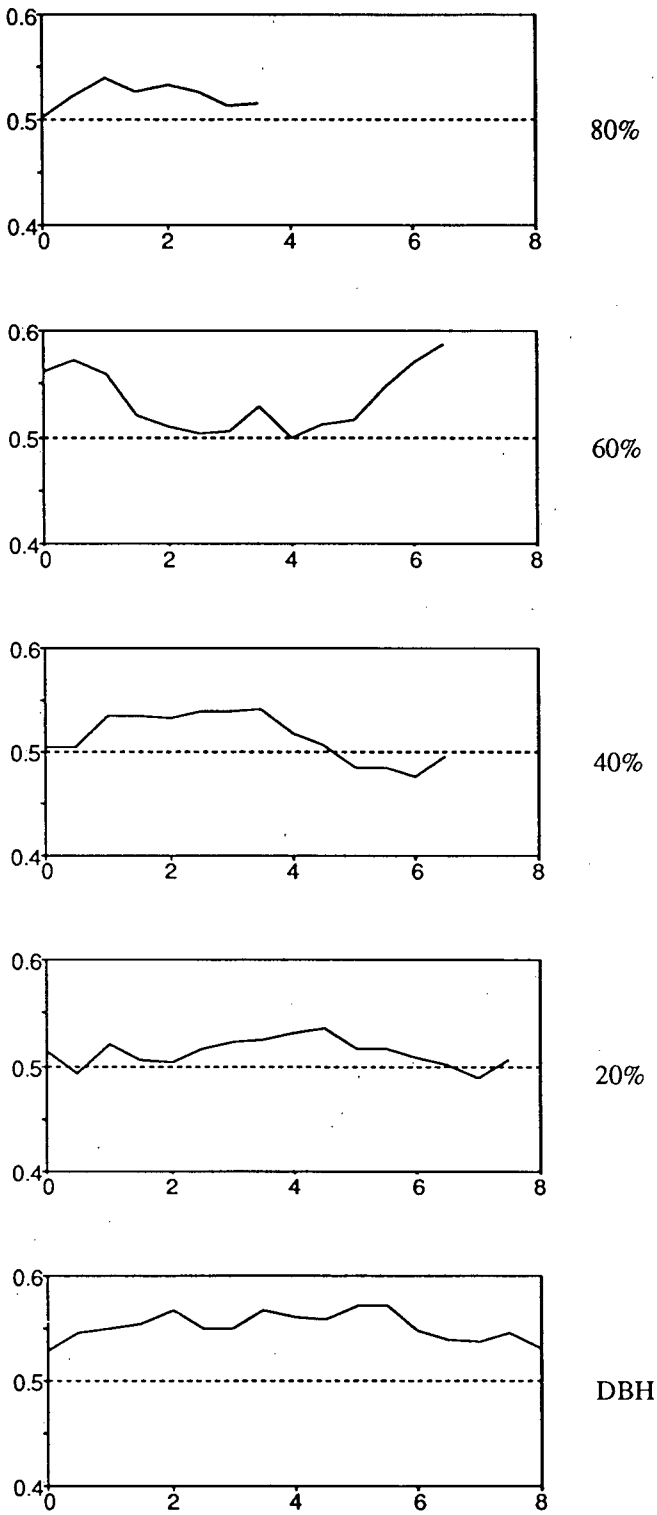
APPENDIX D (continued)

SG DISTRIBUTION, ASPEN TREE FROM WILLIAMS LAKE, B.C.



APPENDIX D (continued)

SG DISTRIBUTION, BIRCH TREE FROM WILLIAMS LAKE, B.C.



APPENDIX E: Calculations involved in the estimation of the origin of the material in the P100 fraction.

Assumptions:

1. The values presented in Tables E.1 and E.2 for aspen and birch are applicable to the wood samples used in this study.

2. All the components of elements in Table E.1 have the same density, so that the values can also be taken as weight percentages.

3. Any loss of material affects evenly all the elements.

4. Any material removed from the fibre surface ends up in the P100 fraction of the pulp.

5. The weight of VE fragments from the frequency tables are proportional to their size.

6. Material from the G-layers is not taken into account.

Procedure:

1. Ray cells
   Since the ray cells were smaller than 150 *um*, it is taken that all this material passed the 100 mesh screen openings. Values of 4 and 10% of the wood weight were taken for aspen and birch, respectively.

2. Vessel elements
   It was considered that all VE fragments larger than the lower confidence limit of the mean from the kraft pulp VE size distribution, could be regarded as whole VE. The rest were considered VE fragments. From these fragments, the portion included in the first class interval, was taken as the weight of VE passing the 100 mesh screen. This portion, multiplied by the percentage of VE that were fragmented, times the values for VE from Table E.1, gave the weight of VE in the P100 fraction.

3. ML material
   Values for ML plus MLcc, as defined in Table E.1, were taken from this table and multiplied by the portion of ML that was removed from the fibres.

4. S_1 layer material
   The proportion of S_1 layer material to that of the secondary wall was calculated from values presented in Table E.2, and multiplied by values for fibre secondary wall in Table E.1. The results were then multiplied by

the portion of S_1 material that was removed from the fibres.

5. S_2 layer material
This was calculated by substracting the values obtained in points 1, 2, 3 and 4 (above) from the weight of the P100 pulp fraction.

Table E.1
PERCENTAGE VOLUME OF COMPONENTS OF ELEMENTS
(Source: Musha & Goring 1975)

	FIBRE			VESSEL		RAY	
	S	ML	MLcc	S	ML*	S	ML*
ASPEN	74.0	6.0	3.0	11.0	2.0	3.0	1.0
BIRCH	73.4	5.2	2.4	8.2	0.8	10.0	0.0

*: Assuming the middle lamella width found for fibres
S: secondary wall; ML: middle lamella; cc: cell corner

Table E.2
FIBRE DIMENSIONS IN
CROSS SECTION (microns)

	DIAMETER (1)	TOTAL SECONDARY WALL (1)	S1 LAYER (2)
ASPEN	18.8	2.55	0.12
BIRCH	19.5	3.60	0.21

(1): Musha & Goring (1975)
(2): Marton et al (1979)

APPENDIX F: ABBREVIATIONS

ML	Compound Middle Lamella
MLr	Retention of Compound Middle Lamella
MLrI	Compound Middle Lamella Retention Index
ML(r=0)	No Retention of Compound Middle Lamella
ML(r<50)	MLr on less than 50%, but more than zero of the fibre cross section
ML(r>50)	MLr on more than 50%, but less than 100% of the fibre cross section
ML(r=100)	Total Retention of the Compound Middle Lamella
S_1r	Retention of the S_1 layer
S_1rI	S_1 layer Retention Index
S_1(r=0)	No Retention of the S_1 layer
S_1(r<50)	S_1r on less than 50%, but more than zero of the fibre cross section
S_1(r>50)	S_1r on more than 50%, but less than 100% of the fibre cross section
S_1(r=100)	Total Retention of the S_1 layer
S_2e	Exposure of the S_2 layer
S_2eI	S_2 layer Exposure Index
S_2(e=0)	Zero exposure of the S_2 layer
S_2(e<50)	S_2e on less than 50%, but more than zero of the fibre cross section
S_2(e>50)	S_2e on more than 50%, but less than 100% of the fibre cross section
S_2(e=100)	Total Exposure of the S_2 layer
RF	Radial Failure
TW	Tension Wood
VE	Vessel Element