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THE MECHANISM OF RUPTURE OF EARLY- AND LATEWOOD IN REFINING
(LE MÉCANISME DE RUPTURE DU BOIS DE PRINTEMPS ET BOIS D'ÉTÉ AU COURS DU RAFFINAGE)

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Summary

The most distinguishable variability in fibre morphology of softwood grown in temperate zone exists between earlywood (EW) and latewood (LW), especially in fibre length, cell wall thickness, cross-section dimension, and pit size. Generally, LW fibres are longer, have thicker cell wall and smaller radial width than EW fibres. EW fibres have more pits and their pits surfaces are larger than those of LW. Because of these morphological differences, the EW and LW breakdown dissimilarly during refining in terms of cell wall rupture modes (trans-wall, intra-wall failures), cell wall thickness reduction, fibre shortening, fibre wall splitting, fibrillation, collapsibility and other physical changes.

The principal objective of this research project is to study the breakdown mechanism of Jack pine (*Pinus banksiana*) EW and LW in thermomechanical pulping (TMP) by means of microscopic observations. Characteristics such as fibre splitting, shortening, delamination (internal fibrillation) and external fibrillation, etc. are evaluated. Physical changes in the EW and LW fibres are qualified and quantified with the aide of light microscopy as well as scanning electron microscopy. The impacts of the observed changes on pulp and paper properties are assessed to establish possible interrelation between the fibre characteristics and paper properties.

In this study, the Jack pine EW and LW chips were firstly separated manually using a chisel. Then, the EW and LW chips were either separately refined or in blend according to the initial proportion of each existing in the wood. The refining was carried out in two stages: pressurized primary stage and atmospheric secondary stage. The pressurized stage was conducted at two temperatures: 120°C and 160°C. Owing to the high refining pressure, the refiner plate clearance at 160°C had to be reduced to half of that at 120°C. Finally, handsheets were made from different pulps which were produced in separate refining of EW, LW and EW/LW mixed furnish. Other furnishes employed include fines-free pulps and pulps of different Bauer-McNett fractions or in combination. We also compared the differences in papermaking properties between "Chip Mixing" in co-refining of EW and LW chips and "Pulp Mixing" or post-refining mixing of separately

refined EW and LW pulps. The physical and optical characteristics of all these handsheets were analysed.

Comparisons between the EW and LW chips indicate that EW is lighter in colour while LW has higher bulk density. In addition, the cell wall thickness of LW is twice as thick as that of the EW. Chemically, the LW has higher holocellulose content while EW contains more lignin and dichloromethane (DCM) extractives.

EW and LW fibres behave differently in thermomechanical pulping (TMP). During refining, the thin-walled EW fibres tend to separate in the P/S1 interface and show little external fibrillation. The thick-walled LW fibres usually separate in the P/S1 and S1/S2 regions, causing considerable external fibrillation. In addition, EW fibres manifest more severe delamination (internal fibrillation) and have higher water retention value (WRV) than the LW fibres. Moreover, LW fibres exhibit mostly intra-wall failure and lower curl and kink indices, while EW fibres tend to fail in trans-wall mode (split) and show higher curl and kink indices. As a result, LW yields higher fibres length while EW suffers more fibre cutting. EW fibres are more collapsible and conformable than the LW counterparts.

The natures of fines and rejects generated from EW and LW are different. The thick-walled LW fibres produce more fibrillar components in the fines while the thinned-walled EW fibres fines contain more unfibrillated and flake-like fragments. As a consequence, the EW fines have higher surface lignin coverage while the LW fines give higher specific volume by sedimentation (SV). The rejects of the LW pulp has more fibrillar elements which tend to entangle with other fibres forming aggregates of fibrils. The aggregates cause an increase in rejects content in the LW pulp. In contrast, the EW rejects contains more bundle-like shives.

Refining at 160°C, which operates with reduced plate gap, produces more rejects and generates more fibrillation from the LW but yields more collapsed fibres form the EW when compared with the refining at 120 °C.

The morphological modifications of EW and LW fibres in refining strongly influence the papermaking characteristics. For a given freeness, the handsheets made from EW

pulps show higher sheet density, lower roughness, porosity and opacity than those prepared from LW pulps. At a given specific energy, the handsheets made from LW pulps yield better physical strengths, in terms of Scott bond, tensile, burst, and tear indices than EW counterparts. In addition, EW pulps have higher brightness, light absorption and light scattering coefficients than LW pulps.

In comparison to the refining at 120°C, for both EW and LW, the refining at 160°C produces pulps having better sheet density, physical strength (Scott bond, tensile, burst, and tear resistances), sheet opacity and light scattering and absorption coefficient while the surface roughness, porosity, and brightness are negatively affected.

Regarding the handsheets made from the Bauer-McNett fibre fractions (R14 to R100), EW pulps always exhibit better physical properties than LW furnishes do. In addition, LW fines (P200) have greater influences than EW fines on the physical properties of handsheets because they contain a higher proportion of fibrils.

The characteristics of the pulps produced in co-refining of pre-mixed chips (EW and LW chips) are different from those made by blending the two separately refined pulps (EW and LW pulps). In the former case, the fibres have greater average fibre length and better tearing resistance as compared to the latter case. In contrast, the fibres produced in separate refining are more developed and have more fines, resulting in increased physical properties and light scattering coefficient.

In terms of sheet structure, thin-walled EW fibres have better collapsibility and conformability while thick-walled LW fibres produce sheets with higher bulk and have greater fibre twists and fibrils in the network.

This study reveals that the majority of long fibre fractions (R14+R28+R48) are derived from LW in refining of Jack pine. These long LW fibres are less developed and have greater coarseness. To improve the pulp quality, we suggest that the long fibres be extracted by fractionation and further developed by refining with additional energy. It may also desirable to refine them at an elevated temperature or treat them with chemicals, which would minimize the cutting effect and improve inter-fibre bonding.

Keywords: *Pinus banksiana*, earlywood, latewood, Thermomechanical pulping, fibre morphology, light microscopy, scanning electron microscopy, physical and optical properties

Résumé

Il existe une grande variation de morphologie des fibres entre le bois de printemps (BP) et le bois d'été (BE) chez les résineux dans la zone tempérée. Ces variations incluent notamment la longueur des fibres, l'épaisseur de paroi cellulaire, la dimension transversale et la taille des ponctuations. En général, les fibres de BE sont plus longues que celles de BP. Dans la section transversale, les fibres de BE ont des parois plus épaisses et des largeurs radiales plus étroites par rapport à celles de BP. Le BP présente des ponctuations plus nombreuses et ayant des surfaces plus grandes que celles de BE. À cause de ces différences structurelles, les fibres du BP et du BE auraient des comportement différents au cours du raffinage au niveau du mode de rupture (inter ou intra cellulaire), de la réduction en épaisseur, du clivage de la paroi cellulaire, de la coupure en longueur, de la fibrillation, de l'aplatissement et d'autres changements physiques.

Cette étude a pour objectif principal d'examiner le mécanisme de rupture du BP et du BE de pin gris (*Pinus banksiana*) au cours de la mise en pâte thermomécanique (PTM) à l'aide de la microscopie. Les caractéristiques telles que le clivage de la paroi cellulaire, de la coupure en longueur, de la délamination (la fibrillation interne) et de la fibrillation externe sont évaluées. Les changements physiques de fibres du BP et du BE sont qualifiés et quantifiés avec l'aide du microscope optique et du microscope électronique à balayage (MEB). Les impacts de ces changements sur les propriétés du papier sont évalués, ce qui pourrait nous aider à établir les rapports possibles entre les caractéristiques des fibres et celles du papier.

Dans cette étude, les copeaux du BP et du BE de pin gris sont d'abord séparés manuellement à l'aide d'un ciseau à bois. Par la suite, les copeaux du BP et du BE ont été raffinés individuellement ou en mélange selon la proportion massique initiale dans le bois entier. Le raffinage s'est réalisé en deux étapes: une 1re étape pressurisée à 120°C et 160°C et une 2^{ième} étape à pression atmosphérique. À cause de la haute pression à 160°C, la distance entre les plaques du raffineur a due être réduite de la moitié par rapport à celles utilisée à 120°C pour obtenir raffinage semblable. Finalement, des

feuilles ont été fabriquées à partir des différentes pâtes produites à partir de copeaux de BP et de BE seuls ainsi qu'avec leurs mélanges. Des formettes ont aussi été fabriquées à partir de pâtes sans fines et de pâtes des différentes de fractions Bauer-McNett ainsi que leurs combinaisons. On a également examiné les caractéristiques des feuilles qui proviennent de deux processus de mélange de BP et de BE, c'est-à-dire, le «mélange des copeaux avant raffinage» et le «mélange des pâtes après raffinage». Les propriétés physiques et optiques ont été évaluées pour toutes les sortes de feuilles.

La comparaison entre les copeaux de BP et de BE démontre que le BP est plus clair que le BE. Ce dernier a une densité basale plus grande que le premier. Par ailleurs, l'épaisseur de paroi cellulaire du BE est presque le double de celle du BP. La composition chimique de ces deux bois est aussi différente. Le BE contient plus d'hollocellulose que le BP, mais ce dernier a plus de lignine et plus des matières extractibles dans le dichloromethane (DCM) que le BE.

Le BP et le BE se comportent différemment dans la mise en pâte thermomécanique (PTM). Le BP présente une paroi plus mince et a tendance à se séparer à l'interface P/S1 en manifestant peu de fibrillation externe. Le BE, à paroi plus épaisse, est souvent brisé dans les régions P/S1 et S1/S2, ce qui cause une fibrillation externe considérable. De plus, les fibres de BP montrent des délaminations (fibrillation interne) et des capacités de rétention d'eau «water retention value» plus importantes que celles de BE. Les fibres des BP exhibent principalement des ruptures trans-paroi cellulaire (clivage) alors que celles de BE sont plutôt intre-paroi. Les fibres de BP ont un indice de courbure «curl index» et un indice en dents de scie «kink index» plus élevés que celles de BE. Par conséquence, au cours du raffinage, le BE produit plus de fibres longues alors que les fibres de BP subissent plus de coupures en longueur. En outre, le BP, à paroi plus mince, manifeste un degré d'aplatissements et de conformabilité plus grand que le BE qui a une paroi plus épaisse.

La production de fines et de rejets du BP et du BE est différente au cours du raffinage. Les fibres de BE produisent plus de fines de type fibrillaire tandis qu l'on retrouve plus de fines en flocons dans les fines de BP. Ces dernières contiennent plus de lignine à leurs surfaces alors les premières ont des volumes spécifiques par sédimentation (VS)

plus grands. Dans les rejets, les fibres de BE ont également plus de fibrilles. Les agrégats de ces fibrilles rendent la production de rejets de BE plus élevée que pour le BP. Par contraste, les rejets de BP contiennent plus de bûchettes.

Lors du raffinage à 160°C, la distance entre les plaques du raffineur doit être réduite pour obtenir le même niveau de raffinage, ce qui produit plus de rejets (emmêlement des flocons et des fibrilles) et génère plus de fibrillation externe pour le BE tandis que, pour le BP, plus de fibres écrasées sont présentes lors de la comparaison avec le raffinage à 120°C.

Les différences mécanisme de rupture du BP et du BE au cours du raffinage influencent fortement les caractéristiques physiques et optiques du papier. Pour un indice d'égouttage donné, les feuilles fabriquées à partir des pâtes de BP ont une densité plus élevée mais une rugosité, une porosité et une opacité plus basses que celles de BE. Pour une énergie donnée, les feuilles faites à partir de pâte de BE ont des résistances physiques, telles que la cohésion interne «Scott bond», l'allongement, l'éclatement ainsi que la déchirure plus fortes que celles de BP. De plus, les feuilles de BP ont des blancheurs supérieures, un coefficient d'absorption de la lumière ainsi qu'un coefficient de diffusion de la lumière plus élevées par rapport à celles de BE.

En comparaison avec le raffinage à 120°C, pour le BP et le BE, le raffinage à 160°C produit des feuilles qui ont une plus grande densité et des résistances physiques (cohésion interne «Scott bond», allongement, éclatement et déchirure) supérieures. L'opacité ainsi que les coefficients d'absorption et de diffusion de la lumière sont plus élevés tandis que la rugosité, la porosité et la blancheur des feuilles sont négativement affectées.

Dans les tests des feuilles fabriquées à partir des fractions Bauer-McNett (de R14 à R100), les BP exhibent toujours propriétés physiques supérieures à celles de BE. En outre, les fines (P200) de BE ont une influence plus importante que celles de BP au niveau des résistances physiques des feuilles car elles contiennent une haute proportion de fibrilles.

Les pâtes démontrent des caractéristiques différentes dans les deux processus de mélange de BP et de BE tels que le «mélange de copeaux» et le «mélange de pâtes». Dans le premier cas, les fibres sont plus longues et ont une meilleure résistance à la déchirure que celles dans le dernier cas. Par contraste, les fibres dans le processus de «mélange de pâtes» sont mieux développées et ont plus de fines. Cela a une influence positive sur les caractéristiques des feuilles au niveau des propriétés physique et du coefficient de diffusion de la lumière par rapport au processus «mélange de copeaux».

En observant la structure des feuilles, les fibres de BP avec les parois plus minces ont un plus grand degré d'aplatissement et de conformabilité. Néanmoins, les feuilles fabriquées de fibres de BE sont plus bouffantes et ont plus de torsion dans les fibres ainsi que plus de fibrilles.

Cette étude révèle que la majorité des fibres longues dans les fractions R14, R28 et R48 proviennent du BE au cours du raffinage. De plus, ces fibres longues du BE sont moins développés et ont des masses linéiques plus élevées. Afin d'améliorer les qualités de la pâte, on conseille de séparer les fibres de BP à partir des fractions de fibres longues par fractionnement avant d'imposer une énergie supplémentaire aux fibres longues (ou aux fibres de BE) pour les développer. Il semble préférable de raffiner ces fibres à température plus élevée ou après un traitements chimiques, ce qui est favorable pour ramollir les fibres et minimiser la coupure de fibre en longueur. Une telle pratique pourrait certainement améliorer les liens inter fibres et la qualité de la pâte.

Mots clés: *Pinus banksiana*, bois de printemps, bois d'été, mise en pâte thermomécanique, morphologie des fibres, microscopie optique, microscopie électronique à balayage, propriétés optique et physique

Résumé substantiel

PROBLÉMATIQUE

Le bois est un matériel ayant des caractéristiques chimiques et physiques variables entre les espèces, entre les arbres de la même espèce et même entre les différences parties de la tige à l'intérieur d'un même arbre. Cependant, dans la zone tempérée, les plus grandes variations morphologiques des fibres se trouvent entre le bois de printemps (BP) et le bois d'été (BE) chez les résineux. En général, les fibres de BP ont un périmètre extérieur plus grand, une paroi cellulaire plus mince ainsi qu'une largeur radicale plus grande par rapport à celles du BE. Ces différences ont des effets sur, non seulement les propriétés des fibres telles que la densité et les résistances mécaniques, mais aussi sur les mises en pâte chimique et mécanique ainsi que sur les propriétés du papier.

Lors du raffinage, les parois cellulaires du BP brisent plus facilement car leur épaisseur est plus mince, elles ne supportent pas les forces mécaniques causées par les barres du raffineur. Par contre, les fibres de BE, aux parois plus épaisses, ont des résistances plus fortes à l'action du raffinage et ainsi subissent des réductions importantes d'épaisseur au cours du raffinage.

Due à leur paroi cellulaire plus mince, les fibres du BP ont tendances à changer de forme transversalement facilement en manifestent un plus grand degré d'aplatissement et de flexibilité. Inversement, les fibres de BE, aux parois plus épaisses sont plus résistantes à de tels changements au cours du raffinage. Des rapports précédents montrent que le BP se brise en fragments de formes irrégulières (bûchettes) tandis que le BE se désintègre en fibres individuelles. Des recherches indiquent aussi que le BP a besoin de plus d'énergie pour atteindre le même niveau d'indice égouttage que le BE.

De plus, à cause de leurs différences morphologiques, le BP et le BE influencent différemment les propriétés du papier. Par exemple, les fibres de BP produisent un papier de haute densité, tandis que les fibres de BE produisent des papiers avec une rugosité et une résistance à la déchirure plus élevées.

Malgré ces découvertes, il y encore des questions sans réponse: comment ces deux types de bois se transforment-ils du bois solide en des fibres individuelles? De quelle manière contribuent-ils aux résistances physiques du papier? Dans cette investigation, nous espérons apporter un peu de lumière sur ces questions. Effectivement, bien que des progrès se fassent au niveau de la compréhension de la théorie du raffinage, il y a très peu de connaissance sur les mécanismes de rupture du BP et du BE au cours du raffinage, particulièrement dans la mise en pâte thermomécanique (PTM). De nombreuses questions ne sont pas élucidées, surtout sur la réaction du BP et du BE en fonction de la température du raffinage, leur mécanisme de rupture (inter ou intra cellulaire) ainsi que sur leur consommation énergétique. Jusqu'à ce jour, nous savons peu de chose sur les rapports entre les changements morphologiques du BP et du BE au cours du raffinage et leur influence sur les propriétés du papier. Dans cette étude, un processus systématique de recherche est proposé, sur la séparation du BP et du BE, sur la mise en pâte thermomécanique à différentes températures, ainsi que sur la caractérisation des feuilles. Des séries d'analyses physique et chimique sont réalisées dans chaque étape expérimentale. Ces études nous aideront à mieux comprendre les mécanismes de rupture de ces deux types de bois pour ainsi améliorer les qualités des pâtes qui proviennent du pin gris puisque il est présentement sous-exploité et moins désirable dans les industries forestières. Le fruit de cette recherche nous aidera également à améliorer l'efficacité du raffinage.

OBJECTIF

Cette étude a pour un objectif principal d'examiner le mécanisme de rupture du BP et du BE dans la mise en pâte thermomécanique (PTM) par le biais d'observations au microscope. Les caractéristiques telles que le clivage des fibres, la réduction de la longueur, la délamination (fibrillation interne) ainsi que la fibrillation externe, etc. sont évaluées. Les changements physiques des fibres de BP et de BE sont qualifiés et quantifiés à l'aide du microscope optique et du microscope électronique à balayage (MEB). Des changements observés sont évalués dans le but d'établir un lien entre les caractéristiques des fibres et les propriétés du papier.

Les points importants dans cette étude sont les suivants:

1. Indentification des fibres de BP et de BE dans les pâtes d'après leurs différences morphologiques telles que l'épaisseur cellulaire, la taille et la distribution des ponctuations et la taille du lumen;

2. Détermination des compositions chimiques du BP et du BE, notamment la cellulose, les hémicelluloses et la lignine;

3. Évaluation des changements physiques des fibres de BP et de BE au cours du raffinage, particulièrement sur le clivage de fibre, la délamination de la paroi cellulaire, la coupure en longueur des fibres, les productions de fines et de rejets;

4. Mesures des propriétés optiques et physiques des feuilles fabriquées par des pâtes de BP et de BE.

Plusieurs analyses sont employées pour caractériser les propriétés physiques des fibres ainsi que les changements structurels résultant de l'action de raffinage. Les aspects d'analyses incluent:

- Modifications morphologiques des fibres telles que les modes de séparation, le changement de longueur, la surface de lumen, l'épaisseur de la paroi cellulaire, le clivage, l'aplatissement ainsi que la fibrillation externe et interne;

- Les caractéristiques de feuille telles que la densité, l'indice de rupture, l'indice d'éclatement, l'indice de déchirure, la porosité, la rugosité, la blancheur, le coefficient de diffusion de la lumière, le coefficient d'absorption de la lumière. Les résultas de ces analyses sont reliées avec les observations microscopiques en espérant établir des liens entre les caractéristiques des fibres et les propriétés du papier.

MÉTHODOLOGIE

Afin d'étudier le mécanisme de rupture de BP et BE dans la mise en pâte thermomécanique (PTM), nous avons élaboré des processus expérimentaux, incluant des méthodes pour caractériser les propriété physiques et chimiques du BP et du BE sous différentes formes, c'est-à-dire sous forme de copeaux, de pâtes et de feuilles.

Préparation des matériaux

Des billes de pin gris (*Pinus banksiana* Lam.) fraîchement coupées sont utilisées dans cette expérimentation. Le pin gris provient d'une plantation de 30 ans de la région de la Mauricie. Les billes sont d'abord coupées en rondelles de 2,5 cm d'épaisseur à l'aide d'une scie mécanique. Par la suite, les rondelles sont écorchées manuellement avec un ciseau à bois. Les copeaux sont préparées à partir de l'aubier en excluant le bois de cœur afin que minimiser les effets des matières extractibles sur les propriétés de la pâte. Des copeaux de BP et de BE d'environ 2-3 cm en largeur et en longueur, sont ainsi préparés manuellement avec un ciseau à bois. L'épaisseur des copeaux varie en fonction de la largeur des anneaux de croissance ainsi que de la proportion de BP et de BE dans cette région. La séparation du BP te du BE est basée sur leur différence en couleur, car le BE est plus foncé que le BP.

Raffinage

Les pâtes de BP et de BE ont été produites avec le raffineur de laboratoire CD 300 (Mesto Inc.) du Centre Intégré en Pâtes et Papiers d l'UQTR. Les copeaux de BP et de BE sont raffinés individuellement ou en mélange selon leur proportion massique initiale dans le bois entier. Les raffinages sont divisés en deux étapes. La première étape s'effectue sous pression à des températures de 120 °C et 160 °C, tandis que la seconde étape est à pression atmosphérique. L'indice d'égouttage de la pâte dans la première étape est d'environ 500 mL, tandis que dans la deuxième une plage de 50-250 mL est ciblée. La consistance de la première étape est plus élevée que celle de la deuxième étape, soient 20-24% contre 10-14%. De plus, dû à la haute pression de raffinage avec la température de 160°C, l'entrefer dans cette condition est plus bas que celui à température moins élevée, soit 120°C.

Fractionnement de la pâte

Les pâtes du deuxième stade de raffinage sont fractionnées sur un appareil de classage Bauer-McNett en 6 fractions: R14, R28, R48, R100, R200 et P200 (fines). Cette méthode de fractionnement de fibre est généralement utilisée pour caractériser les

distributions en longueur des fibres des pâtes. Les fibres dans chaque fraction sont morphologiquement différentes, ce qui pourrait avoir une influence sur les propriétés du papier. Par exemple, les fibres longues de la fraction R14 sont rigides et leur surface moins fibrillée offre peu de contact pour les liaisons inter-fibres. Les matières fines P200 sont définies comme les particules qui passent à travers un tamis 200 mailles au pouce du classificateur Bauer-McNett. Les fines sont celles que l'on retrouve dans la pâte, soit les cellules de parenchyme, les rayons ainsi que les débris de la paroi cellulaire S1 et S2. Les fines ont une haute surface spécifique (de 10 à 50 m^2/g alors les fibres ont une surface de 1 m^2/g), ce qui favorise une meilleure consolidation de la feuille lors du séchage en augmentant les forces de compression sous l'effet de la tension de surface. De plus, les fines augmentent la surface de contact en remplissant les cavités ou pores.

Caractérisation de la pâte

Les fibres dans chaque fraction sont caractérisées sur leur mode de rupture (inter et intra cellulaire), la délamination de la paroi, le clivage des fibres, les fibrillations externes et internes, ainsi que l'aplatissement. Ces analyses sont réalisées par microscopies optique et électronique. De plus, le volume spécifique par sédimentation (VS) des fines est évalué. Il présente une mesure du degré de développement de la pâte, surtout au niveau de la fibrillation externe. Ce concept est toutefois utilisé pour les pâtes après un traitement mécanique tel le raffinage. Aussi, la fibrillation interne peut se qualifier par une mesure de rétention d'eau de gonflement «Water Retention Value». La fibrillation interne (délamination) correspond à un gonflement de la paroi cellulaire suite à une action mécanique telle que le raffinage. Le gonflement de la fibre s'effectue par un clivage tangentiel des fibrilles élémentaire de la macrofibrille. De toute évidence, la présence de ces caractéristiques sur les fibres implique une augmentation appréciable de la flexibilité des fibres dans les feuilles.

La longueur des fibres, l'indice de courbure «curl index», l'angle de brisure «kink index» et la masse linéique sont déterminées à l'aide d'appareil optique sur des suspension de fibres dans l'eau tel que le FQA «Fibre Quality Analyser, Optest Equipment». Au cours du raffinage, les fibres peuvent êtres coupées (i.e. réduction de la longueur), courbées (i.e. indice de courbure «curl» et indice en dents de scie «kink») ou

fibrillées en diminuant la masse linéique de la fibre. La facilitée avec laquelle les fibres changent morphologiquement dépend de l'épaisseur de la paroi cellulaire ainsi que de la flexibilité des fibres. Toutes ces modifications sont d'une grande importance pour obtenir une meilleure formation du papier. Par exemple, la porosité et l'absorption d'eau augmentent avec la courbure des fibres. Une diminution de longueur entraîne une augmentation au niveau de l'opacité et du fini de surface des feuilles.

Le pourcentage de rejets est déterminé par l'appareil «Pulmac Shive Analyzer, MasterScreen) tandis que le pourcentage des fines est obtenu par le classificateur Bauer-McNett.

L'épaisseur de paroi cellulaire de fibre est évaluée à l'aide d'un appareil tel l'analyseur d'épaisseur de paroi de fibre (Techpap, France). Ce paramètre est une caractéristique morphologique importante qui est liée avec la rigidité des fibres ou à leur habileté à créer des liaisons entre les fibres. Cette mesure nous apporte des informations sur la réponse de fibre au raffinage.

Fabrication de feuille

En général, les fibres longues des fractions R14, R28 et R48 sont rigides, moins développées et présentent peu de contacts inter fibres. Les fines (P200) comprennent des particules en rubans, des rayons ainsi que des fibrilles. Les fines de pâte mécanique jouent un rôle important pour les résistances physiques de la feuille. Afin de mieux comprendre l'influence des caractéristiques de fibres sur les propriétés papetières, plusieurs catégories de feuilles sont faites à partir de différentes fractions Bauer-McNett de BP et de BE ainsi que leurs mélanges. Les feuilles sont fabriquées à partir des pâtes comme suit:

- Pâte entière;
- Pâte sans fines (P200);
- Combinaison des fractions de fibres longues (R14+R28+R48);

- Combinaison des fractions de fibres courtes (R100+R200+P200);
- Fractions individuelles R14, R28, R48 et R100;
- Mélange des pâtes de BP et de BE après le raffinage. Ce mélange a la même proportion massique que le mélange des copeaux de BP et de BE avant le raffinage. Les comparaisons nous permettent de comprendre l'effets de mélange pour les deux processus, soient le co-raffinage et le raffinage individuel de BP et BE.

Caractérisation de feuille

Les propriétés physiques, chimiques, et optiques sont analysées sur les feuilles des pâtes du deuxième stade de raffinage. À partir de ces mesures, on espérait établir des liens possibles entre les caractéristiques de fibres et les propriétés papetières.

a. Propriétés physiques telles que l'allongement, la déchirure, l'éclatement, la densité, la porosité, la rugosité, et la liaison inter fibres (Scott Bond);

b. Propriété chimique telle que la teneur de lignine à la surface de fibre (spectroscopie par rayon X);

c. Propriétés optiques telles que la blancheur, le coefficient de diffusion de la lumière, le coefficient d'absorption de la lumière et l'opacité;

d. Propriétés structurelles telle que l'angle de torsion de la fibre dans la feuille, qui pourraient être mesurées par microscopie électronique à balayage (MEB).

e. Les caractéristiques intrinsèques telles que l'épaisseur de parois cellulaires, la taille du lumen, l'aplatissement, le clivage des fibres sont aussi analysées par la MEB.

RÉSULTAS ET DISCUSSION

1. Caractéristiques des matières primaires

La couleur des copeaux de BP est plus claire que celle du BE. Ce dernier a une densité basale plus élevée. De plus, il contient plus de fibres longues dans la matrice du bois. Les fibres de BE ont une double épaisseur de paroi cellulaire plus grande que celle du BP. Les feuilles produites à partir de pin gris ont des résistances physiques inférieures à celles fabriquées d'essences traditionnelles telles que l'épinette car la première contient une proportion importante de BE.

Quant aux compositions chimiques, le BE a plus d'holocellulose que le BP, alors que ce dernier contient plus de lignine. De plus, le BP a plus de matières extractibles dans le dichlorométhane (DCM) puisque il a plus de canaux résinifères, abondants en résine. Ceci pourrait être à l'origine des problèmes de poix associés au pin gris dans la mise en pâte.

2. Caractéristiques des pâtes, pour un indice d'égouttage donné

À cause de leurs différences morphologiques, le BP et le BE se comportent différemment lors de la mise en pâte thermomécanique (PTM), aussi bien au niveau de la séparation que du développement des fibres. Le BP, à la paroi mince, a tendance à se séparer dans l'interface P/S1 en manifestant peu de fibrillation externe tandis que le BE, à paroi plus épaisse, est souvent brisé dans les régions P/S1 et S1/S2, ce qui cause une fibrillation externe considérable. De plus, les fibres de BP présente une délamination (fibrillation interne) et une capacité de rétention d'eau «water retention value» plus importantes que celles de BE.

Au cours du raffinage, les fibres de BE sont plus résistantes que celles de BP aux forces mécaniques. Les fibres de BP exhibent principalement des ruptures trans-paroi cellulaire (clivage) alors que celles de BE sont plutôt de type inter-paroi. Les fibres de BP ont un indice de courbure «curl index» et un indice en dents de scie «kink index» plus élevés que celles de BE. Par conséquent, au cours du raffinage, le BE produit plus de fibres longues que les fibres du BP, qui subissent plus de coupures en longueur.

Face aux forces de compression existant dans le raffinage, les fibres de BP et de BE manifestent des déformations transversales différentes. Grâce à leur paroi cellulaire plus mince et à un lumen plus large, les fibres de BP montrent plus d'aplatissement et de conformabilité que celles de BE qui ont des parois plus épaisses.

La production de fines et de rejets du BP et du BE est fortement influencée par leur modification morphologique au cours du raffinage. Les fibres de BE sont fibrillées (fibrillation externe) considérablement en produisant plus d'éléments fibrillaires dans leurs fines tandis qu'on trouve plus de flocons dans les fines de BP. Par conséquent, les fines de BP contiennent plus de lignine à leur surface que celles de BE qui ont un volume spécifique (VS) plus grand. Principalement en raison de leur développement de fibre différent, les rejets de BP et de BE sont qualitativement et quantitativement différents. Dans les rejets, les BE ont également plus de fibrilles. Les agrégats de ces fibrilles rendent la production de rejets de BE plus élevée que le BP. Par contraste, les rejets de BP contiennent plus de bûchettes.

Lors du raffinage à 160°C, la distance des plaques du raffineur doit être réduite pour obtenir le même niveau de raffinage, ce qui produit plus de rejets (emmêlement des flocons et des fibrilles) et génère plus de fibrillation externe pour le BE tandis que, pour le BP, plus de fibres écrasées apparaissent en comparaison avec le raffinage à 120°C.

3. Caractéristiques des feuilles

La différence dans la mode de rupture du BP et BE au cours du raffinage influence fortement les caractéristiques physiques et optiques du papiers. Pour un indice d'égouttage donné, grâce à leur conformabilité des fibres supérieure, les feuilles fabriquées à partir des pâtes de BP ont une densité plus élevée, mais une rugosité, une porosité et une opacités plus basse que celles de BE. Pour une énergie donnée, en raison de leurs fibres hautement fibrillées ainsi que leur abondance d'élément fibrillaires dans leur fines, les feuilles de BE ont des résistances physiques plus fortes que celles de BP telles que le «Scott bond», l'allongement, l'éclatement ainsi que la déchirure. De plus, les feuilles de BP ont des blancheurs supérieures dues à leurs valeurs initiales plus élevées. En outre, les pâtes de BP ont un coefficient d'absorption de la lumière plus

grand que celles de BE car elles contiennent plus de lignine. Les pâtes de BP ont aussi un coefficient de diffusion de la lumière plus élevée par rapport à celles de BE puisqu'elles produisent plus de particules en flocons dans les fines, ce qui est favorable pour la diffusion de la lumière.

En comparaison avec le raffinage à 120°C, pour le BP et le BE, le raffinage à 160°C produit des feuilles qui ont une plus forte densités et des résistances physiques telles que la cohésion interne «Scott bond», l'allongement, l'éclatement et la déchirure, l'opacité, ainsi que les coefficients d'absorption et de diffusion de la lumière plus élevés. La rugosité, la porosité et la blancheur des feuilles sont négativement touchées. Ces caractéristiques sont expliquées par l'augmentation du contenu en fines et un plus grand aplatissement des fibres dans le raffinage à 160°C par rapport à celui à 120°C.

Dans les tests des feuilles fabriquées à partir des fractions Bauer-McNett (de R14 à R100), malgré leur bas degré de fibrillation dans ces fractions de fibres, le BP exhibe toujours des propriétés physiques supérieures à celles de BE. Il semble que les fibres de BP manifestent un plus grand indice de conformabilité pendant la consolidation de feuille en formant de meilleures liaisons inter-fibres que celles de BE.

Les fines de pâte thermomécanique (PTM) jouent un rôle important pour les propriétés physiques des feuilles telles que la densité, l'allongement, l'éclatement, la déchirure, la rugosité, la porosité ainsi les propriétés optiques. En raison des différences dans les compositions de fines, les fines du BP et du BE démontent des contributions différentes aux propriétés physiques des pâtes. Les fines (P200) de BE ont des influences plus importantes que celles de BP aux niveaux des résistances physiques des feuilles car elles contiennent une haute proportion de fibrilles, ce qui est plus bénéfique aux liaisons inter fibres que les flocons produit par le BP.

Les pâtes démontrent des caractéristiques différentes dans les deux processus de mélange de BP et de BE tels que le «mélange de copeau» et le «mélange de pâtes». Les interactions du BP et du BE sont différentes dans ces deux processus. Cela rend les propriétés des feuilles différentes. Lors du raffinage en mélange des copeaux, probablement en raison d'un effet tampon possible, les fibres à parois minces de BP sont

protégées par les fibres à parois épaisses de BE contre le traitement mécanique sévère au cours du raffinage. En conséquence, les fibres sont plus longues et ont une meilleure résistance à la déchirure que lorsque les pâtes sont mélangées après raffinage. Par contraste, les fibres dans ce dernier cas sont mieux développées et ont plus de fines. Cela a une influence positive sur les caractéristiques des feuilles aux niveaux des propriétés physiques et du coefficient de diffusion de la lumière par rapport au raffinage des copeaux en mélange.

En observant la structure de feuilles, on note que les fibres du BP à paroi plus mince présentent un plus grand aplatissement et une meilleure conformabilité. Néanmoins, les feuilles fabriquées de fibres de BE sont plus bouffantes. Les fibres ont un niveau de torsions plus grand et ont plus de fibrilles que le BP.

Cette étude révèle que la majorité fibres longues dans les fractions telles que les (R14+R28+R48) provient du BE. De plus, ces fibres longues du BE sont moins développées et ont des masses linéiques plus élevées. Afin d'améliorer la qualité de la pâte, on conseille de séparer les fibres de BP de ces fractions de fibres longues par fractionnement avant d'appliquer une énergie supplémentaire sur ces fibres longues (ou de BE) pour les développer. Il est désirable de raffiner ces fibres à une température plus élevée ou après un traitement chimique, ce qui est favorable pour ramollir les fibres et minimiser la coupure de fibre en longueur. Une telle pratique pourrait certainement améliorer les liens inter fibres et la qualité des pâtes.

D'autres efforts génétiques tels que la technique de clonage, croisement de différentes espèces d'arbres seraient favorables pour l'exploitation de pin gris. Ces pratiques ont pour but de diminuer ou de retarder la production de BE à paroi plus épaisse ainsi que diminuer la teneur en matières extractibles.

List of Contents

List of Figures

List of Tables

List of Equations

Abbreviations

APMP	Alkaline-peroxide mechanical pulping
CML	Compound Middle Lamella
CTMP	Chemithermomechanical Pulping
DDJ	Dynamic Draining Jar
DP	Degree of Polymerization
ESCA	Electron Spectroscopy for Chemical Analysis
EW	Earlywood
FC	Form Circle
FQA	Fibre Quality Analyzer
FS	Form Shape
IN	Inter-cell
IW	Intra-wall
LCC	Lignin-Carbon-Hydrogen Complex
LW	Latewood
RBA	Relative Bonded Area
RMP	Refiner Mechanical Pulping
SEC	Specific Energy Consumption
SEM	Scanning Electron Microscope
SG	Stone Grinding
SV	Specific Volume
TEM	Transmission Electron Microscope
TMP	Thermomechanical Pulping
TW	Trans-wall
WRV	Water Retention Value
XPS	X-ray Photoelectron Spectroscopy

Chapter 1 - Introduction

1.1 Background

Wood is a heterogeneous material in which the chemical and physical characteristics vary between species, between trees of the same species, and even between different parts of the stem within a tree. However, in temperate zone, the most distinguishable variability in fibre morphology exists between early- and latewood of softwood. Generally, earlywood (EW) fibres have larger outer perimeter, thinner cell wall and larger radial width compared with those of latewood (LW) [1, 2]. These differences affect not only fibre properties, such as density [3, 4] and mechanical strength [5, 6], but also the chemical [7, 8] and mechanical pulping [9, 10], and paper properties [10].

EW fibres are more flexible [11, 12, 13], and collapsible [6, 14, 15, 16, 17] than those of LW; the latter being stiffer and less collapsible. In refining the LW fibres exhibit a greater reduction in cell wall thickness. What's more, the EW fibres tend to split especially in the 1st stage refining [9], and they require more refining energy to reach the same freeness as compared to the LW fibres [10]. These findings indicate that EW and LW respond differently to mechanical actions. Due to this dissimilarity, the property of the final pulp or paper would depend on the proportion of EW and LW fibres in the furnish. To minimize the impact of the differences in refining between the EW and LW, some suggestions [18, 19, 20] have been made to separately refine EW and LW. However, this approach is not practical since EW and LW are naturally and intimately grown together, making it economically and technically difficult to achieve a clear separation of the two growth tissues.

In view of the variability of wood and to gain a better understanding on the breakdown mechanism of EW and LW in refining, this proposed research has been initiated to examine the influence of various fibres properties, physical and chemical, on refining. The possible different behaviour of EW and LW (e.g. fibre splitting, cell-wall thickness reduction, collapsibility, etc.) in refining will be investigated.

1.2 Characteristics of EW and LW

In the temperate region, when the atmospheric temperature rises in the spring tree growth activities begin with cambium cells division. This activity is believed to be regulated by a growth hormone, auxin. The early growth is fast and gives rise to fibres or tracheids in conifers (softwood) with large diameter and relatively thin cell wall. This wood tissue is called springwood or EW. As the late summer approaches the temperature falls and the tree growth gradually slows down, producing fibres with smaller cell lumen and thicker cell wall. This zone of thick-walled fibres is called summerwood or LW. The tree growth season ends in the fall as the trees begin to shed leaves or needles. The most visible differences between the EW and LW fibres are that the former tend to have larger diameter, thinner cell wall than the latter. Other differences can be classified into three categories: fibre morphology, physical property and chemical composition, as described in the following paragraphs.

1. Morphology of fibre

Wood fibre has a hollow structure with closed ends. The fibres are connected by a lignin-rich middle lamella (ML). Fibre wall itself is composed of a primary wall (P) and a secondary wall. The latter has three distinct layers: S1, S2 and S3. Among these layers, S2 is the thickest (1-5 μm), while the S1 and S3 are similar in thickness (0.1-0.2 μm). The three layers of the secondary wall are constituted of several laminas of microfibrils. The orientation of the microfibrils is different in each layer. The S1 and S3 tend to have larger fibril angles while the S2 has a relatively smaller angle [21]. Due to the dramatic change in fibril angle between S1 and S2, the interface between them constitutes a weak zone [22]. As such, the S1 layer or S2 layer would be exposed under the influence of mechanical refining, allowing a better development of fibre properties.

In comparison with thin-walled EW fibre, LW fibre would have a larger proportion of S2 layer [23] due to its thicker cell wall. But the fibril angle of the S2 layer is greater for EW fibre than for LW fibre [24]. EW fibre is relatively shorter but has larger cell lumen compared to LW fibre [25]. In addition, EW fibre has larger pit surface and effective pit pore radius compared with the LW fibre [26, 27, 28]. This suggests that EW has higher

permeability, and is more favourable to water and chemical transfers. The differences in morphology render EW fibre more collapsible under mechanical action, improving the fibre bonding in paper.

2. Physical property

The inherent structural organization of wood plays an important role in determining its physical properties. In refining, it is of great importance to understand the mechanical behaviour of the wood structure under the influences of various types of mechanical stresses, such as compression, tensile, shear, and torsion. Mechanically, EW fibre shows greater flexibility [11], elongation [29], collapsibility [14, 15] when compared to that of LW, mainly due to its thinner cell wall and larger lumen. On the contrary, LW fibre is stiffer [10], more brittle [30]. Consequently, the EW fibre is more vulnerable to longitudinal splitting, causing trans-wall failure. On the other hand, LW shows mostly intra-wall failure due to its thick cell wall [31].

3. Chemical composition

Basically, wood is made of three natural polymers: (1) cellulose, a linear and partly crystallized macromolecule, (2) amorphous molecules branched called hemicelluloses, and (3) lignin, a cross-linked amorphous polymer. Besides, there are some minor components, such as extractives and ash. The distribution of these chemical components seems to be different in EW and LW. For instance, the concentration of lignin in EW is higher than that in LW while the reverse is true for the concentration of holocellulose [32,33]. These differences could have some effects on the chemical pre-treatment of wood chips.

In sum, the differences between EW and LW in terms of morphology, physical and chemical characteristics are evident. These differences could influence their responses to refining actions.

1.3 Response of EW and LW to refining

The mechanical pulping industry (e.g. chip refining) has made great progresse during the last three decades or so [34]. This industry has developed various processes: refiner mechanical pulp (RMP), thermomechanical pulping (TMP), chemithermomechanical pulping (CTMP), and alkaline-peroxide mechanical pulping (APMP) to fulfil diverse needs of consumers. The main flowsheet of the various processes is similar as shown in Figure 1.1 [35]. There are, however, some variations in pre-treatment of chips or in the refining stage. According to the flowsheet, the main structural changes of wood chips take place in the pre-treatment and refining (including 1st and 2nd refining stages) stages. In this project, the study will focus on the behaviour of EW and LW in the 2nd stage.

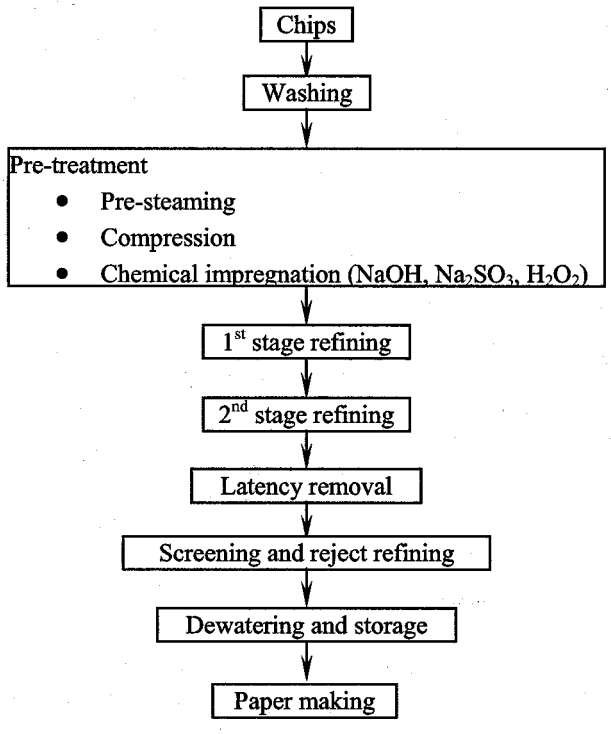

Figure 1.1 Typical flowsheet of refiner pulping

1.3.1 Response to pre-treatment

The aim of pre-treatment is to change the physical properties or chemical structure of wood chips, to reduce the specific energy consumption (SEC) and to improve the pulp

properties. There are various measures to fulfil these requirements, such as treating the chips with mechanical compression, thermal softening, enzyme, and chemicals before refining [36]. Among these measures, the mechanical compression and chemical treatment have particularly received much attention for their efficacy.

The purpose of mechanical compression of wood chips is to de-structure the wood matrix, reducing their particle size and thus increasing their specific surface. This has two main effects: first, it lowers the subsequent refining energy requirement, and second, it improves the chemical impregnation efficiency. In the refining industry, chip compression is achieved by means of a plug screw. However, static compression [37] can also used with less fibre damaged. But its industrial application has yet to be implemented.

On the other hand, the main purpose of chemical treatment is to degrade or soften the lignin, facilitating fibre separation. As a result, more intact fibres can be preserved (longer average fibre length) and the fibre flexibility can be improved (increased fibre conformability in paper). Common impregnation chemicals include sodium hydroxide, sodium sulphite, and hydrogen peroxide. Since EW fibre has thinner cell wall and bigger pit area, it has greater permeability for chemical agents when compared with LW fibre. Consequently, the chemical reaction is expected to be more efficient in EW.

1.3.2 Behaviour in refining stage

According to the structural changes of wood fibres, refining can be classified into two concurrent stages: fibre defibration and fibre development [38, 39,40]. In refining, the wood chips first break down into individual fibres and undefibrated fibre bundles (shives). The fibre defibration stage consumes relatively little energy [41]. In the fibre development stage, the primary wall (P) and S1 layer of the secondary wall are removed, and parts of the S2 are stripped off [9]. The reduction in fibre wall thickness decreases fibre stiffness. These physical changes improve inter-fibre bonding.

Due to their differences in morphology, the EW and LW fibres behave differently in refining. Studies [1, 42] indicated that cell wall splitting occurs principally in EW fibres,

particularly in the first stage of refining, and wall thickness reduction takes place more often on the thick-walled LW fibres. Moreover, EW fibre tends to change its form easily because of its greater compressibility and flexibility. Conversely, the LW fibre is more resistant to the refining action including flexion and kneading, which makes it difficult to change its cross-sectional shape. It was reported that EW absorb energy easily and require more energy to reach the same freeness in comparison to LW [9]. EW tends to break into fragments of irregular forms and sizes (shives) while LW disintegrates into slender bundles or individual fibres [2].

1.3.3 Influences on paper property

Fibre morphology plays an important role in determining the properties of paper. Studies [43, 44] suggested that EW fibre increases some physical strengths, such as density, tensile and burst strength, while the stiffer LW fibre produces bulky sheet with rough surface. Since EW fibre is much more flexible and conformable than LW fibre, it has larger bonding surface, giving better paper strength. However, LW fibre improves tearing resistance.

In summary, the different responses of EW and LW to refining are closely related to their differences in morphological characteristics. Due to these differences, the final pulp quality is dependent on their proportion in wood. This study will examine their behaviour in greater depth, hoping to further advance the understanding of the refining mechanism.

1.4 Objective of research

The main objective of this study was to investigate the mechanism of mechanical failure of EW and LW in relation to their morphology, physical and chemical properties in refining. More precisely, the following aspects were examined:

1) Difference between EW and LW (morphology, physical property and chemical composition);
2) Response of EW and LW in refining;

3) Influence of EW and LW on sheet properties.

Chapter 2 - Physical structure of wood

Woody plants are classified into two main groups, coniferous and deciduous species [45]. The coniferous are also commonly called softwood species and the deciduous hardwood species. The use of the terms of softwood and hardwood is not necessarily related to the hardness of wood. Softwood and hardwood are anatomically different [46]. Within the scope of this study, only the characteristics of softwood will be discussed.

In the temperate zone, the most distinguishable variability of softwood is the transition of EW to LW within a growth ring. The differences of EW and LW are responsible for their different behaviours in refining, and also in the end product properties. In this chapter, the definition, formation and physical structure of EW and LW are discussed.

2.1 General definition of EW and LW

Within a growth ring, the portion of the growth formed in the early season or spring is commonly designated as EW (also called springwood). While the denser and hence frequently darker wood produced later in the growing season/summer is termed LW (or summerwood). EW cells have relatively thinner cell wall when compared with those of LW (Figure 2.1). As a general rule, the difference between EW and LW in hardwood is not as distinguishable as that of softwood [47].

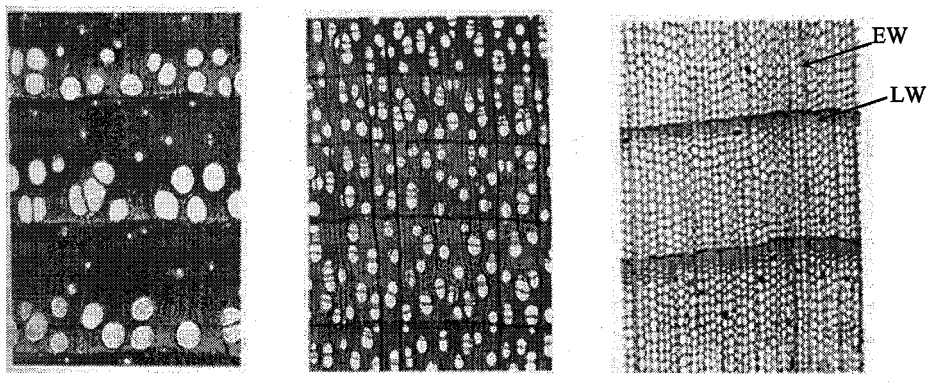

Figure 2.1 EW and LW within hardwood and softwood growth rings [46]
(Left: ring-porous hardwood, Centre: diffuse-porous hardwood, Right: softwood)

2.2 Formation of EW and LW

Studies [48, 49, 50] indicated that the presence of different concentrations of auxin (a growth regulator or hormone) in different stage of growing season was the main factor controlling the formation of EW and LW. The cell division is faster in the early season due to its higher level of hormone in the cambial zone. The fast growth produces cells with large radial diameter, wide lumen and thin walls. While the level of hormone decreases later during the growing season, cell division slows down and the cell diameter gradually decreases while the cell wall increases in thickness. This wood tissue is called LW. Besides the effect of auxin, other studies [47] indicated that the environmental factors, e.g., nutrients, water, temperature, and light, also have indirect influences on the developments of EW and LW.

2.3 Macro-structure of wood

In macroscopic level, a wood block can be characterized by three planes: the transverse plane (cross-section) (X), radial plane (R), and tangential plane (T), as shown in Figure 2.2.

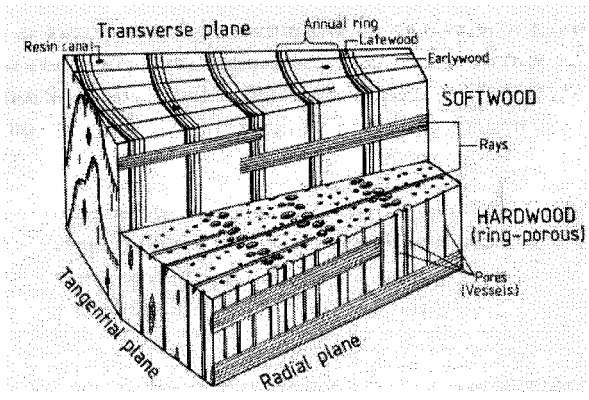

Figure 2.2 Macrostructure of softwood and hardwood [45]

Generally, hardwood is more porous than softwood because of the presence of vessels in the former. The resin canal can be seen in most softwood species [46] while the pores (vessels) are the feature of hardwood (Figure 2.2). In the temperate zone, trees form annual rings (growth ring) that are visible in cross-section as concentric rings. In most softwoods, the successive rings are distinguished by the darker colour of the wood

produced during the latter portion of the growth season. While in hardwood, the size and arrangement of "pores" (vessels) separate the rings from one another [51].

2.4 Micro-structure of wood

In this part, micro-structure of softwood and hardwood are compared. The micro-structural differences of EW and LW within softwood growth rings are also discussed.

2.4.1 Micro-structure of hardwood and softwood

Microscopically, hardwood and softwood consist of different type of cells, as indicated in Table 2.1.

Table 2.1 Cell-types of hardwood and softwood [46]

Hardwood	**Softwood**
Longitudinal fibre and vessel elements	
Vessel element Fibre tracheids Libriform fibres (Vascular, vasicentric) tracheids	Tracheids Resinous tracheids Strand tracheids
Longitudinal parenchyma element	
(Strand, fusiform, epithelial) parenchyma	(Strand, epithelial) parenchyma
Horizontal fibre elements	
None	Ray tracheids
Horizontal parenchyma elements	
Ray parenchyma (upright, procumbent cells) Epithelial parenchyma	Ray parenchyma Epithelial parenchyma

The main cell types in softwood are longitudinal tracheids, which make up approximately 90% by volume of wood. The rest are brick-like, short parenchyma cells and ray tracheids. Compared with softwood, the structure of hardwood is more complicated, it may contain several cell types: libriform fibres, fibre tracheids, vessel elements and parenchyma cell. In most hardwood species, the fibre occupies 40-75%

[52] of wood volume, while the vessels 10-40% [47]. Some characteristics of the major cell types in softwood and hardwood are listed in Table 2.2.

Table 2.2 Cell dimension of hardwood and softwood [53]

Cell type	Orientation[a]	Main Function[b]	Amount in Xylem[c], % by vol.	Length, mm	Radial width[d], μm
Softwood					
Tracheids	V	S,C	90	1.4-6.0	20-50
Ray tracheids[e]	H	C	< 5		
Ray parenchyma	H	ST	< 10	0.01-0.16	2-50
Epithelial parenchyma	V,H	E	< 1		
Hardwood					
Fibres[f]	V	S	55	0.4-1.6	10-40
Vessel elements	V	C	30	0.2-0.6	10-300
Longitudinal parenchyma	V	ST	< 5	< 0.1	< 300
Ray parenchyma	H	ST	15		

[a] Orientation of cell's major axis in the tree; V: vertical (longitudinal) and H: horizontal (radial).
[b] S support, C conduction, ST storage, and E excretion of resin.
[c] Average value depends greatly on tree species.
[d] Typical range, depends greatly on tree species.
[e] Absent in some species.
[f] including all fibre-like cells and tracheids.

In this study, the research scope is limited to softwood tracheid. For convenience, softwood tracheid is also called softwood fibre in this report.

2.4.2 Micro-structure of EW and LW

Due to their different stages of formation during the growing season, the EW and LW have different morphological characteristics, such as fibre length, cell width, cell wall thickness, and pit size, etc.

- Fibre length

Within a growth increment (growth ring), the fibre length of EW is different from that of LW (Figure 2.3). The figure indicates that: within a growth ring, the fibre length decreases sharply at the beginning of the formation of EW, then gradually increases

from EW to LW region. Generally, the softwood fibre length of EW ranges from 2 to 4 mm while that of LW from 3 to 6 mm [45].

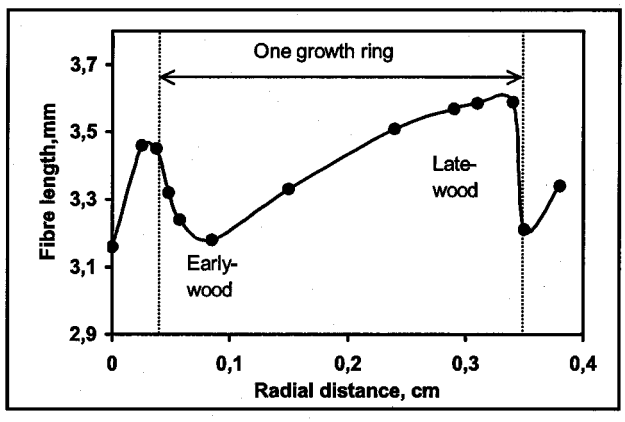

Figure 2.3 Fibre lengths across one growth increment of *Pinus Radiata* [54]

- Cross-section dimension of fibre

The most apparent morphological differences between EW and LW fibres are in their cross-section dimensions, such as cell wall thickness, radial lumen width and radial cell width (Figure 2.4). Generally, LW fibres have thicker cell wall and narrower radial width when compared to EW fibres. Thus, EW fibres have comparatively larger cell wall perimeters and smaller cell wall areas (Table 2.3).

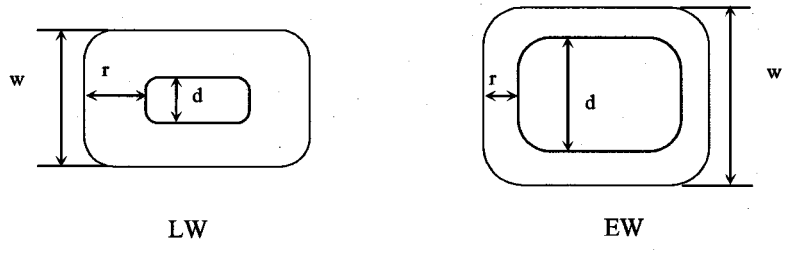

Figure 2.4 Cross-section of EW and LW tracheid
(w: radial cell width, r: cell wall thickness, d: radial lumen width)

Table 2.3 Cross-section dimensions of softwood fibre [55]

	Outer perimeter μm	Cell wall thickness μm	Cell wall area μm^2
EW	130-140	1.7-2.2	200-260
LW	100-112	4.1-6.2	300-450

When observing the cross-section of a growth ring in a kind of hybrid larch, Law [2] discovered that there are still some variations of cell diameter and cell wall width between EW and LW fibres in different direction (e.g. radial and tangential). Figure 2.5 shows that the tangential fibre width varied little throughout the growth ring while the radial width decreased gradually from EW to LW in the later part of the growing season. The double-wall thickness of EW fibres was relatively constant, about one fourth of that for LW fibre.

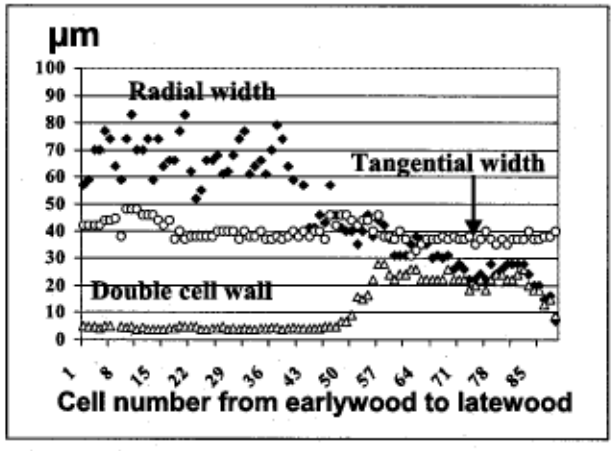

Figure 2.5 Variation of cell diameter and cell wall thickness within a growth ring [2]

- Pit structure

Wood fibres are oriented vertically and inter-connected by small openings (pits) in the cell wall. A pit in an adjacent cell wall usually forms a pit pair. Pit pairs can be classified into three types: simple pit pair, bordered pit pair and half-bordered pit pair, as showed in Figure 2.6. In wood anatomy, pits can be useful indexes in wood identification due to their differences in structures in different wood species [56]. Pits in wood are served to

allow conduction of water and inorganic nutrients from the roots throughout the tree; they also serve as passageways for the impregnation of wood with pulping liquor [47].

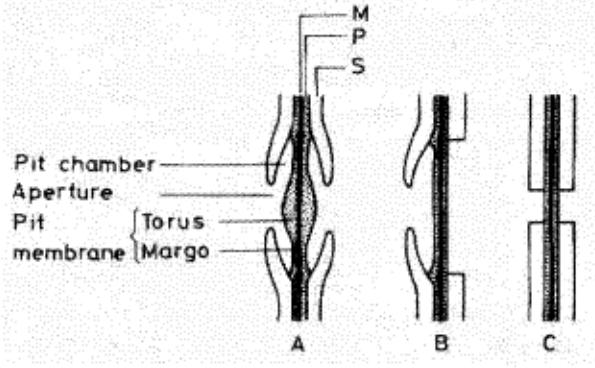

Figure 2.6 Profiles of pit pairs [57]
(Bordered (A), half-bordered (B), and simple (C) pit pairs.
M = middle lamella, P = primary wall, S = secondary wall)

Pits structures in early and LW are different. When compared with LW, EW fibres have more pits, larger pit surfaces and more effective pore diameter [26, 27, 28] (Figure 2.7). In the study of ultrastructure of Jack pine pit, Yang and Benson [58] indicated that EW pit diameters (16-20 μm) are greater than those of LW (12-14 μm). Some researchers [59, 60] reported that EW fibres have more weak spots at pits than in LW, which suggests that EW fibres could disintegrate and split more easily in refining when compared with LW fibres. Note that the ray cross-field area is larger in EW than in LW, as showed in Figure 2.7.

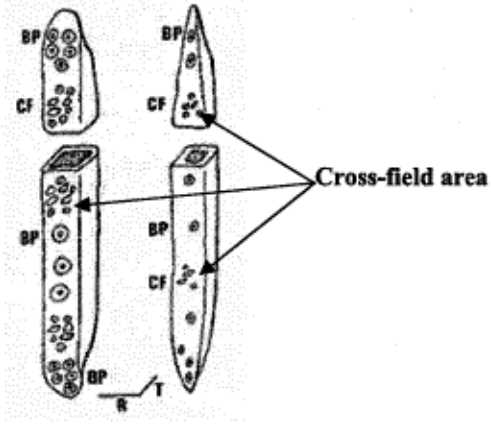

Figure 2.7 Diagrams of EW and LW fibres of southern yellow pine [61]
(EW (left), LW (right)
BP: interfibre bordered pits; CF: ray cross-field area)

2.4.3 Ultra-structure of EW and LW fibres

The ultra-structure of a wood fibre can be visible with the aid of high magnification microscope, e.g. scanning electron microscope (SEM), transmission electron microscope (TEM).

Essentially, the cell wall of a softwood fibre consists of concentric layers, namely, a primary wall (P) and a three-layered secondary wall (S1, S2 and S3), as illustrated in Figure 2.8. The lignin-rich middle lamella (ML) glues the fibres together. The combination of middle lamella and primary wall is called compound middle lamella (CML).

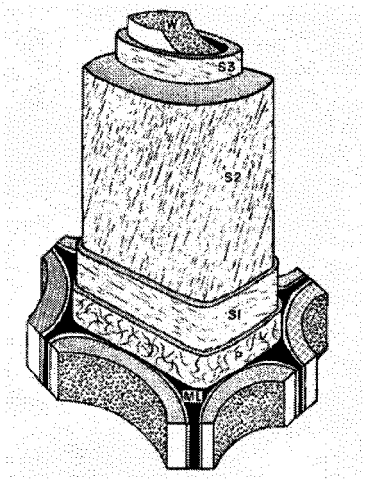

Figure 2.8 Typical cell wall structure of softwood fibre [62]

The primary wall has randomly oriented microfibils. The three layers of the secondary wall have different fibril angles: S1 and S3 having large fibril angles while S2 has a small fibril angle to the fibre axis [22]. Among these three secondary layers, the S2 is the thickest, representing about 70% of the total volume of the cell wall material, while the S1 and S3 are relatively thinner, as indicated in Table 2.4. For this reason, the characteristics of S2 layer exert a major influence on the properties of the entire cell wall. The structure of the cell wall, together with the changes, which occurred in various process stages (cooking, refining, etc.) determine the swellability, conformability, and thus the physical properties of paper [52].

Table 2.4 Physical structure of fibre wall layers [63]

	Fibril angle °	Thickness μm
ML	-	0.1-0.2
P	irregular	0.1-0.2
S1*	50-70	0.2-0.3
S2**	10-30	2.0-4.0 [64]
S3*	50-90	0.1

* The microfibrils in S1 layer form either Z or S helix.
** The microsfibrils in S2 layer form both Z and S helices.

The ultra-structure of EW fibre is different than that of LW fibre, such as the thickness of secondary layers and fibril angles (Table 2.5). It can be noted that the most apparent differences of cell wall ultra-structure between EW and LW fibres are concentrated on the S2 layer thickness and fibril angle. LW tends to have relatively thicker S2 layer and smaller S2 fibril angle. Doubtlessly, these differences will influence their behaviours in pulping and papermaking.

Table 2.5 Comparison of EW and LW cell wall ultra-structure

	EW		LW	
Cell wall [65]	thickness μm	percentage of total wall %	thickness μm	percentage of total wall %
P	0.09	4.3	0.09	2.1
S1	0.26	12.4	0.38	8.8
S2	1.66	79.0	3.69	85.8
S3	0.09	4.3	0.14	3.3
Total wall	2.10		4.30	
Fibril angle, °				
S2 [24, 66]	20-38		10-28	

2.5 Anatomical definition of EW and LW

To characterize wood properties, it is desirable to define the distinction between EW and LW. In the past, some definitions had been put forward basing on the anatomical differences between the EW and LW, especially the cross-section dimension. Some commonly used definitions are described below.

- Mork's rule [67]

A Norwegian wood anatomist, Mork, defined LW fibres as those that have a double cell wall thickness equal to or greater than the radial lumen width, as illustrated in Figure 2.4. However, in some species the transition from EW to LW is relatively gradual. In this case there exists a transition zone where some fibres do not meet the requirement of the Mork's rule and yet they possess relatively thick cell wall and have LW-like mechanical properties.

- Parameter *Z* [68]

Parameter *Z* is defined as the cross-section fibre wall area divided by the area of a circle with the same outer perimeter (Eq. 2.1).

$$Z = \frac{4A_w \times \pi}{P^2} \qquad \text{Eq. 2.1}$$

A_w = cross-sectional fibre wall area

P = fibre outer perimeter

The value of the parameter *Z* ranges from 0 to1. A value close to 0 would indicate an EW fibre; conversely a value near 1 indicates LW fibre. The drawback of the Parameter Z is that there is no definite value defining distinction between the EW and LW.

- Mohlin's *F/L* ratio [69]

Mohlin used a ratio of outer cell wall perimeter of fibre (*F*) and outer lumen perimeter (*L*), *F/L*, to indicate the amount of LW fibres. The larger the ratio indicates the more LW fibres in the sample. On the contrary, smaller ratio represents more EW fibres. To be useful it requires knowing the upper value for 100% LW fibres and the lower value for all EW fibres. As in the case of the Parameter *Z*, there is no clear demarcation between EW and LW fibres. However, it does give an indication of fibre quality of a pulp furnish.

In reality, as indicated earlier, in most species there is a transition zone between EW and LW (Figure 2.1, Figure 2.9) within a growth ring. None of the above definitions has taken this into account, suggesting the weakness of these definitions. However, the

Mork's rule, which is commonly accepted for wood characterization, does provide a clear distinction between the EW and LW. Hence, this definition will be employed in this study.

2.6 Other physical properties of EW and LW

As mentioned earlier, the transition from EW to LW may be gradual or abrupt (Figure 2.9). Additionally, the volume ratio of EW to LW varies depending on species, as indicated in Table 2.6. Due to its thicker cell wall, LW has higher specific density when compared to EW (Table 2.7). Consequently, higher percentage of LW donates higher specific gravity of wood [47].

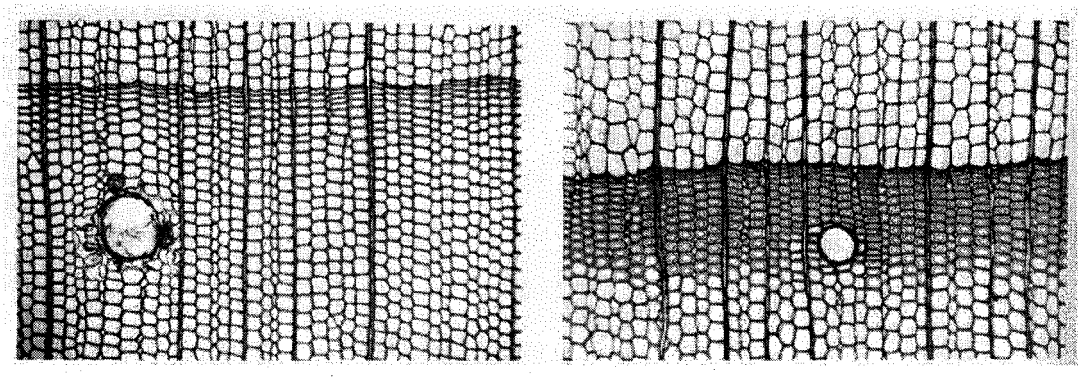

Figure 2.9 Transition from EW to LW [46]
(Left: Gradual transition (western white pine), Right: Abrupt transition (western larch))

Table 2.6 LW percentage in selected Canadian softwood (by volume)

Species	LW %	Density	Basic density, kg/m3	Reference
Black spruce	20	0.40	400	70
Red cedar	16	0.31	310	71
White spruce	25	0.35	350	71
Alpine fir	24	-	-	71
Western larch	39	0.53	530	71
Douglas fir	45	0.46	460	71

Table 2.7 Specific gravity of EW and LW in some softwood species

	Specific Gravity	
	EW	LW
Douglas fir [72]	0.216	0.626
Redwood [73]	0.259	0.583
Shortleaf pine [74]	0.429	0.858
Norway spruce [74]	0.298	0.609
Scots pine [74]	0.316	0.763

2.7 Summary

From the morphological differences between EW and LW discussed above, the amount of LW in wood constitutes an important parameter determining not only the mechanical properties of the wood itself but also the processing of wood, for example mechanical pulping, and the characteristics of the final product.

Chapter 3 - Chemical properties of wood

In this chapter, the properties of the major wood components (lignin, cellulose and hemicelluloses) are briefly discussed in respect to pulping and papermaking.

3.1 Chemical composition of wood

Wood has three main building components, lignin, cellulose and hemicelluloses. It also contains small amounts of extraneous materials such as extractives and ash. Softwood has different chemical composition compared with hardwood (Table 3.1). The distribution of the three major chemical components across the fibre wall is also different, as shown in Figure 3.1. The compound middle lamella (CML) is rich in lignin. The S2 layer, which is the thickest among the three secondary layers, is abundant with cellulose and hemicelluloses, while the relatively thinner S1 and S3 have less cellulose. The distribution of chemical components within the fibre wall could have great influences on chemical pulping (e.g. chemical penetration and deligninification rate) and mechanical pulping (e.g. fibre separation and fibrillation, etc.).

Table 3.1 Approximate chemical composition of softwood and hardwood [75]

	Constituent, %			
	Lignin	Cellulose	Hemicelluloses	Extractives [76]
Hardwood	20 ± 4	45 ± 2	30 ± 5	5 ± 3
Softwood	28 ± 3	42 ± 2	27 ± 2	3 ± 2

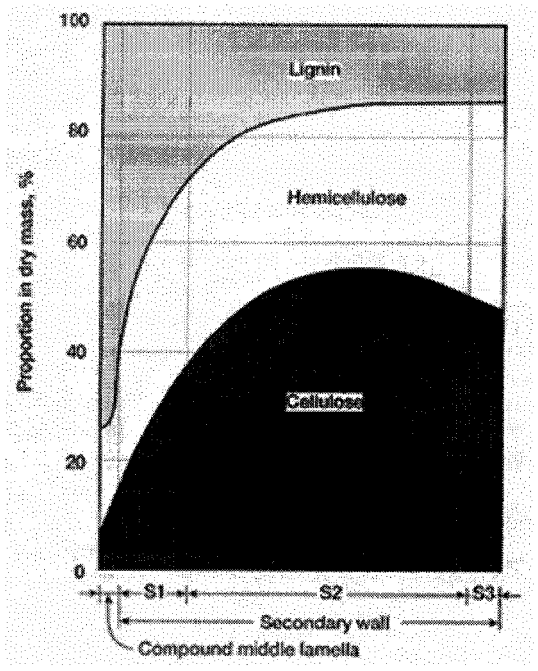

Figure 3.1 An approximate percentage distribution of the chemical components in different layers of softwood tracheid [47]

The distribution of chemical components is different in EW and LW fibre walls, as indicated in Table 3.2.

Table 3.2 Distribution of chemical composition in spruce tracheids [77]

Region	Cellulose	Hemicelluloses	Lignin
Wall layer	% of total cellulose	% of total polyoses	% of total lignin
EW compound			
CML	4.1	20.6	26.8
S1	8.9	23.2	10.4
S2+S3	87.0	56.1	62.8
LW compound			
CML	2.5	15.0	18.4
S1	5.2	15.6	7.9
S2+S3	92.3	69.4	73.7

When compared with EW, LW has higher lignin content in S layer (S1+S2+S3, 81.6%), but lower in CML (18.4%). Studies [32, 78, 79, 80, 81, 82] indicated that the total lignin

content (S+CML) is higher in EW as contrast to LW. LW has higher cellulose content due to its thicker and cellulose-rich S2 layer [33]. The distribution of hemicelluloses is uneven either in EW or in LW. According to the calculated results, the hemicelluloses content is higher in EW than in LW [83].

3.2 Chemical properties of major wood components

The general characteristics of cellulose, hemicelluloses and lignin, are discussed briefly in the followings.

Cellulose is the main component of wood fibre, located mainly in the S2 layer. Cellulose is a long-chain unbranched polymer composed of β-D-glucopyranose units linked together by (1-4)-β-glycosidic bonds. The degree of polymerization (DP) of wood cellulose is around 500-10000 glucose units, and its molecular weight is in the order of 600,000 [84, 85]. Cellulose molecules have a high tendency to form intra- and intermolecular hydrogen bonds and thus aggregate together into microfibrils [63]. Highly ordered (crystalline) regions alternate with less ordered (amorphous) regions in the microfibrils, and the crytallinity is around 60-70% [45]. The softening temperature of dry cellulose and in water-saturation state are 220°C and 20°C, respectively [35]. As such, these characteristics of cellulose have a strong influence on the fibre flexibility, physical strength, swelling and water absorption capacity.

The main softwood hemicelluloses are galactoglucomannan, arabinoglucuronoxylan, and arabinogalactan [52]. The softening temperature of hemicellulose under dry and water-saturated state is 180-220°C [86] and 20°C [87], respectively. Hemicelluose and cellulose are mutually attached by hydrogen bonds and Van Der Waals force. The lignin and hemicelluloses form a so-called lignin-carbon-hydrogen complex (LCC) [45, 63]. Generally, in refining, most of the hemicelluose is retained, improving pulp yield and fibre properties. As a hydrophilic polymer [88], hemicelluloses have a high ability to absorb water and swell. Thus, high hemicelluloses content improves fibre flexibility and increases the final sheet density [52].

Lignin is an amorphous cross-linked three-dimensional polymer (DP 20,000) with phenylpropane units [89]. The phenylpropane units are joined together with stable covalent bonds (ether and C-C linkages) and less stable hydrogen bonds. Lignin also contains functional groups such as methoxyl, phenolic hydroxyl, benzyl alcohol, and carbonyl groups. Functional groups are of major influence in the reactivity of lignin in chemical pulping. The typical softwood lignin is predominately guaiacyl type, whereas guaiacyl and syringyl groups are present in hardwood [52]. The phenylpropane units render lignin hydrophobic while its three-dimensional structure makes the cell wall resistant to external forces. The softening temperature of water-saturated lignin is 80-90°C. Additional water does not result in considerable further softening of the lignin. In mechanical pulping, the *in situ* softening temperature of lignin (as part of the wood fibre matrix) is in the range of 100-130°C [35]. Thus, refining at temperatures equal or higher than the softening temperature is beneficial to maintain fibre length.

3.3 Summary

Wood is mainly composed of lignin, cellulose and hemicelluloses. The distribution of these major components is different in EW and LW. This uneven distribution in EW and LW would influence their behaviours in pulping and papermaking.

Chapter 4 - Effects of EW and LW on pulping and papermaking

EW and LW fibres could behave differently in pulping and papermaking due to their considerable differences in physical structure and chemical composition. In this chapter, the effects of EW and LW on pulping, particularly refining, and their influences on paper properties are discussed.

4.1 Effects on pulping

There are two main types of pulping: chemical and mechanical pulping. Generally, the chemical pulping dissolves out the lignin, which binds the cellulosic fibres together while the mechanical pulping separates the fibres from wood chips or logs by mechanical forces with little loss of wood components. Evidently, the fibre characteristics are different for the chemical and mechanical processes. Due to their low yield, the chemical fibres are flexible and produce paper with excellent strength properties. Furthermore, because of the dissolution of lignin in chemical pulping, the process requires little mechanical forces to separate the fibres, minimizing fibre breakage. On the other hand, the mechanical fibres contain practically all the lignin component, rendering them stiff and yielding bulky paper with good optical properties [34]. Additionally, the mechanical fibres suffer severe fibre breakage, shortening the average fibre length. The behaviour of EW and LW in both mechanical and chemical pulping is discussed respectively in the followings.

4.1.1 Effects on mechanical pulping

Mechanical pulping can be classified into two processes: stone grinding (SG) and refining. In SG, debarked wood logs are forced against a revolving abrasive stone which grinds the fibres off the logs and release them from the matrix. The process is harsh and energy efficient [40]. In refining, wood chips are fed between two metal discs with at least one of them rotating. The wood fibres are separated by the action of grooves and bars on the surfaces of the two discs [34]. These two types of mechanical pulp have different properties: the SG pulp has a higher content of fines due to the abrasive action,

while the refiner pulp has a lower content of fines and its long fibres tend to be more ribbon-like [34]. Generally, the refiner pulp has better strength property than the SG pulp because of its greater average fibre length and external fibrillation.

Within the scope of this study, the research emphasis is on thermomechanical pulping (TMP). The mechanism of TMP and the variables affecting this process will be discussed. The behaviours of EW and LW in TMP will also be compared.

4.1.1.1 Refining mechanism

It is generally accepted that there are two concurrent stages in refining: a fibre defibration stage and a fibre development stage [38, 39, 40] (Figure 4.1). In the defibration stage, the chips are disintegrated into fibres and undefibrated bundles (shives). This stage requires relatively little energy [41]. The fibre development takes place through repeated compressive-shear action in the refiner [2]. This stage is the most important stage for the final pulp quality, and consumes most of the total energy input. In this stage, the middle lamella, primary wall and S1 are removed, and part of the S2 is stripped off [9]. These changes induce external and internal fibrillations. In addition, the refining action also shortens fibres [90]. As shown in Figure 4.1, the breakage of fibres into shorter fragments and fines (path I) and peeling off of cell wall (delamination) (path II) exist in the fibre development stage. According to Karnis [39], the path II dominates in this stage. Hence, refining changes the fibre length, specific surface and flexibility of the resultant fibrous elements.

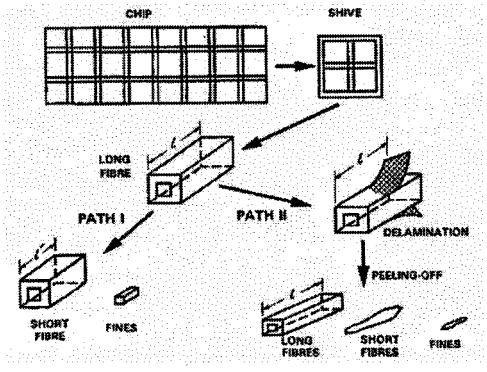

Figure 4.1 Mechanism of chip refining [39, 90]
(Path I: part of the long fibres is broken to shorter fibres and fines,
Path II: fibre wall material is first delaminated and eventually peeled off to form short fibres and fines.)

4.1.1.2 Variables affecting chip refining

In refining, the refiner design and process conditions determine the pulp quality (Table 4.1).

Table 4.1 Variables affecting chip refining [91]

Refiner design	Disc diameter, plate pattern, etc.
Process conditions	Pressure, temperature, consistency, refiner speed, residence time, etc.

The combination of all these variables results in different specific energy and refining intensity. For a given specific energy, an increase in rotation speed and a decrease in plate gap will lead to an increase in refining intensity, and a reduction in fibre length (Figure 4.2). Higher pressure and temperature reduce fibre shortening, and make fibres having less fibrillation. In addition, refining consistency also plays an important role in refining. A consistency range of 45-50% provides the maximum energy saving while producing high quality pulp [92].

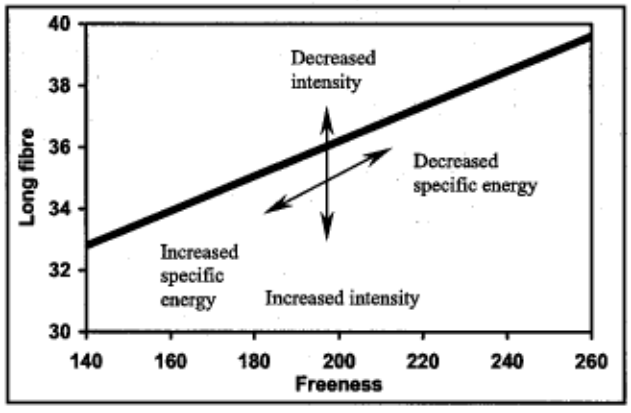

Figure 4.2 The relation between freeness and long fibre content [93]

4.1.1.3 Pre-treatment in refiner mechanical pulping

The purpose of mechanical pre-treatment of wood chips in refining is to change the physical structure and chemical properties of chips, aiming at reducing the specific energy consumption (SEC) and improving the pulp strength [94]. There are various measures to fulfil these requirements, such as chemical, mechanical and biological pre-

treatment of chips before refining [95, 96, 97]. Among these measures, the chemical and mechanical pre-treatments have received much attention and are proved to be effective.

As discussed in Chapter 1, the mechanical pre-treatment reduces the SEC through compression of wood chips before refining, while the chemical pre-treatment, especially sulfonation (Na_2SO_3) can change the modes of fibre rupture through lowering the softening temperature of lignin (Figure 4.3). The softening effect of sulfonation in CTMP facilitates fibre separation mainly in the middle lamella (ML), producing pulps with good fibre length. Using thermal softening such as in TMP also preserves respectable average fibre length by inter-cell (IC) separation. Trans-wall (TW) and intra-wall (IW) failures and fibre breakage are common fibre failures in RMP, where there is no chemical or thermal softening is used Thus, the chemical and thermal pre-treatments tend to better maintain fibre length in refining.

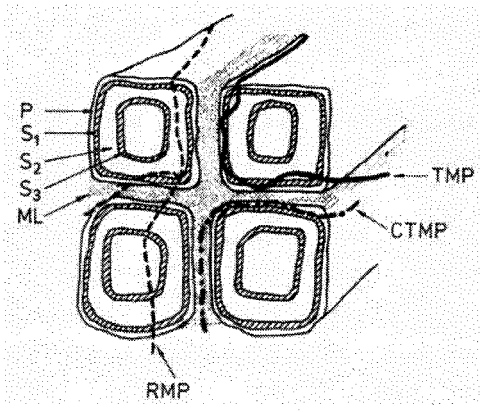

Figure 4.3 Typical ruptures in high-yield pulping processes [98]

4.1.1.4 Behaviour of EW and LW in refining

Due to their differences in physical and chemical nature, EW and LW could respond differently in refining, yielding various structural changes such as shape deformation, splitting, and energy absorption. The principal characteristics are discussed in the followings.

- Load deformation

Law [2] has done some research works about EW and LW deformation under different radial compression loads which were similar in the mechanical pre-treatment of refining. When pressed in radial direction, the specimen (wood block) showed an elastic characteristic in the early stage of deformation. As the load increased the elastic behaviour was followed by a plastic plateau which is characterized by the collapse of EW fibres, Figure 4.4 shows. When the strain further increased, the stress increased gradually, as observed earlier by Easterling et al. [99]. The area of rapid increases in stress is usually characterized as the densification zone [100, 101]. At this stage, LW fibres buckled and fractured. However, LW fibres showed little collapse even with an overall 70% deformation of specimen containing EW and LW when the compression was done without restraint.

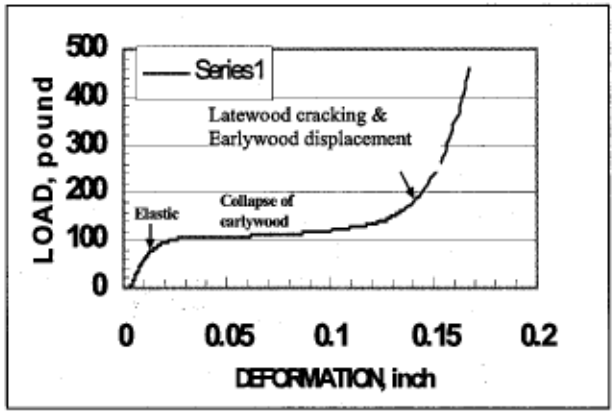

Figure 4.4 Load deformation curve for radial compression [2]

- Splitting

In refining, longitudinal splits occur in fibre wall. In such a case trans-wall failure is expected especially with thin-walled EW fibres, particularly in the first stage of refining where the fibres are subjected to severe mechanical actions [1, 9]. The comparatively lower elastic modulus (Young's modulus) of EW fibre is also considered responsible for such splitting [102, 103, 104]. The fibre splitting reduces the stiffness of mechanical fibres and thus improves the surface smoothness of paper [105, 106].

- Cross-section dimension

Under the influence of refining actions, the fibres change their cross-sectional dimension in terms of lumen shape and cell wall thickness. Having thin cell wall and large lumen, EW fibres tend to collapse easily (Figure 4.5).

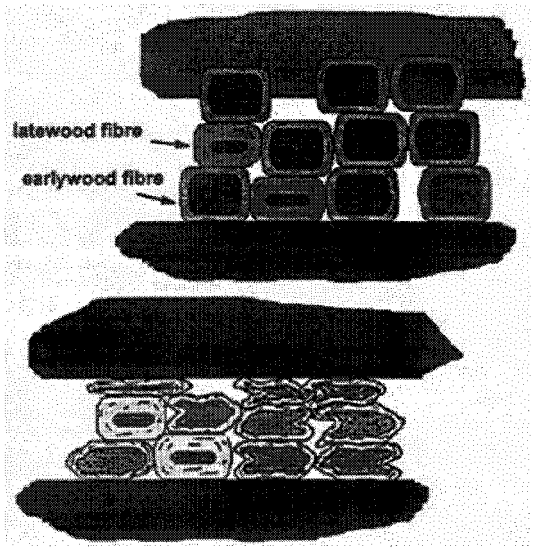

Figure 4.5 Collapse of EW and LW fibres in refining [19]

The degree of fibre collapse or deformation can be expressed in terms of Form Circle (*FC*) [1], as calculated in Eq. 4.1. The Form Circle is defined as the area of the fibre wall and lumen, divided by a circle with the same outer perimeter as the fibre's cross-section. Its values varies from 0 to 1, 1 being a circular cross-section.

$$FC = \frac{4\pi A_t}{P^2} \qquad \text{Eq. 4.1}$$

A_t = lumen area and cross-sectional wall area

P = outer perimeter of cell wall

Within a growth ring, EW fibres tend to have larger *FC* values than those of LW (Figure 4.6) since the former have relative thinner cell wall and larger lumen than the latter. Thus, in refining the collapsed EW fibres have smaller FC values and show better flexibility (Table 4.2).

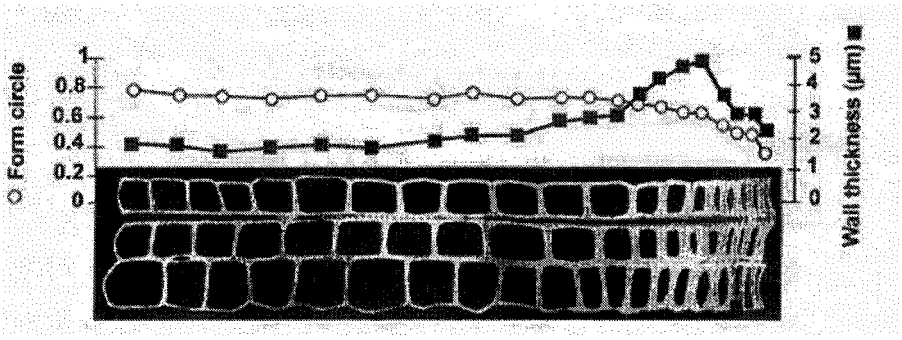

Figure 4.6 Form circle of EW and LW in a growth ring of Norway spruce [1]

Table 4.2 Form Circle values of EW and LW fibres in refining [1]

	After 1st stage	After 2nd stage
EW fibres	0.41±0.01	0.36±0.01
LW fibres	0.65±0.01	0.60±0.01

In addition, LW fibres have greater reduction in cell wall thickness than those of EW fibres (Figure 4.7) [1, 9]. Thus, fibrillation is more evident in LW fibres [12].

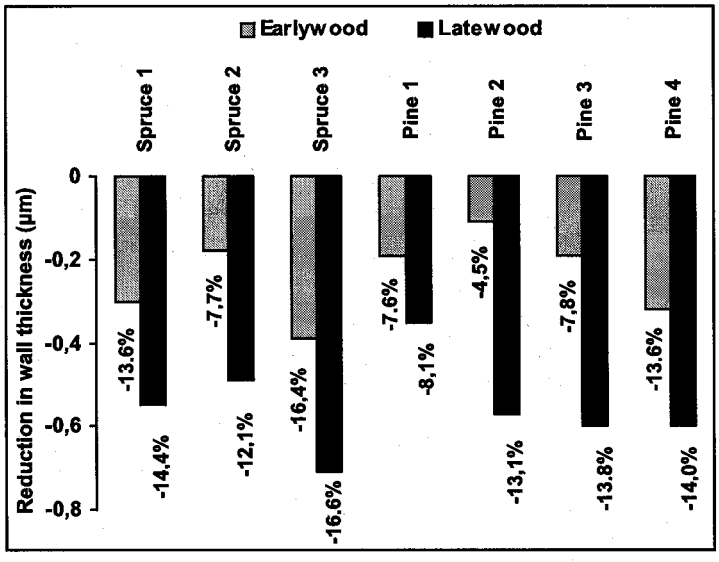

Figure 4.7 Reduction of fibre cell wall thickness in refining [9]
(The numbers below the bars show relative reduction in fibre cell wall thickness)

- Shives content

Cowan [107] proposed that a projected area of 0.7~1.25 mm^2 can be measured to indicate shives content. Some research work [108] indicated that LW defibrated more easily than EW. The thicker walled LW may yield TMP pulp with less shives content than the thinner walled EW [9]. Furthermore, Law [2] found that EW shives were much chunkier while those of LW were slender (Figure 4.8) in a kind of larch RMP pulp.

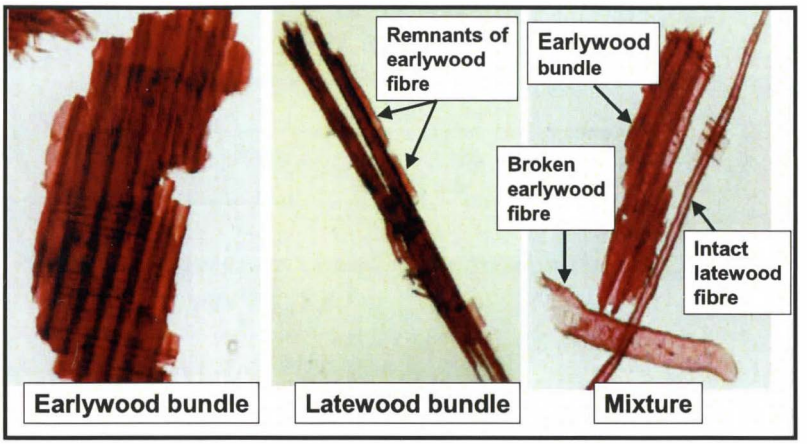

Figure 4.8 Typical shive in larch RMP [2]

- Fine content

The fine fraction (passed 200-mesh fraction of Bauer-McNett classification) is comprised of thin, ribbon-shaped lamellae from the fibre walls and flakes from the compound middle lamellae. It also contains short ray cells which have poor bonding properties. The fine particles have a great effect on the strength and drainage characteristics of mechanical pulp [34]. Some recent research works [10] indicated that thin-walled EW fibres produced more fines than the thick-walled LW fibres in high intensity refining.

- Energy absorption

EW fibres are more flexible and collapsible due to their large lumen and thin cell wall. As a result, in refining, they absorb more energy than LW fibres do [10]. A repetitive cyclic compression test (Figure 4.9) [109], which is similar to the refining action, indicated a rapid temperature rise in EW portion of the test specimen, confirming

preferential energy absorption by EW portion of the growth ring. The consequence is that EW fibres would yield to the refining actions more easily than those of LW and rupture more readily.

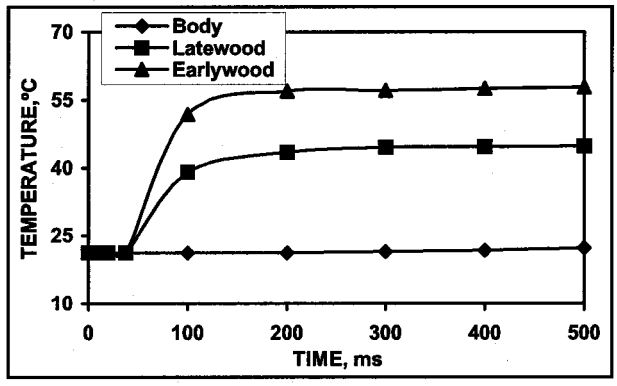

Figure 4.9 Temperature record of southern pine in repetitive cyclic compression [109]

4.1.1.5 Separation of EW and LW

Since EW and LW are naturally and intimately bonded together, it's difficult to examine each of their behaviour during refining. In order to further advance the understanding of their rupture mechanism in refining, some researchers suggested to separate EW and LW [18, 19, 20]. The separation may be conducted either before or after refining.

- Separation before refining

Murton et al [10] proposed to separate EW and LW by means of a veneer machine. As Figure 4.10 shows, 2-mm thick veneers are peeled off from the wood log by the machine. The LW was distinguished from EW by its darker colour. Then the two kind of wood veneers were chipped for refining.

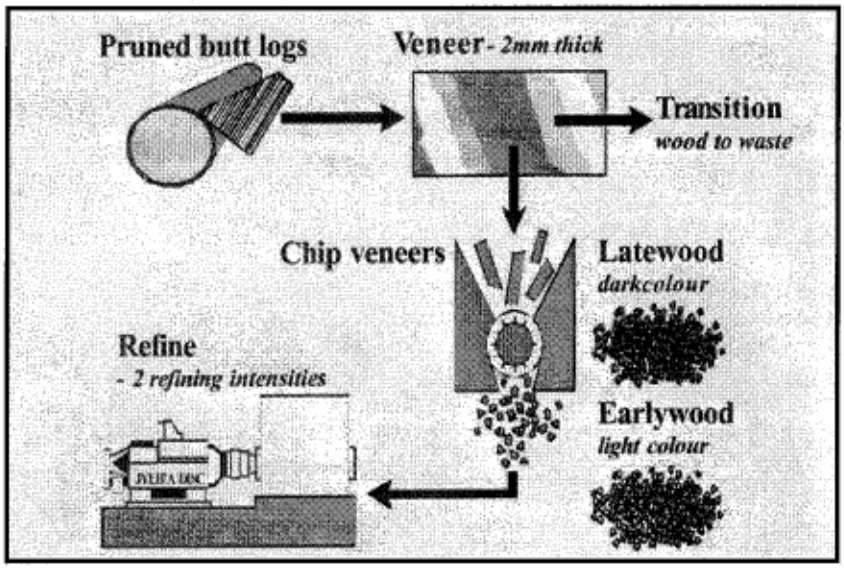

Figure 4.10 Separation of EW and LW before refining by veneer machine [10]

- Separation after refining

Kure [106] proposed to separate EW and LW after refining by a hydrocyclone (Figure 4.11). The separation is based on the fact that the thick-walled LW fibres have higher density than that of EW and they tend to travel faster to the periphery of the cyclone, and are discharged through the bottom orifice. On the other hand, the lighter EW fibres are discharged through the upper orifice of the cyclone.

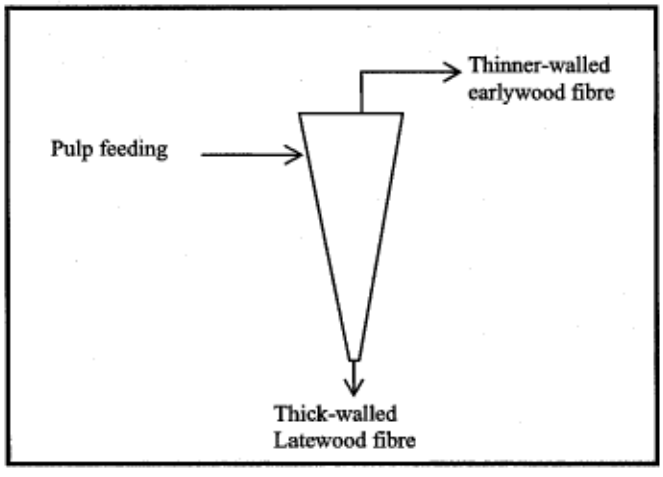

Figure 4.11 Separation of EW and LW after refining by hydrocyclone

In comparison the former technique is more efficient than the latter, but yield much more waste since most of transition wood are excluded. As for the latter one the efficiency of this method is doubtful because refining could minimize the differences in density between EW and LW fibres, complicating the separation.

4.1.2 Effects on chemical pulping

The differences in physical structure (pit structure, cell wall thickness) and distribution of chemical components between EW and LW are expected to have great influences on their behaviour in chemical pulping.

- Pit structure and cell wall thickness

Due to its greater numbers of bordered pits, larger lumen and thinner cell wall, EW fibres have high permeability in chemical pulping, which is favourable to water or chemical transfers. In contrast, LW fibres with thicker cell wall and less bordered pits area would have lower permeability. Therefore, these structural differences of EW and LW could induce heterogeneous kinetics transfer and chemical reactions in chemical pulping [28]. Studies indicated that EW fibres show better chemical absorption [7] and higher delignification rate [8, 110] than those of LW.

- Chemical distribution

In chemical pulping, most of the lignin is dissolved out while the cellulose is retained. As seen earlier, LW has higher cellulose content than EW. Consequently, it shows higher pulp yield than the EW when cooked under similar conditions [111, 112].

4.2 Influences on paper strength

The relations between the properties of raw material, pulp fibre, fibre network and paper are shown in Figure 4.12. The figure indicates that the thin-walled EW fibre favours fibre flexibility and collapsibility, which in turn increases inter-fibre bonding (e.g. the relative bonded area (RBA)) [113] and improves paper strength (e.g. tensile, and burst indices) [52]. On the other hand, the thick-walled LW fibre, which has smaller S2 fibril angle, would show greater Young's modulus, and thus greater stiffness. This

characteristic of LW fibre would contribute to bulky sheet and usually with better tearing resistance when compared to the thin-walled EW fibre. As shown in Figure 4.13, with LW content greater than 20% the bulk of paper increases and the fibre bonding ability decreases.

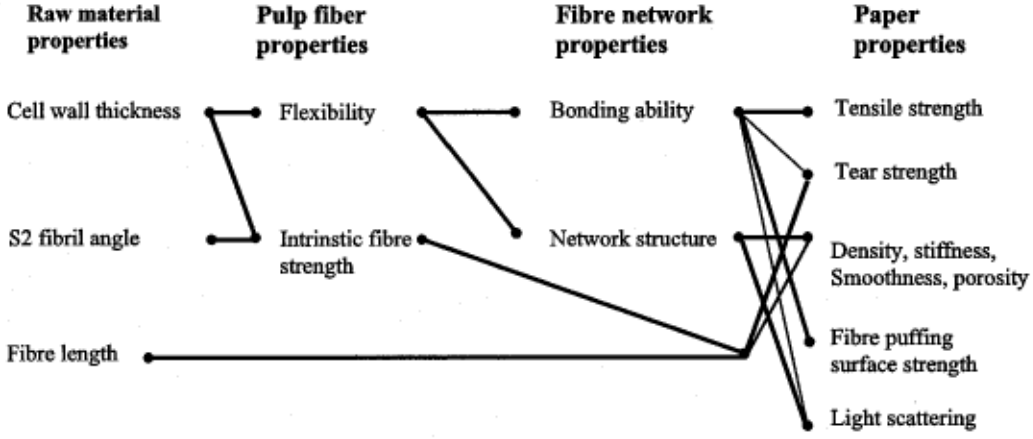

Figure 4.12 Influences of softwood properties on fibre network and paper properties [52]
(The main effects are presented as thick lines.)

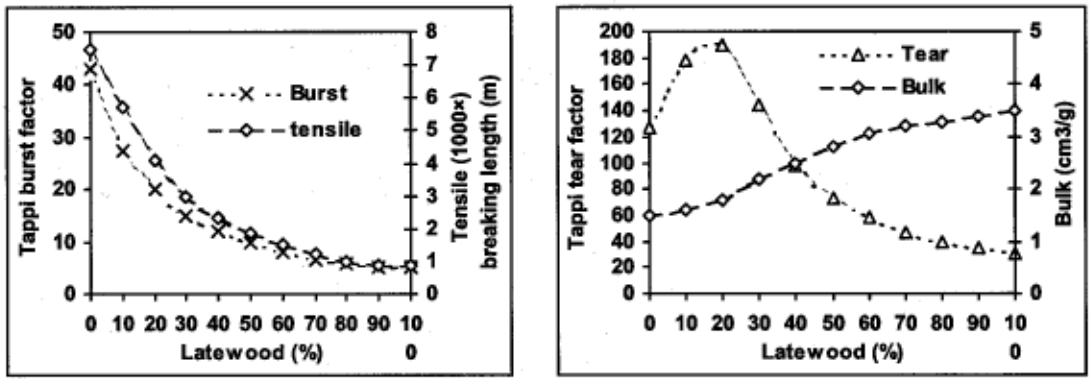

Figure 4.13 Physical properties of Douglas fir pulp vs. LW content [72]

4.3 Summary

Due to their differences in physical and chemical characteristics, EW and LW behave differently in pulping and papermaking. In refining, thin-walled EW fibres tend to split, collapse and absorb energy more easily than those of LW. They form fibrous network

with good strength properties. In contrast, the thick-walled LW fibres show greater reduction in cell wall thickness in refining, producing sheets with high bulk and tearing resistance.

Chapter 5 - Research proposal

5.1 Background

As discussed in the preceding chapters, the EW and LW have different morphological characteristics, such as cell wall thickness, fibre length, cross-section dimension, and pit size, etc. Previously reported research works [54] indicated that the fibre length decrease sharply at the beginning of the formation of EW, then gradually increase from EW to LW region. In cross-section, LW fibres have thicker cell wall and narrow radial width when compared to EW fibres. Furthermore, the pit structure of EW and LW are also different: EW fibres have more pits and their pits surfaces are larger and are more effective in liquid transportation in contrast to those of LW fibres.

Other previous studies also indicated that the differences in these structural characteristics of EW and LW have significant impacts on refining and papermaking properties.

In refining, the cell wall of EW fibres split readily due to its thinness; it cannot sustain the mechanical actions of the refiner bars. In contrast, the thick-walled LW fibres resist relatively well against the refining actions and they experience significantly higher degrees of reduction in cell wall thickness.

Again due to its relatively thin cell wall, EW fibre tends to change its form easily and shows greater compressibility and flexibility, as reported [19]. Conversely, thick-walled LW fibre is more resistant to such changes in refining action. We also learn from other research's works [2] that EW tends to break into fragments of irregular form and sizes (shives) while LW disintegrates into slender bundles of individual fibres. It has also been reported that EW absorbs energy more readily and requires more energy to reach the same freeness in comparison with the EW counterpart.

Additionally, EW and LW fibres also influence differently the paper properties due to their morphological discrepancies. For example, EW fibres give higher sheet density,

tensile and burst strengths, while the stiffer LW fibres produce bulky sheet with high surface roughness. However, LW fibres yield greater tearing resistances.

In spite of these findings, it remains unclear how these two types of wood tissue are transformed from solid wood into individual fibrous elements and in what manner they contribute to the sheet characteristics. We hope to shed some light on these aspects in the present investigation. In fact, although some progresses have been made in the understanding of the refining mechanism, there is little published information regarding the breakdown mechanism of EW and LW in refining, especially in thermomechanical refining. Their responses to various refining temperature, modes of rupture (inter-cell, trans-wall and intra-wall), energy consumption, etc. are yet to be elucidated. Up to date, little is known about the relations between the morphological changes of EW and LW in refining and their influences on paper properties. In this study, systematic research procedures are proposed, including the separation of EW and LW, thermomechanical refining at various temperatures, and handsheet characterization. A series of physical and chemical analyses are conducted in each experimental step. Evidently, with the results obtained from this research, we could gain better understanding of the mechanical failures of these two types of wood tissue and will also certainly help improve the quality of the pulps produced from jack pine, an under-exploited and less desirable species among the Canadian forest resources. The expected results would also help us ameliorate the refining efficiency.

5.2 Objective and approaches

The principal objective of this research project is to study the breakdown mechanism of EW and LW in thermomechanical pulping (TMP) by means of microscopic observations. Characteristics such as fibre splitting, shortening, delamination (internal fibrillation) and external fibrillation will be evaluated. Physical changes in EW and LW fibres will be qualified and quantified by means of light microscopy as well as scanning electron microscopy. The impacts of the observed changes on pulp and paper properties will be assessed to establish possible interrelation between the fibre characteristics and paper properties.

The main points of the study are as follows:

1. Identification of EW and LW pulp fibres according to their morphological differences (cell wall thickness, pit size and pit distribution, lumen size, etc.);
2. Determination of chemical composition of EW and LW (e.g. cellulose, hemicelluloses and lignin, etc.);
3. Evaluation of physical changes of EW and LW fibres in refining (e.g. fibre splitting, cell wall delamination, length reduction, fines and shives contents, etc.);
4. Measurements of sheet properties of EW and LW fibres (e.g. optical and physical properties).

Several analyses are employed to characterize the physical properties of fibres and the subsequent changes of fibre structure as a result of refining. The analyses include the following aspects:

1) Morphological modifications: fibre separation modes, changes in fibre length, width, pit structure, lumen area and cell wall thickness, cell wall splitting, fibre collapsibility, and external fibrillation, etc.

2) Handsheet characteristics: density, tensile, burst, tear, TEA, stretch, light scattering coefficient, roughness, porosity, etc. The results of these analyses will be related to those of the microscopic analyses to establish possible interrelations between the fibre characteristics and paper properties.

Chapter 6 - Methodology

To study the breakdown mechanism of EW and LW in thermomechanical pulping (TMP), the following experimental procedures have been elaborated, including methods used for characterizing the physical and chemical properties of EW and LW in different forms, such as chip, pulp and handsheets.

6.1 Experimental design

Figure 6.1 shows the main experimental procedures which are further described in details in the following sections.

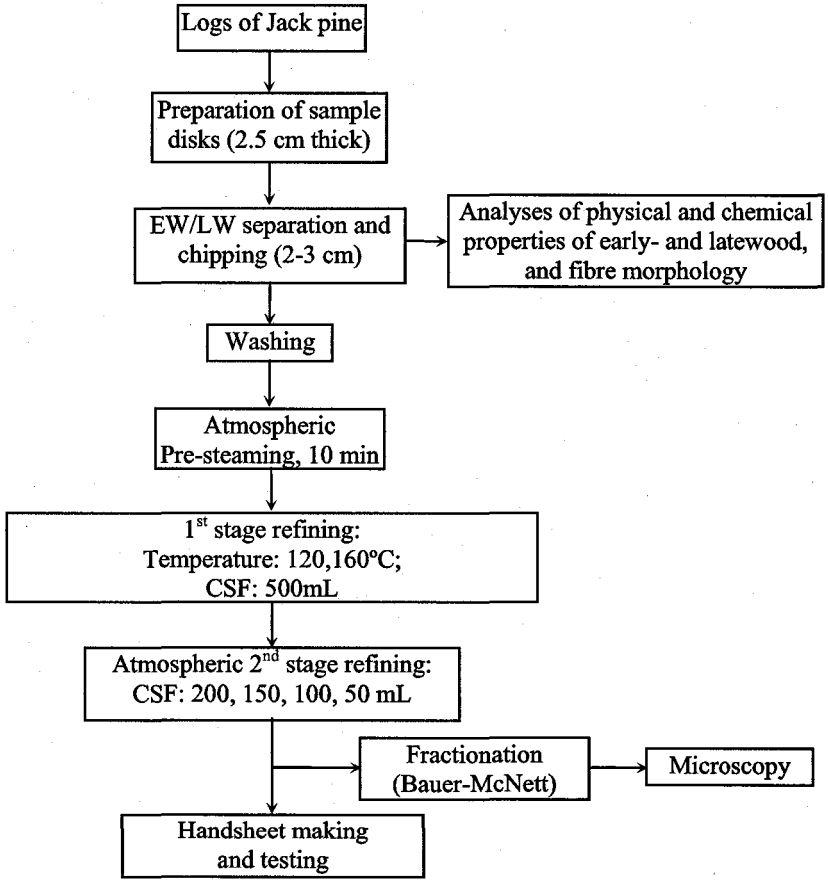

Figure 6.1 Experimental procedures

6.2 Material and preparation

Logs of freshly felled jack pine (*Pinus banksiana* Lam.) were used in this work. The jack pine trees were taken from a 30-years old plantation in St. Maurice region of Quebec. The logs were sawn into disks of about 2.5 cm thick in longitudinal direction. The disks were then debarked manually by means of a chisel. Chips were prepared from the sapwood portion, excluding the heartwood to minimize the possible effects of its high extractives content on pulp properties. The chips of EW and LW, approximately 2-3 cm in width and length, were also prepared manually using a chisel. The thickness of the chips varied depending on the width of the growth rings and the proportion of EW and LW in the growth increment. The separation of EW from LW was based on the difference in colour: LW in jack pine was relatively broad and much darker than the EW counterpart.

6.3 Physical and chemical characterization of raw material

Some optical, physical and chemical analyses were conducted for EW and LW chips, including basic density, lignin, holocelluloses, extractives and ash content.

6.3.1 Basic density

As discussed in the previous chapter, generally, LW has higher basic density in contrast to EW due to its thicker fibre cell wall. In this study, the basic density (weight o.d./volume) of the chips was measured following the Tappi method T258 om-02. In this method, the volume of the water-saturated samples is determined by water displacement technique and the weight is oven-dry weight of samples. Basic density can be expressed in the same terms as bulk density (TAPPI UM 23 "Bulk density of wood chips").

6.3.2 Chemical analysis

In this study, the chemical composition (e.g. lignin, holocelluloses, 1% NaOH solubility, dichloromethane extractives and ash) of EW and LW chips were determined following the Tappi test methods (Table 6.1)

Table 6.1 Test methods for chemical analysis

Components, %	Methods	
Lignin	Tappi T-222 om-98	Acid insoluble lignin in wood and pulp
Holocellulose	Tappi T-9 wd-75	Holocellulose in wood
1% NaOH solubility	Tappi T-212 om-98	One percent sodium hydroxide solubility of wood and pulp
Dichloromethane (DCM) extractives	Tappi T-204 cm-97	Solvent extractives of wood and pulp
Ash	Tappi T-204 cm-97	Solvent extractives of wood and pulp

6.3.3 Characterization of EW and LW fibres

To study the fibre characteristics, such as fibre length, and cell wall thickness, it is necessary first to liberate the fibres from the wood matrix by means of chemical maceration. In this process, the chips were split into match-stick sized splinters and then cooked at 70°C in test tubes containing a solution of equal parts of glacial acetic acid and hydrogen peroxide (35%), according to a modified Franklin method [114]. The test tubes were in turn placed in a water-filled beaker heated with a hot plate. The splinters were cook until they turn white. The treated specimens were thoroughly washed in running tap water. The fibres were separated by gentle shaking with some glass beads [115].

Fibre length and cell wall thickness measurement are conducted for these chemical separated EW and LW fibres. The methods are discussed in section 6.6.1 and 6.6.5 respectively.

6.4 Refining

A Sunds Defibrator 300 CD pilot plant (Metso Paper) was used for refining the chips. The refining capacity is 2 Mt/day. The models of this refiner rotor and stator plate are R3809BG and R3803, respectively. EW and LW chips were either separately refined or in blend according to the initial proportion of each exiting in the wood. In the process, the chips were pre-steamed atmospherically for 10 min and then screw-fed into a digester using a 2:1 compression ratio.

The refining was carried out in two stages, as indicated in Table 6.2. The first-stage was pressurized at 120 and 160 °C and the pulps were produced with a freeness of about 500 mL. The primary pulps were refined atmospherically to a freeness range of 50-200 mL. The specific energies were recorded for the pulps of each freeness level. The 1st stage refining consistency is about 20-24%, while the 2nd stage is around 10-14%. In addition, the refiner plate clearance is different between the two temperature levels in the 1st stage refining: 1.20-1.40 mm in lower temperature refining (120°C), but 0.40-0.60 mm in higher temperature refining (160°C).

Table 6.2 Refining conditions

	1st stage		2nd stage
Refining consistency, %	20-24		10-14
CSF, mL	500-600		50-250
Temperature, °C	120	160	Atmospheric
Refiner plate clearance, mm	1.20~1.40	0.40~0.60	0.50-0.25

The reasons of these refining condition choices are explained as follows.

6.4.1 Temperature

The common wisdom taught us that refining wood chips at elevated temperature, well above the softening temperature of lignin, results in fibre separation in the lignin-rich middle lamella [116, 117, 118, 119]. Hence, mechanical pulping processes that use temperature above the lignin softening point produces fibres with smooth surfaces coated with lignin. On the other hand, higher temperature has been exploited in combination with high refining intensity during the first stage operation, which also leads to improved pulp quality and reduced energy [120, 121, 122, 123]. In the first stage, we used a relatively low TMP temperature of 120°C and an elevated temperature of 160°C. The main purpose was to see the thin-walled EW and thick-walled LW fibre morphological changes (i.e. fibre collapsibility, rupture mode, etc.) during the two refining temperature operations and the subsequent influences on the handsheets properties.

6.4.2 Consistency

Alami et al [92] showed that, ideally, a refining system should operate within a consistency range of 45-50%. This condition will provide the maximum energy saving while producing high quality pulp with minimum dependence on consistency variations. It is reported [92, 124] that using high consistency in the fist refining stage could efficiently break down the material and separate the fibres. In this study, due to the restrictions of operation, the refining consistency is much higher in 1st stage (20-24%) to facilitate the fibre separation. In the 2nd refining stage, the consistency is relatively lower (10-14%).

6.4.3 Refiner plate gap clearance

It has to be noted that in the first stage, the refiner plate gap clearance in the high temperature (160°C, 0.40~0.60 mm) operation is reduced to more than 1/2 of that in the low temperature operation (120°C, 1.20~1.40 mm). During the high temperature operation, due to the high refining pressure, the fibres are more flexible and collapsible when compared with those in low temperature operation. In order to get the same freeness as that in low temperature operation, it is necessary to reduce the refiner plate clearance during the high temperature operation.

The reduced refiner plate gap clearance will bring some changes on the refining actions. As discovered by Law [125], there are two major forces in refining: shear and compression. Illustratively, when two refiner bars cross each other, the wood matrix or fibre mass is subjected to a major shear force and minor compression force, as indicated in Figure 6.2. These two mechanical actions occur simultaneously, as shown in Figure 6.3, the refining action of shear increase with increasing compression. Evidently, the reduced refiner plate gap increases both shear and compression forces.

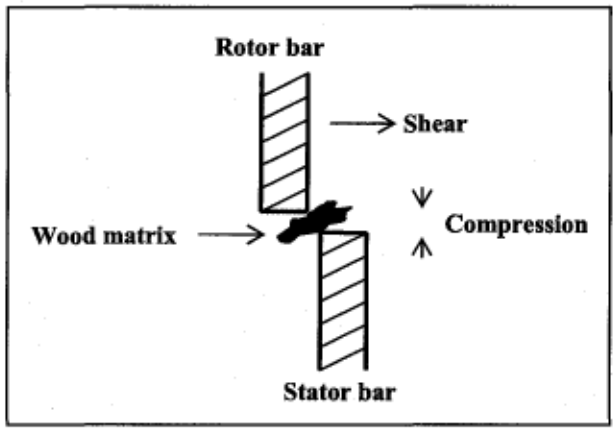

Figure 6.2 Refining actions on wood matrix

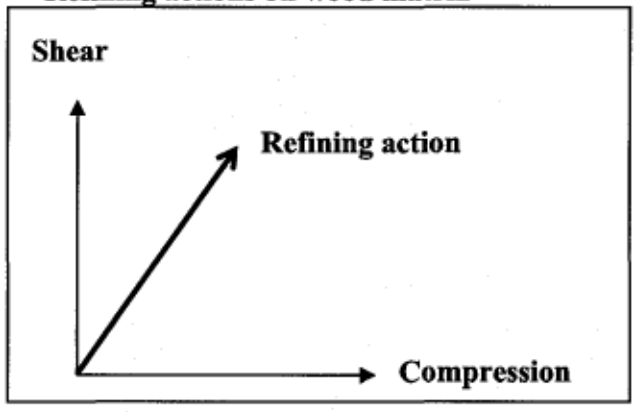

Figure 6.3 Combined action of shear and compression

6.5 Fractionation of pulps

The experimental second-stage pulps were fractionated in a Bauer-McNett classifier to obtain 6 fractions denoted as R14, R28, R48, R100, R200 and P200 (fines).

The Bauer-McNett fibre classification is a commonly used method to characterize the fibre length distribution of mechanical pulps. The fibres in different Bauer-McNett fraction are morphologically different and have different effects on the paper properties. For example, the R14 fraction contains long and stiff fibres which have poor bonding characteristics. The fines (P200) fraction comprises flake-like particles and fibrils, which would strengthen the fibre network. Based on these reasons, it is necessary to fractionate the pulps before the characterization.

6.6 Pulp characterization

Each fibre fraction was characterized in terms of cell wall rupture modes (e.g. inter-cell, trans-wall, and intra-wall failures), cell wall peeling, fibre breakage, fibre wall splitting, external fibrillation and collapsibility. These analyses were conducted by means of microscopic analysis, including optical and electronic microscopic analysis.

Meanwhile, specific volume (SV) and water retention value (WRV) of pulp fibres were determined as described in sections 6.6.3 and 6.6.4 respectively.

The fibre length and coarseness, curl index, kink index were measured by means of a FQA (Optest Equipment).

Shives content was determined using a Pulmac shive analyzer (Pulmac II, MasterScreen) while the fines content was obtained from the Bauer-McNett classifier.

6.6.1 FQA analysis

The Fibre Quality Analyzer (FQA, OpTest Equipment Inc.) employed in this study is an optical device used to measure the fibre length, fibre coarseness, fibre curl and kink index, etc. It uses circular polarized light to project images of the fibres in the sample to gain information on fibre length and contour. As the fibre joins the water sheaths and passes through the optics box, a digital camera takes pictures of the fibre projections, as showed in Figure 6.4.

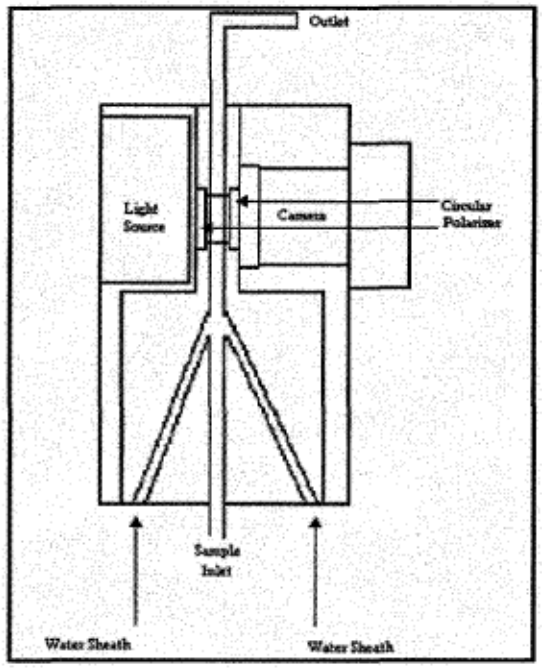

Figure 6.4 Flow cell configuration of FQA [126]

6.6.1.1 Fibre length

As discussed previously, in the fibre development stage of refining, the fibres are broken into short fragments and fines, which lead the fibre length reduction. The fibre length measurement will directly reflect this mechanical response of fibre in refining.

In FQA length measurement equation, fibres are grouped into various length classes ℓ, n_i is the number of fibres in class ℓ_i. According to different statistic methods, three types of mean length data can be obtained from FQA: arithmetic, length-weighted, weight-weighted mean, showed as follows [127].

$$\ell_n = \sum_i n_i \ell_i / \sum_i n_i \qquad \text{Eq. 6.1}$$

$$\ell_{lw} = \sum_i n_i \ell_i^2 / \sum n_i \ell_i \qquad \text{Eq. 6.2}$$

$$\ell_{ww} = \sum_i n_i \ell_i^3 / \sum_i n_i \ell_i^2 \qquad \text{Eq. 6.3}$$

where,

ℓ_n = arithmetic mean length

$\ell_{\ell w}$ = length-weighted mean length

ℓ_{ww} = weight-weighted mean length

The arithmetic mean length (ℓ_n) is applicable for the fibre samples with fairly uniform length. If the sample contains a high proportion of short fibres and fines, such as TMP fibres, the weighted mean length is then preferred. There are two forms of weighted mean length: the length-weighted mean length ($\ell_{\ell w}$) and weight-weighted mean length (ℓ_{ww}). The length-weighted mean length ($\ell_{\ell w}$), is defined for the case where the fibre weight per unit length (fibre coarseness) is assumed to be constant. The weighted-weight mean length (ℓ_{ww}) is defined for the case where the fibre coarseness is proportional to the fibre length. Compared with the weight-weighted mean length, the length-weighted mean length is considered to give a better prediction of the papermaking potential of the fibres [128]. Accordingly, in this study, the length-weighted mean length was employed.

6.6.1.2 Fibre coarseness

Fibre coarseness is a measure of cell wall mass per fibre length, as showed in Eq. 6.4. During the fibre development stage of refining, fibre coarseness decreases due to the peeling of cell wall. Generally, in refining, thick-walled LW fibres result in coarser fibres than those of EW. The measurement of fibre coarseness will help us understand the breakdown behaviours of EW and LW in refining, such as external fibrillation and cell wall reduction.

$$C = \frac{M}{N \times \ell_n} \qquad \text{Eq. 6.4}$$

where,

C = coarseness (mg/m)

M = dry weight of the fibres

N = number of the fibres

ℓ_n = arithmetic mean length of the fibres

6.6.1.3 Curl index

Curl is the gradual and continuous curvature and is defined by the curl index, as shown in Figure 6.5 and Eq. 6.5 [129]. Fibres in wood are straight. They become curly during pulping, mixing and refining, when exposed to bending and axial compressive stresses, particularly at high consistencies.

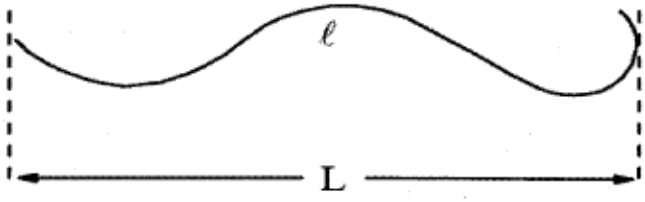

Figure 6.5 Definition of fibre curl index

$$Curl\ \ index = \frac{\ell}{L} - 1 \qquad \text{Eq. 6.5}$$

The mechanisms of curl induction, removal and retention in pulp fibres have been discussed by Page et al. [129]: in mechanical pulps, the curl manifests as stress or deformation in the lignocellulosic matrix of the fibre wall. Fibres straighten as the stress is relaxed. This occurs during latency removal, when fibres are stirred at low consistencies and at elevated temperatures. Fibre curl index is related to its morphological characteristics: the thick-walled LW fibres are expected to be less curlable than those of thin-walled EW fibres. Measurements of curl index can indicate the fibre flexibility or reciprocal stiffness.

6.6.1.4 Kink index

Kink or the abrupt change in fibre curvature, is another parameter used for usual measurements of fibre shape (Figure 6.6). Kink index or Kibblewhite's Kink index is the weighted sum of the number, Nx, of kinks within a range of "x" kink angles, as indicated in Eq. 6.6.

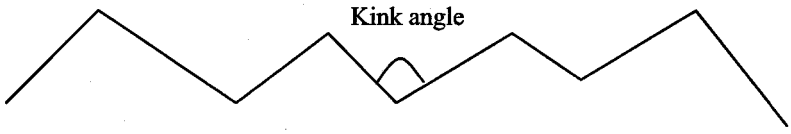

Figure 6.6 Illustration of kink in fibre

$$Kink\ \ index = \frac{\{N_{(21-45°)} + 3N_{(46-90°)} + 4N_{(91-180°)}\}}{Total\ \ fibre\ \ length} \qquad \text{Eq. 6.6}$$

Kink index is an important parameter which is also related to fibre morphologic characteristics. Previous research work revealed that in refining, the thin-walled fibres are less resistant to the mechanical action and have much greater kink index than thick-walled fibres [130]. Measurements of the kink index could yield a useful indication of fibres' behaviour during refining.

In sum, FQA is an efficient and multi-purpose equipment to quantitatively evaluate EW and LW fibre morphological changes in refining. Finally, it has to be noted that: since the fibres of R14 fraction tend to jam the flow tube of pulp suspension of the instrument, these fibres were not characterized. Regarding the R200 and P200 fractions, they tend to yield unreliable results for their tiny size [131] and therefore, they were also excluded from the measurements. For these reasons we characterize only the R28, R48 and R100 fractions.

6.6.2 Shives analysis

The shives contents were determined by means of a Pulmac shive analyzer equipped with a 0.1 mm screen plate. The technique is given in the TAPPI test method T274 sp-97: 2g pulp slurry (0.1-0.15 g/L) is needed for a 10 min screening cycle. The retained shives are collected, dried and weighted for determining the percentage of shives content. Some shives were also collected for microscopic analysis, such as fibre types and rupture modes. The expected results will help us to understand the breakdown behaviour of EW and LW in refining.

6.6.3 Specific volume (SV) analysis of fines by sedimentation

The quality of the fines is an important characteristic and it can be qualified by the SV of these particles. The mechanical pulp fines consist of three types of particles: fibril and flake-like elements, and ray cells. The fibrils consist of cellulose-rich material with a high specific surface area (volume), while the flakes are lignin-rich cell outer layer of cell (e.g. the compound middle lamella) with low surface area [132, 133]. EW and LW are morphologically different, thus under refining actions, they could produce different sizes of particles in the fines and yield different SV values. The SV analysis of fines will provide information of EW and LW breakdown in refining, especially in the morphological changes in the fines fraction. As described by Marton *et al.* [133] and Luukko [134], the SV analysis of fines could be divided into two steps: fines collection and SV measurement, as showed in Figure 6.7.

6.6.3.1 Fines collection

The fines were collected by passing the pulp suspension (0.2-0.3 %) through a 200-mesh wire in a 2-litre Dynamic Draining Jar (DDJ). The pulp suspension was prepared as following: 10 g pulp was disintegrated at 1.2% consistency in hot (90°C) demineralised water for 5 min (about 3530 rev.) in a British disintegrator to remove latency. The pulp was then diluted to 0.2-0.3% consistency with demineralised water. Due to the volume limit of DDJ, the 10 g diluted pulp was divided into several batches for the fines collection; each batch consisted of 1500 mL of pulp suspension. During the separation, the pulps are firstly agitated (1000 rpm) in DDJ for 2 min with the bottom valve closed. After that, the bottom valve was opened to let pass the fines which was collected from the filtrate. The filtrate was then placed in a graduated cylinder and kept still for 24 hours, after which the supernatant was decanted by siphoning. The concentrated fines suspension was then centrifuged (6000 rpm) for 5 min to increase its consistency to 0.4-0.5 %. The condensed fines suspension was for SV measurement.

6.6.3.2 SV measurement

Since the fines suspension is colloidal, some effect of possible flocculation could take place in the suspension, influencing the settling. To prevent the occurrence of flocculation, the metal ions and pH of the suspensions should be strictly controlled [133]. In this work, 50 mL of fines suspension was firstly mixed with a solution containing Na_2SO_4 120 mg/L and $CaCl_2$ 30 mg/L (mixing volume ratio: 1:2) to equilibrate the ions content and then pH was adjusted to 6-6.5. A 1 g/L NaOH and 1 g/L HCl were employed for the pH adjustment. After that, the treated fibre (or fines) suspension was transferred to a 100 mL graduated glass cylinder, and the surplus was discharged. Before the settling test, the air in the suspension was removed by a vacuum pump for 10 min. After the air removal stage, the cylinder was sealed by a parafilm and manually well shaken to disperse the suspension. After that, this suspension was kept still for 24 hours, after which the volume of the settled fines was read (accuracy ±1 mL). And then, this suspension was filtered to recovery the fines. The mass of fines was obtained by drying at 105 °C; the weight of the recovered fines is needed to determine the SV value. The SV was calculated as shown in Eq. 6.7. Three replicates were taken for determining the average value of SV.

$$SV(cm^3/g) = \frac{V}{W} \qquad \text{Eq. 6.7}$$

where

SV = specific volume
V = volume of fine suspension
W = oven dry weight of recovered fines

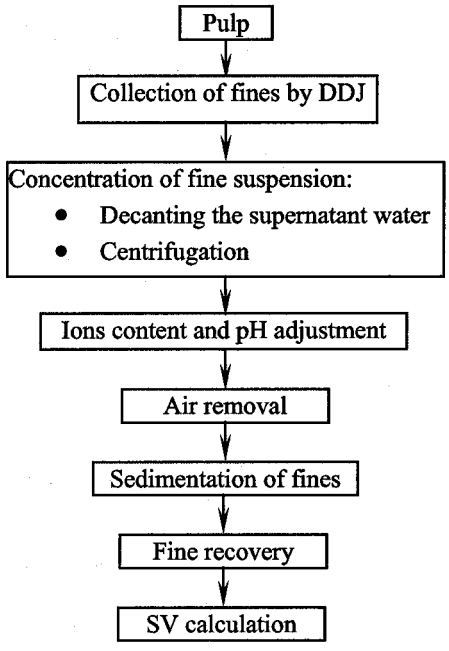

Figure 6.7 Procedures for the measurement of specific volume of fines

6.6.4 Water retention value (WRV) determination

Water retention value (WRV) of pulp is related to fibre internal fibrillation. Under mechanical refining action, the fibre walls split and delaminate, which facilitates the water absorption. As a result, the fibres swell and yield higher water retention value. The WRV measurement could reflect the fibre cell wall damage in refining, such as split and internal fibrillation (delamination).

In this study, WRV analysis was conducted on four Bauer-McNett fractions (R14, R28, R48 and R100). The method is described by Allender and Waterhouse [135]: briefly, a fibre mat was formed (using a pulp slurry of 52 mL at 0.25% consistency) onto the 100-mesh wire in the centrifuge tube and centrifuged (IEC UV Centrifuge, Diameter 60 cm) at 2500 rpm for 15 minutes. The centrifuged samples were weighted before and after drying overnight at 105 °C. The WRV is expressed as gram of water per gram of oven-dry pulp, as indicated in Eq.6.8.

$$Water\ \ retention\ \ value = \frac{W1 - W2}{W2} \qquad \text{Eq. 6.8}$$

where,

$W1$ = sample weight before drying

$W2$ = sample weight after drying

6.6.5 Cell wall thickness measurement

Cell wall thickness is an important morphological characteristic which is related to the fibre's stiffness or its ability of inter-fibre bonding. The measurement of cell wall thickness also provides us with the information on the mechanical response of fibres in refining. In this study, the cell wall thickness was measured by means of a MorFi cell wall thickness analyzer (Techpap, France) [136]. This instrument, an independent table unit, has two major parts: hydraulic part and electrical part. The hydraulic component includes a pulp sampling system, a pump, circuits and valves, a measuring cell and a camera equipped with a microscope having a resolution of 0.5 μm. The electric component integrates mainly the electrical supplies, relays and a PC (Figure 6.8).

In the process of the measurement, a sample of 0.01 g/L was poured into a sample beaker of 500 mL and the fibres are dispersed mechanically by an agitator. The dispersed fibre suspension was then pumped through a measuring cell where a camera grabs the fibre's microscopic image. Image analysis software calculates the average cell wall thickness and produces a thickness distribution chart.

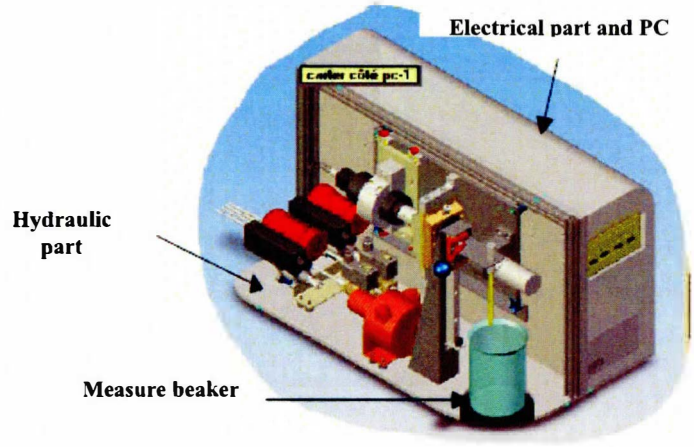

Figure 6.8 Profile of MorFi cell wall thickness device [137]

6.6.6 Light microscopy and image analysis

Light microscopy and image analysis were used to quantitatively evaluate the characteristics of rupture of EW and LW fibres in refining, such as, external fibrillation, trans-wall and intra-wall failures in fibre cross-sectional dimension [138]. For the statistics reason, at least 300 fibres per sample were measured.

1. External fibrillation

External fibrillation is calculated by the comparison of fibre outer perimeter between the fibres with and without fibrils (Figure 6.9), calculated as shown in Eq. 6.9. The perimeter measurement was conducted by means of a Plan Wheel (Scalex Corporation139): roll the wheel along the contour of fibre in the microscope image, and read the measurement on the plan wheel display.

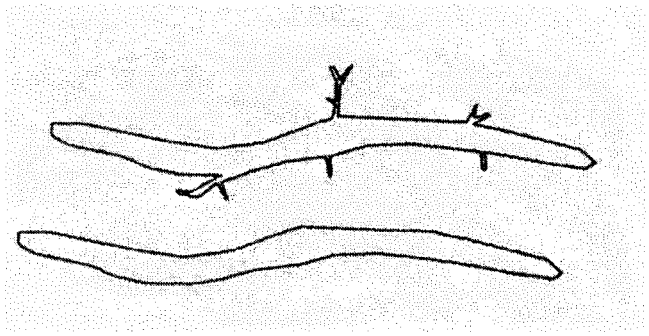

Figure 6.9 External fibrillation of fibre

$$Degree\ of\ External\ Fibrillation = (\frac{P_F}{P} - 1) \times 100 \qquad \text{Eq. 6.9}$$

2. Trans-wall and intra-wall failure

In the cross-section image of TMP fibres, we can see two types of physical changes taken place in the fibre wall: trans-wall and intra-wall failure, these characteristics are shown in Figure 6.10 [140].

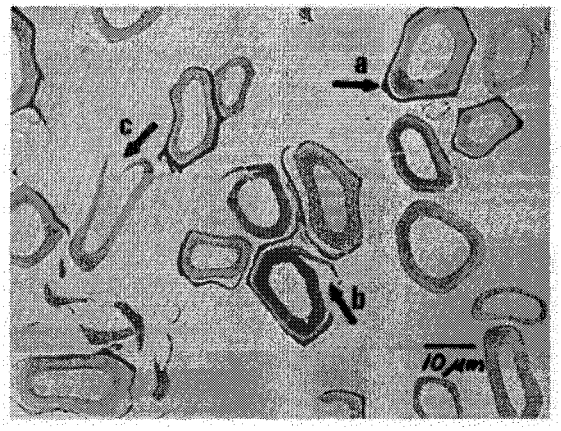

Figure 6.10 Cell wall damage in refining [140]
((a) CML retention (b) intra-wall failure (c) trans-wall failure

In this study, the proportion of damaged fibres was assessed by counting the fibres which present the feature of interest, i.e., trans-wall and intra-wall failure. These fibre cross-section characteristics were carried out by image analysis. The sample preparation procedure was described as following: first, the fibre were aligned according to the method described in [141]; second, the aligned fibres were progressively dehydrated using water-ethanol mixtures with increasing concentration of ethanol (10, 30, 60, 80, 90

and 100%); then the dehydrated fibre were impregnated with a resin medium as described in [142]; third, the resin-impregnated fibres were imbedded in plastic capsules filled with resin medium and were cured at 70°C for at least 8h or longer. The cured samples were sectioned in 3 μm-thick sections using a sledge microtome. The thin sections were stained with Toluidine Blue O (T161, Fisher Scientific Co.), and then mounted on microscope slides for observation using a Zeiss photomicroscope. In brief, the sample preparation can be described in Figure 6.11.

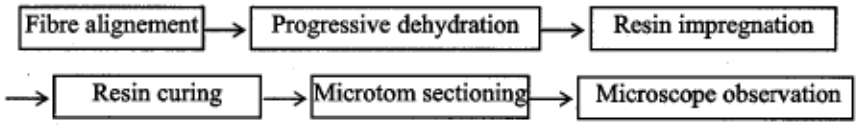

Figure 6.11 Sample preparation for image analysis

6.6.7 Scanning electron microscopy (SEM) and image analysis

Scanning electron microscopy (SEM) was employed to qualitatively examine the breakdown of EW and LW fibres, e.g., fibre separation mode (S1, S2, S1/S2 exposure), splitting, collapsibility etc. [138, 143]. An ImageJ algorithm (Image Processing and Analysis in Java) [144] was employed to directly measure the following fibre properties in SEM images: cell wall thickness, cell wall area, area of filled fibres (fibre wall area + lumen area), outer and inner fibre perimeter of cell wall [145]. The fibre collapsibility was determined as follows.

Form Circle (*FC*) and Form Shape (*FS*) were used to evaluate the fibre collapse in refining [1, 52, 146, 147], the collapsibility of EW and LW fibres could be compared by means of these parameters (Figure 6.12).

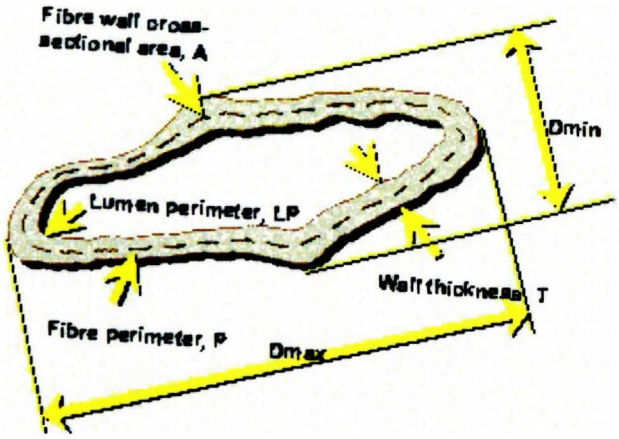

Figure 6.12 Fibre cross-section analysis

$$FC = \frac{4\pi A_t}{P^2} \qquad \text{Eq. 6.10}$$

$$FS = \frac{D_{\min}}{D_{\max}} \qquad \text{Eq. 6.11}$$

where

A_t = area of filled fibres (fibre wall area + lumen area);

P = outer perimeter of cell wall without fibrils;

D_{min} = minimal distance measured between parallel tangents to the cross-section;

D_{max} = maximal distance measured between parallel tangents to the cross-section;

Both FC and FS are useful parameters to evaluate the fibre collapsibility. The cell wall area, lumen area and the outer perimeter of fibre are used in defining the FC equation (Eq.6.10). On the other hand, only the minimum and maximum distances measured between parallel tangents to the cross section are employed to define the FS (Figure 6.12 and Eq. 6.11). Due to the differences in the cell wall thickness and lumen size between EW and LW fibres, their cell wall areas and outer perimeters are quite different. Therefore, when compared with the FS, the FC reflects more precisely the cross-section deformation. However, in this study, both of FS and FC are measured for the cross-section analysis.

The initial stages of sample preparation for SEM analysis were similar to those used for conventional thin sectioning (as showed in Figure 6.11). Since SEM images are based mainly on the topography of the surfaces, a smooth sample block face would show a faint image of poor clarity of the embedded specimen. To resolve this problem, a modified method was adopted to remove a part of resin in the sample surface. In this method, a thin layer of the supporting resin was dissolved away from the block face to expose the specimen structure. The solvent used in this work was a saturated solution of NaOH in a mixture of absolute ethyl alcohol and propylene oxide [148, 149]. The treated samples were observed using a SEM (JOEL, JSM-500). The ImageJ was used to calculate the *FC* and *FS* from the SEM images. The sample preparation procedures are shown in Figure 6.13.

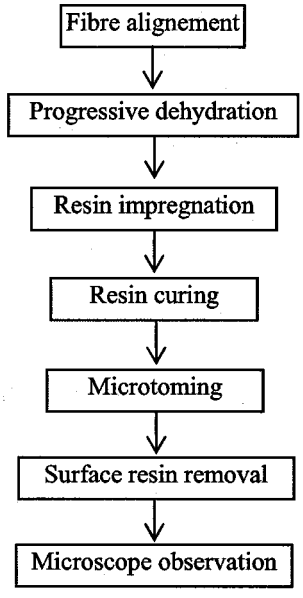

Figure 6.13 Sample preparation for SEM image analysis

6.7 Handsheet formation

Generally, the relatively long fibre fraction (e.g. R14, R28, and R48) contain long and stiff fibres with poor bonding characteristics while the fine fraction (P200) is comprised mostly of fibrils from S layer and flakes from compound middle lamella. The fines (P200) favour the bonding in fibre networks [34]. In order to examine the influences of

fibre characteristics on paper properties, standard handsheets of 60 g/m^2 (Tappi method T205 sp-02) were made from different Bauer-McNett fractions of EW, LW and the mixture of EW and LW. The handsheets included:

- Whole pulp;
- Pulp without fines (P200);
- Combined long fibre fractions (R14+R28+R48);
- Combined short fibre fraction (R100+R200+P200);
- Individual fractions of R14, R28, R48 and R100;
- Blends of EW and LW pulps after refining. This pulp mixture has the same weight proportion of EW and LW as in the chip mixture. The results were compared with those of mixture of EW and LW refined together.

6.7.1 Characterization of handsheets

Some physical, chemical, and optical properties and microscopic structures were measured for these handsheets of the second-stage refining pulp, hoping to establish some possible interrelations between the fibre characteristics and paper properties.

1. Physical properties: tensile, tear, burst, stretch, TEA, density, porosity, roughness, inter-fibre bonding (Scott bond);
2. Chemical properties: Oxygen/carbon atomic ratio of fibre surface (X-ray photoelectron spectroscopy) [150, 151];
3. Optical properties: brightness, light scattering coefficient, light absorption coefficient, opacity;
4. Structural properties, such as the twist angle of EW and LW fibres in handsheets (SEM);

5. Basic characteristics such as cell wall thickness, lumen size, collapsibility, form factor and cell splitting were also examined by SEM, as discussed in section 6.6.

6.7.1.1 X-ray photoelectron spectroscopy (XPS) analysis

During refining, the EW and LW fibre cell walls were peeled off in different ways (e.g. inter-cell, trans-wall or intra-wall), which would influence their surface chemical composition, especially the lignin. X-ray photoelectron spectroscopy (or Electron Spectroscopy for Chemical Analysis, ESCA), is a well-known method to measure the chemical surfaces [150, 151]

Dorris and Gray [152, 153, 154] were pioneers in the field of characterizing mechanical pulp surfaces with ESCA. The ESCA measurements were made with an AXIS ULTRA electron spectrometer equipped with an Al (Mono) X-ray source. The analysed area had a diameter of 2x2 mm, and the angle from the X-ray detector to the sample was 45°. Peak intensities were determined by peak area integration. The sensitivity factors used were 0.278 for C1s and 0.780 for O1s. The matching of the curve fitting to the C1s peaks was carried out with Gaussian curve fitting program. Sheets with a layer grammage of 60 g/m^2 were made for the ESCA analysis. In the ESCA technique the chemical composition was normally evaluated either using the C1s and O1s peaks by calculating the total O/C ratio. The surface coverage of lignin, ϕ_{lignin}, was calculated from the O/C atomic ratio, Eq.6.12 [155]

$$\phi_{lignin} = 100 \times [\frac{O/C_{(pulp\ sample)} - O/C_{(carbonhydrate)}}{O/C_{(lignin)} - O/C_{(carbonhydrate)}}] \qquad \text{Eq. 6.12}$$

where

$O/C_{(carbonhydrate)}$= 0.833

$O/C_{(lignin)}$= 0.333

6.7.1.2 Twist angle determination

The twist angle was the angle measured between the longest dimension of the fibre cross section and the horizontal frame of the image [156]. As showed in Figure 6.14, β is the

twist angle, which indicates the rotation extent of fibre cross-sectional major axis towards to the paper plane. The orientation of the fibre with respect to the paper plane (fibre twisting) could strongly affect the structure of the paper. Decreasing the number of twist in fibres will increase sheet strength [157]. The reduction in fibre twist would reduce the amount of space taken by the fibre in the paper structure, thereby reducing the void space and increasing the density of paper. The reduction in fibre twist could also increase the potential bonding surface area of the fibres, especially for collapsed fibres [156].

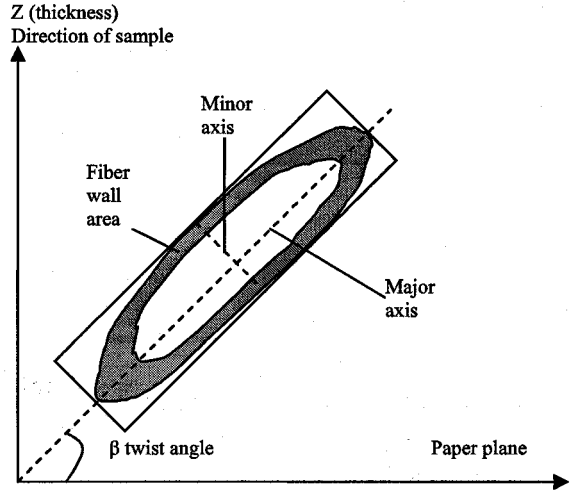

Figure 6.14 Definition of fibre twist angle [156]

Twist angle determination was conducted by SEM image analysis, as discussed in section 6.6.6. The sample preparation was the same as that described in Figure 6.13: paper samples were embedded in resin, and then a thin layer of resin were dissolved by an organic solution to expose the observing surface. An angle tool was applied to measure the twist angle in the SEM images.

6.7.1.3 Optical and physical properties analysis

Some paper properties of the sheet made from the whole pulp and each Bauer-McNett fraction were be measured according to the methods listed in Table 6.3.

Table 6.3 Test methods of paper properties.

Properties	Methods	
Thickness	Tappi T411 cm-97	Thickness (calliper) of paper, paperboard, and combined board
Physical properties (density, tensile, burst, tear indices)	Tappi T220 sp-01	Physical testing of pulp handsheets
Tensile energy absorption (TEA)	Tappi T494 om-01	Tensile breaking properties of paper and paperboard
Interfibre bond strength (Scott bond)	Tappi T569 pm-00	Test for interfiber bond using the internal bond tester
Brightness	Tappi T452 om-98	Brightness of pulp, paper, and paperboard
Light scattering	Tappi T220 sp-01	Physical testing of pulp handsheets
Opacity	Tappi T425 om-96	Opacity of paper
Roughness	Tappi T538 om-01	Roughness of paper and paperboard
Porosity	Tappi T547 om-97	Air permeance of paper and paperboard

In summary, the principal aspects analyzed are shown in Figure 6.15.

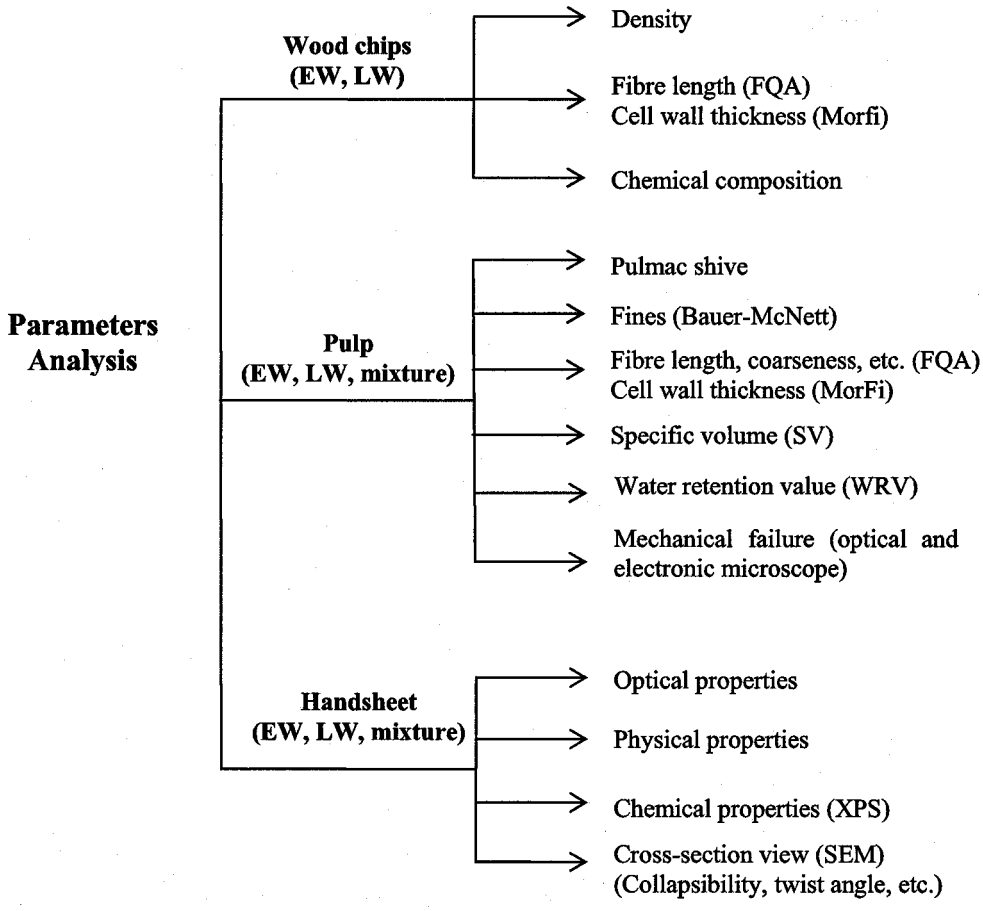

Figure 6.15 General outline of analysis parameters

Chapter 7 - Results and discussion (1) — raw material

In this chapter, the physical and chemical characteristics of EW and LW Jack pine (*Pinus banksiana* Lamb.) used in the present study are discussed. The physical properties include bulk density of chips, fibre length, cross-section dimension and cell wall thickness, while the chemical components include lignin, holocellulose, extractives, 1% NaOH dissolution and ash contents.

Wood characteristics play an important role in determining the pulp qualities [158]. Jack pine is a species which has thick-walled fibres when compared with the traditional pulp wood species such as black spruce (*Picea mariana*) and usually gives low tensile and burst strength [159, 160, 161, 162, 163]. For its morphological difference and its comparatively high pitch content, the use of Jack pine in mechanical pulping is relatively limited [159]. In this study, we compared the physical and chemical characteristics of Jack pine EW and LW with the whole wood of Jack pine and black spruce. The objective of this investigation was to study the contribution of EW and LW to the papermaking problems, in terms of low physical strength and high pitch content.

7.1 Physical characteristics

7.1.1 Cross-section view of Jack pine

As discussed in the previous experimental section, the EW and LW were separated manually using a chisel. The separation of EW from LW was based on the difference in colour. Shown in Figure 7.1 is a cross-sectional view of a wood block of Jack pine. LW zone was much darker than the EW counterpart. As seen in the figure, the zone of LW in Jack pine was rather broad, representing approximately 30 to 40% in volume. Due to its thick-walled fibres (Figure 7.1 and Table 7.1), the mass proportion of LW could be well above 60% in a given growth increment. Because of this high proportion of LW, Jack pine is considered as a high density species (Table 7.1). For this same reason, Jack pine always poses problem in terms of paper properties and refining energy in mechanical pulping [130].

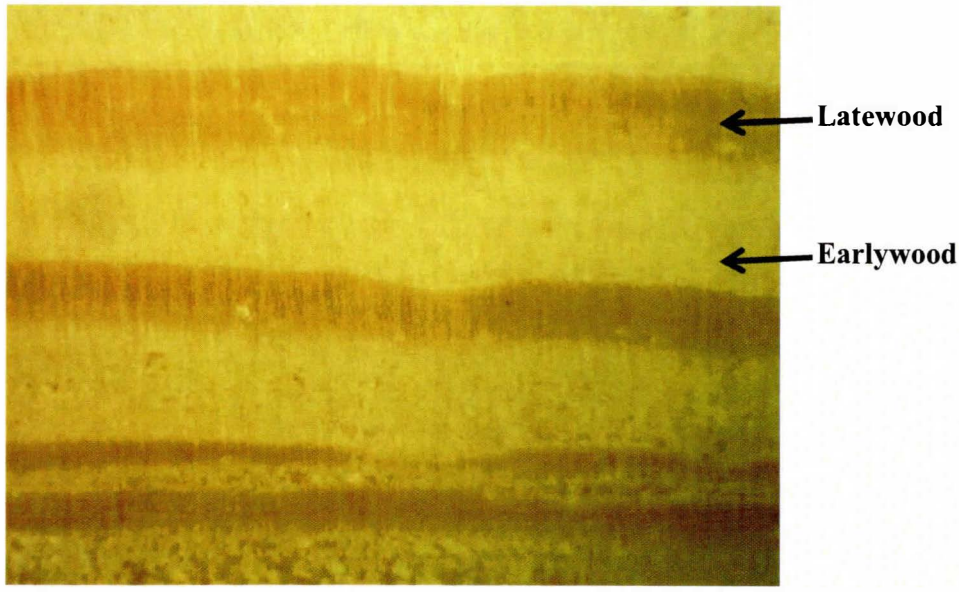

Figure 7.1 **Incident light micrograph showing the cross-sectional view of Jack pine (x10)**

7.1.2 Physical characteristics

Table 7.1 indicates that the bulk density of LW is nearly 1.6 times as high as that in EW. When compared with thin-walled EW fibre, thick-walled LW fibre has a greater cell wall volume and smaller lumen area. As a result, LW density is greater than that of EW. In addition, the LW fibre (3.55 mm) is longer than EW fibre (3.34 mm).

Figure 7.2 shows the cross-sectional view of a growth ring of Jack pine. The photomicrograph reveals that the EW fibre has thinner cell wall and larger lumen in comparison with LW counterpart. These differences were also quantitatively compared in Table 7.1. The cell wall thickness of LW (4.75 μm) is more than 2 times as that in EW (2.12μm). EW fibre has greater outer perimeter and lumen area than LW. LW fibre has larger cell wall area due to their thicker cell wall and smaller lumen. Again thanks to its relatively thicker cell wall, the LW fibre's coarseness (19.5 mg/100m) is much higher than that of EW (16.8 mg/100m). These findings are well in line with those reported earlier [54, 55]. However, it has to be pointed out that the coarseness values presented in Table 7.1 were obtained using macerated fibres (approximately 50% yield) and, hence, the real coarseness values of fibres in wood or those of mechanical fibres should be much higher.

Table 7.1 Physical characteristics of Jack pine and black spruce

	Experiment data	Reference data	
	Jack pine	Jack pine	Black spruce
Basic density, g/cm^3	0.458	0.46 [164]	0.45 [164]
EW	0.298	-	-
LW	0.485	-	-
LW percentage, % in mass	60	-	50 [74]
Length, mm	3.48	3.5 [164]	3.5 [164]
EW	3.34	-	-
LW	3.55	-	-
Cell wall coarseness, mg/100m	18.5	18.0 [165]	15.8 [165]
EW	16.8	-	-
LW	19.5	-	-
Cell wall thickness, µm	3.98	-	-
EW	2.12	2.75 [165]	2.30 [165]
LW	4.75	4.85 [165]	3.45 [165]
Outer perimeter, µm	-	-	-
EW	130	-	-
LW	105	-	-
Lumen area, µm^2	-	-	-
EW	400	-	-
LW	260	-	-
Cell wall area, µm^2	-	-	-
EW	240	-	-
LW	350	-	-
Flexibility ratio	-		
EW	-	0.77 [165]	0.82 [165]
LW	-	0.39 [165]	0.51 [165]
Rigidity coefficient			
EW	-	1.2 [165]	1.1 [165]
LW	-	3.2 [165]	2.9 [165]

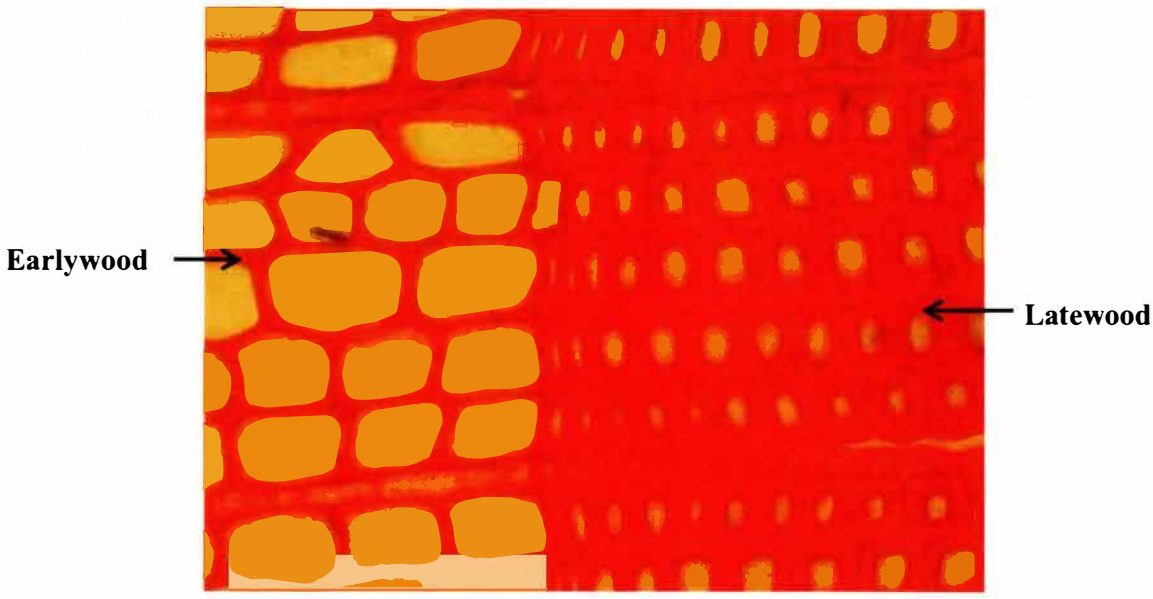

Figure 7.2 Photomicrograph showing the cross-section morphology of EW and LW of Jack pine (x200)

In the comparison of reference and experimental data, Jack pine has a slightly higher basic density than black spruce because it has higher percentage (60%) of thicker LW fibres than that of spruce (50 %). In addition, the cell wall thickness of Jack pine LW fibre (4.85 μm) is much greater than that of spruce (3.45 μm), which is mainly responsible for its higher fibre coarseness. High percentage of coarser LW fibre contributes to the higher rigidity and less flexibility of Jack pine wood fibre as compared with those of black spruce, and could account for higher energy requirement in refiner pulping of Jack pine [160]. High percentage of coarser LW fibre also renders the Jack pine TMP pulps inferior in strength property when compared with black spruce [166]. Research indicated that biological, chemical or mechanical pre-treatment of Jack pine could improve its refining performances and pulp qualities [159].

7.2 Chemical components

The experimental data of chemical components of EW and LW of Jack pine are given in Table 7.2. Due to its thicker cell wall, LW has about 3.5% more holocellulose content when compared with EW. However, it has lower lignin content. Note that LW also has higher 1% NaOH solubility. On the other hand, EW has 4.5% higher lignin content in

comparison with the LW. This is probably attributed to its relatively thicker lignin-rich CML as explained by Fengel [65]. Interestingly, the EW shows 20.4% higher dichloromethane (DCM) extractives than LW does. This difference in DCM extractives might probably due to the fact that the EW contain more resin-rich canals than LW, as shown in Figure 7.3. In addition, there are no significant differences in the ash content between EW and LW.

Table 7.2 Chemical components of Jack pine and black spruce

	Experimental data	**Reference data**	
	Jack pine	**Jack pine**	**Black spruce**
Klason lignin, %	27.50	27.6 [167]	26.9 [167]
EW	28.30	-	-
LW	27.09	-	-
Holocellulose, %	69.77	69.7 [167]	73.3 [167]
EW	68.68	-	-
LW	71.01	-	-
Dichloromethane (DCM) extractives, %	1.88	2.0 [168]	1.0 [168]
EW	1.95		
LW	1.62		
1% NaOH solubility, %	12.60	12.7 [167]	13-15 [167]
EW	11.64		
LW	13.13		
Ash, %	0.17	0.16 [167]	0.22 [167]
EW	0.17		
LW	0.17		

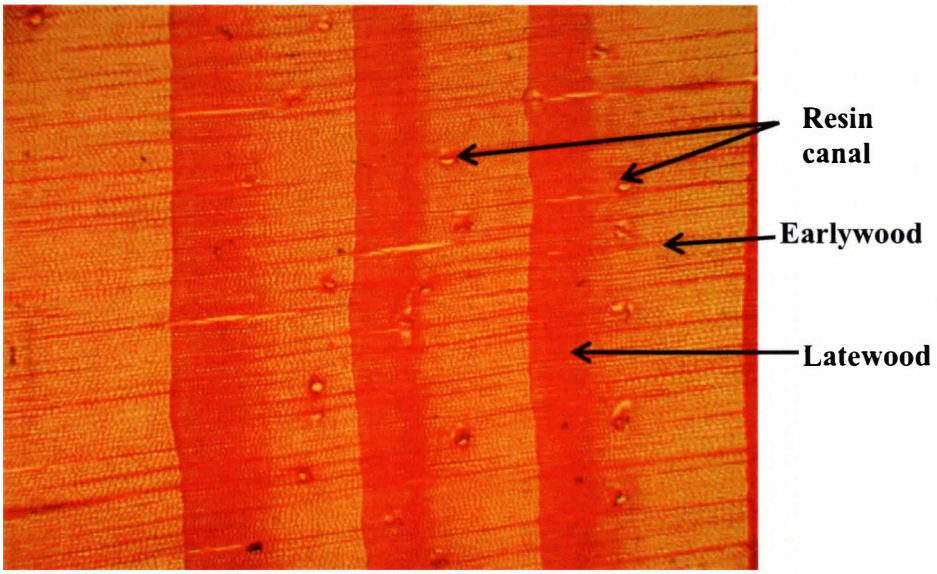

Figure 7.3 Resin canals in Jack pine (x40)

As shown in Table 7.2, the experimental data correlated well with the data reported by other workers with the exception of dichloromethane (DCM) extractives. The DCM extractives value obtained in this study is slightly lower than the reference data. This might be due to the fact that we had eliminated the extractive-rich heartwood during the chip preparation. Note that the DCM extractives content in Jack pine is about twice as high as that in black spruce, indicating that Jack pine has more serious pitch problem in pulping [169] and difficulties in bleaching [170, 171]. As indicated in Table 7.2, in comparison to LW, EW is mainly responsible for this problem since it contains more extractives than the former. Recent report indicated that the pitch problem could be minimized by biological, mechanical, physical or chemical treatment before pulping [159].

7.3 Summary

Jack pine EW chips are lighter in colour than those of LW. LW has higher basic density than EW and has relatively longer fibre length when compared to EW. In addition, the cell wall thickness of LW fibre is twice as that of EW. The high proportion of LW is responsible for its inferior paper strength in comparison with black spruce.

Regarding the chemical compositions, LW has higher content of holocellulose while EW shows greater lignin content. Furthermore, EW has higher dichloromethane (DCM) extractives than LW because the former has more resin-rich canals.

Chapter 8 - Results and discussion (2) — pulp characterization

In this chapter, we describe the morphological changes of EW and LW fibres incurred in refining. The experimental pulps were classified into various fractions using a Bauer-McNett classifier. The fibres of each fraction were characterized in terms of fibre length, cell wall thickness, fibre coarseness, curl and kink index, water retention values (WRV), and specific volume (SV) of fines. In addition, the surface lignin coverage of EW and LW pulp fibres was analyzed by means of an X-ray photoelectron spectroscope (XPS). Optical and electron microscopic analyses were also conducted to examine the rupture characteristics of EW and LW fibres.

8.1 Refining energy

Figure 8.1 clearly shows that refining EW required more energy than refining LW at a given freeness. As discussed later, the EW was defibrated into pulp fibres with relatively little fibrillation when compared to the LW. As a result, EW pulp had higher freeness for a given energy consumption. This finding is in agreement with that reported by other researchers [10]. It was also observed that the energy consumption of the mixed furnish containing both EW and LW was found between those of EW and LW.

Note that the refining temperature had also important influence on refining energy consumption. In comparison with low temperature refining at 120 °C the high refining temperature (160°C) decreased the friction between fibre mass and refiner plate surface due to the fact that the wood matrix is softer at 160°C than at 120°C. For this reason it is necessary to reduce the plate gap to improve refining efficiency. As a consequence, the reduced plate clearance had unfortunately increased fibre cutting, producing more fines and decreasing more rapidly the freeness of the pulp produced.

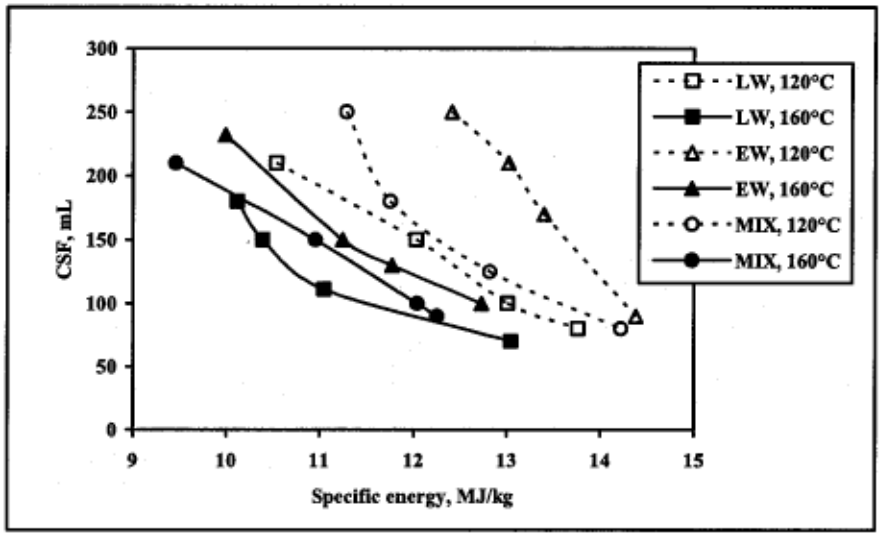

Figure 8.1 Freeness as a function of specific refining energy

8.2 Fibre fraction

The distribution of length fractions constitutes an important nature of mechanical pulps. It reflects the resistance of wood fibres to the mechanical forces exerted by the refiner plates. It also indicates the responses of fibres to different refining conditions such as temperature, plate gap and chemical treatments.

The longest Bauer-McNett fraction, R14, represent the intact fibres, separated or in bundles. The quantity of this fraction remaining in a mechanical pulp would indicate the extent of refining. Figure 8.3 shows that the fibres were shortened as a result of refining, particularly for the EW fibres due to their comparatively thinner cell wall. The effect of fibre cutting aggravated with increasing refining energy.

Interesting fact, refining at 160°C promoted cutting because of additional plate gap reduction at this high temperature when compared to the refining at a lower temperature such as 120 °C. According to the findings of Law [125], there are two principal forces in refining: compression and shear, and the two forces have different effects on the fibres, as shown in Figure 8.2. The shear force, due to its inherent nature, is mainly responsible for fibre separation, and fragmentation including peeling, cutting, splitting and external fibrillation while the compression force contributes to fibre deformation, flexibilization,

delamination (internal fibrillation). The latter increases the fibre flexibility. During high temperature refining, as the refiner plate gap decreases, the shear force increases, causing more fibre cutting.

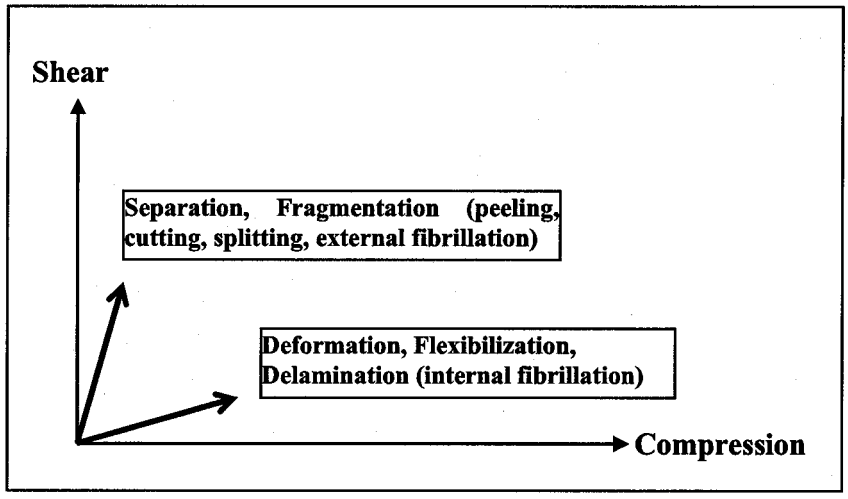

Figure 8.2 Contributions of shear and compression actions in refining

The particular effect of fibre shortening in high temperature refining was also evidenced in other fractions such as R28 and R48, as shown in Figure 8.4 and 8.5, respectively. Since the sample MIX consisted of EW and LW, its curve tended to lie between those of EW and LW.

While the R14 fraction represents the intact fibres remaining after refining, the short fibre fractions such as R100 (Figure 8.6) and R200 (Figure 8.7) correspond to the accumulated effect of fibre cutting induced by the refining action. Again, as seen in Figures 8.6 and 8.7, refining at 160°C promoted fibre cutting in comparison with the refining at 120 °C, and EW suffered more physical destruction as compared to LW.

The fines, P200, consist of ray cells, flake-like outer layer of cell wall and fibrils resulted from the cutting and/or peeling effects of refiner plates. Quantitatively, refining at 160 °C generated more fines from LW, as shown in Figure 8.8. However, there was little difference between LW and EW when the refining was carried out at 120°C, implying that there was less severe fibre cutting or peeling when the refining was conducted with a large plate clearance at this temperature.

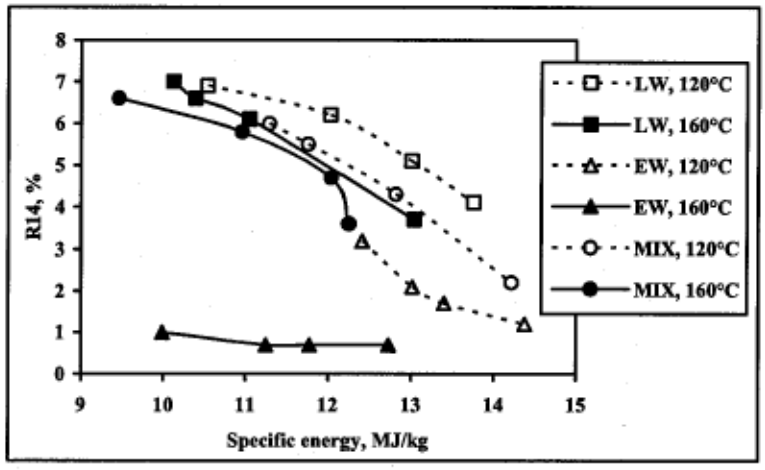

Figure 8.3 R14 fraction as a function of specific energy

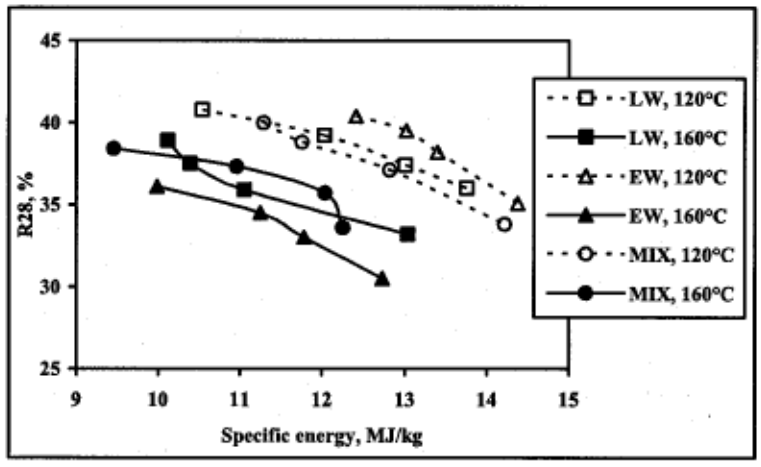

Figure 8.4 R28 fraction as a function of specific energy

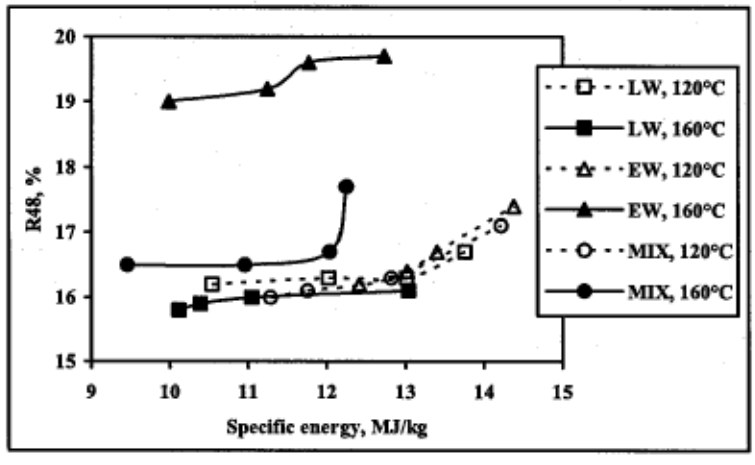

Figure 8.5 R48 fraction as a function of specific energy

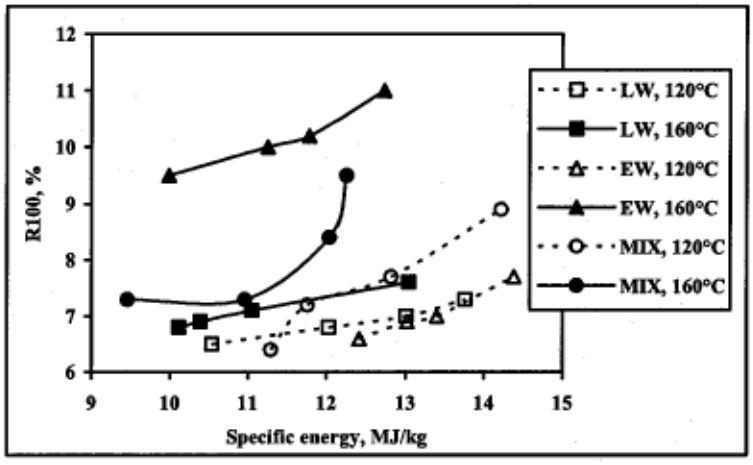

Figure 8.6 R100 fraction as a function of specific energy

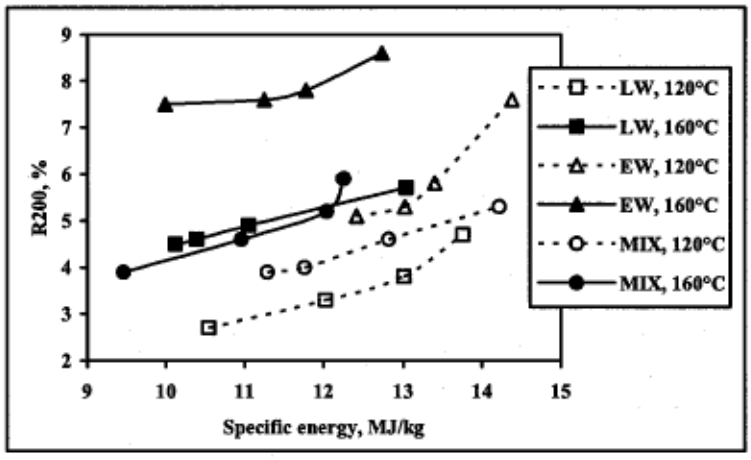

Figure 8.7 R200 fraction as a function of specific energy

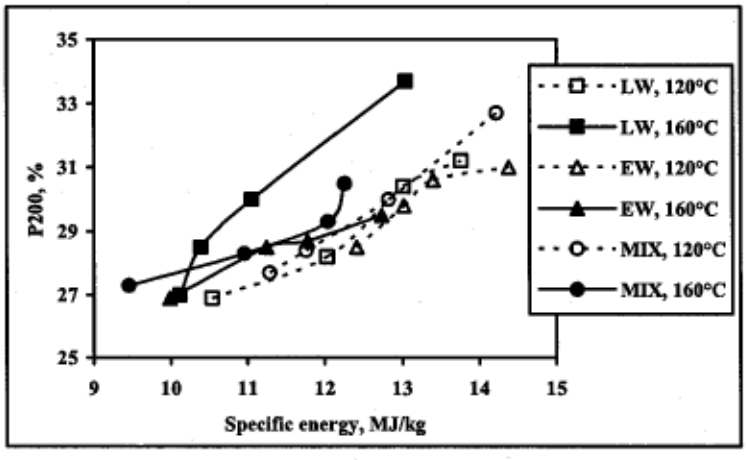

Figure 8.8 P200 fraction as a function of specific energy

8.3 Microscopic study of fibre fraction

Microscopic studies allowed us to gain useful information on the fibre development during refining. The general major characteristics observed for each Bauer-McNett fraction of EW and LW are presented by a series of photomicrographs, as represented in Figures 8.9-8.15. According to the microscopic observation, between the two temperature refining process (120°C and 160°C), within the EW or the LW fibres, the characteristics of fibre failure are very similar but different in quantity. For example, when compared with the low temperature refining (120°C), the high temperature refining (160°C) produces more fibrillation in LW fibre while more fibre splits for EW fibres mainly due to the reduced refiner plate gap. For the convenience of comparison in quality, LW and EW pulps employed in this study were produced at 160°C with a freeness of 150 mL.

The nature of the R14 fibres of LW (Figure 8.9) ranged from fibre bundles to individual fibres with a large variation of surfaces properties ranging from a smooth surface to a completely exposed S2 layer. Some fibres have most of their outer layer removed, exposing the S2 layer while the others remained undisrupted. Peeling of the outer layer or other types of rupture never occurred uniformly along the fibre length. For instance, a fibre could have its outer layer removed over most of its length, while the rest remain undamaged. Moreover, some fibres were compressed and twisted, even completely fibrillated (so-called "sleeve rolling" effect) by the refiner bar. As seen in the R14 fraction of EW (Figure 8.10): under the refiner forces, thin-walled EW fibres were readily ruptured, split and broken down in shorter segments. In addition, the fibrillation of EW fibres was not as noticeable as in the LW fibres. The diversity of surface structure of LW and EW TMP fibres render fibre characterization complicated. This also reveals a fact that the refining action is not uniform.

The R28 fraction of LW (Figure 8.11, A-D) also showed a complex nature of fibre surface ranging from "sleeve rolling" effect to complete exposure of S2 with intact or rupture cell wall. Often, two or more of these characteristics occurred over the fibre length. As for the R28 fraction of EW (Figure 8.11, E-H), splitting of fibre wall and

fibre cutting were the most evident features, some fibrillations had also taken place on the fibre surface.

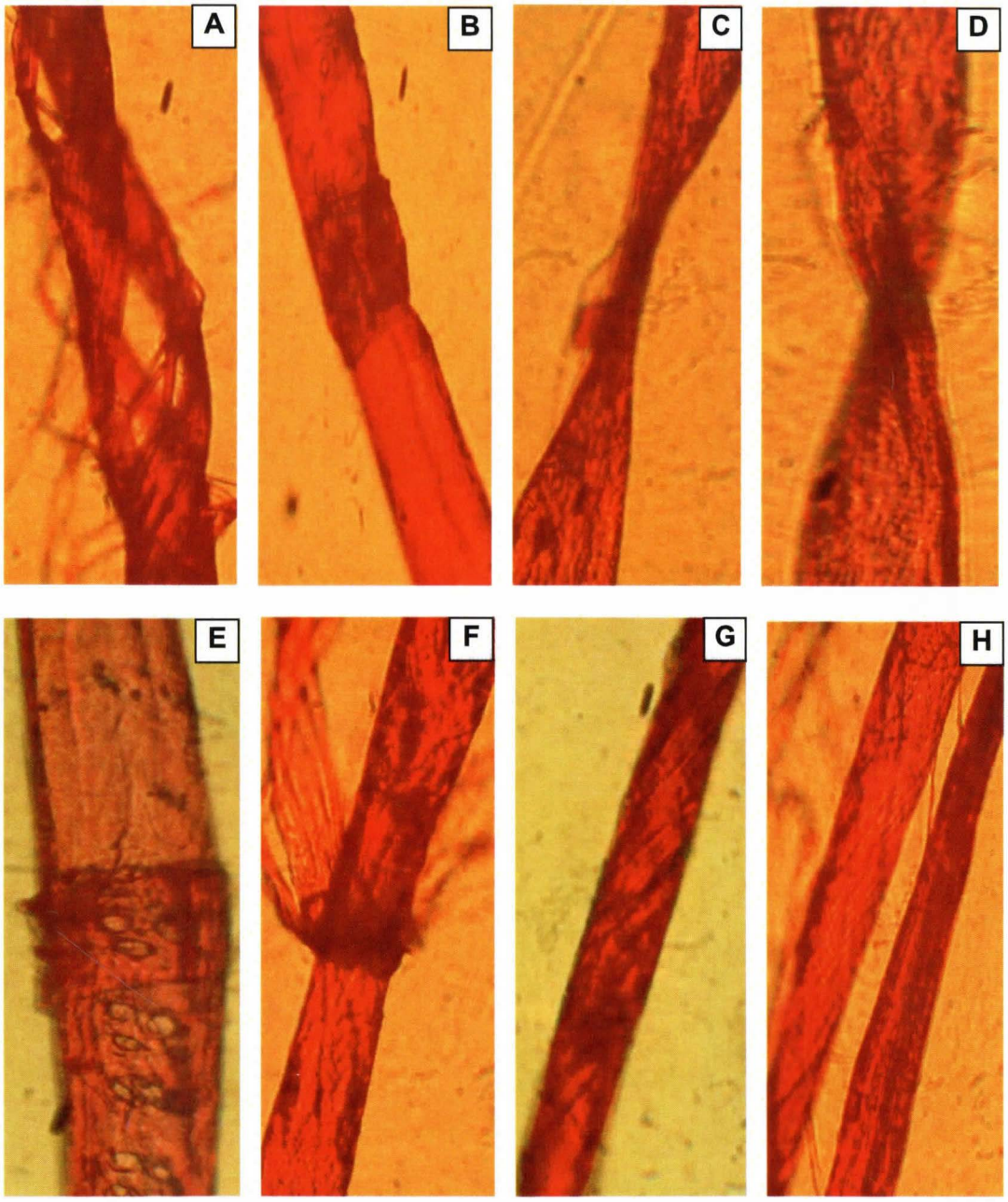

Figure 8.9 Micrographs showing the surface characteristics of LW fibres in R14 fraction (x400)

(A): Complete fibrillation of S2;
(B): A partly peeled fibre (the whole surface has been peeled at different degrees);
(C): Compressed and twisted zone probably caused by the edges of refiner bars; (D): A twisted fibre;
(E): Localized peeling around the fibre; (F): "Sleeve rolling" effect; (G): Fibrillation of S2;
(H): A non-peeled fibre and completely peeled one.

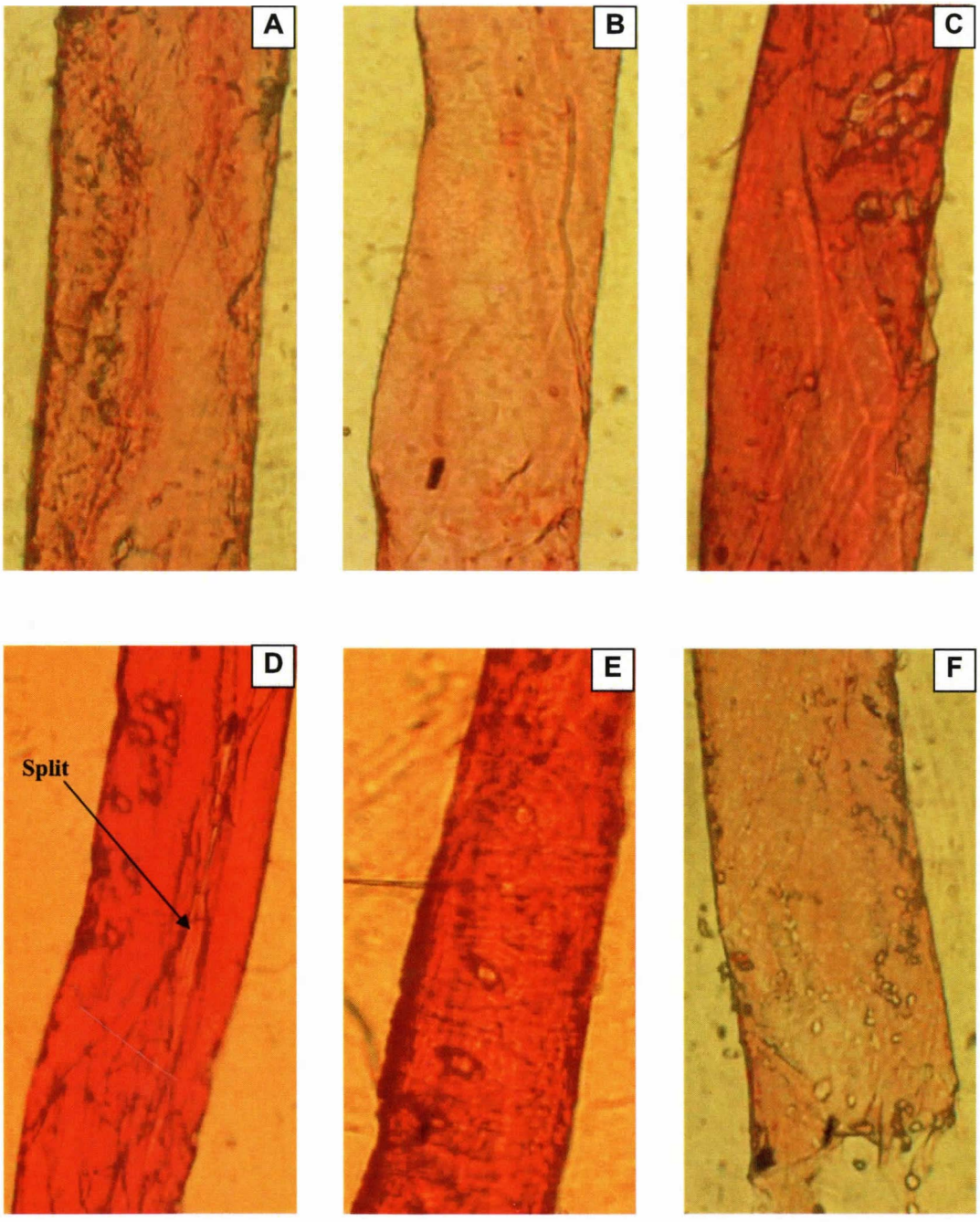

Figure 8.10 Micrographs showing the surface characteristics of EW fibres in R14 fraction (x400)

(A-C): unfibrillated fibres, showing smooth surface;
(D): A split fibre;
(E): A fibre with some fibrils on the surface;
(F): Cut fibre end with little fibrillation.

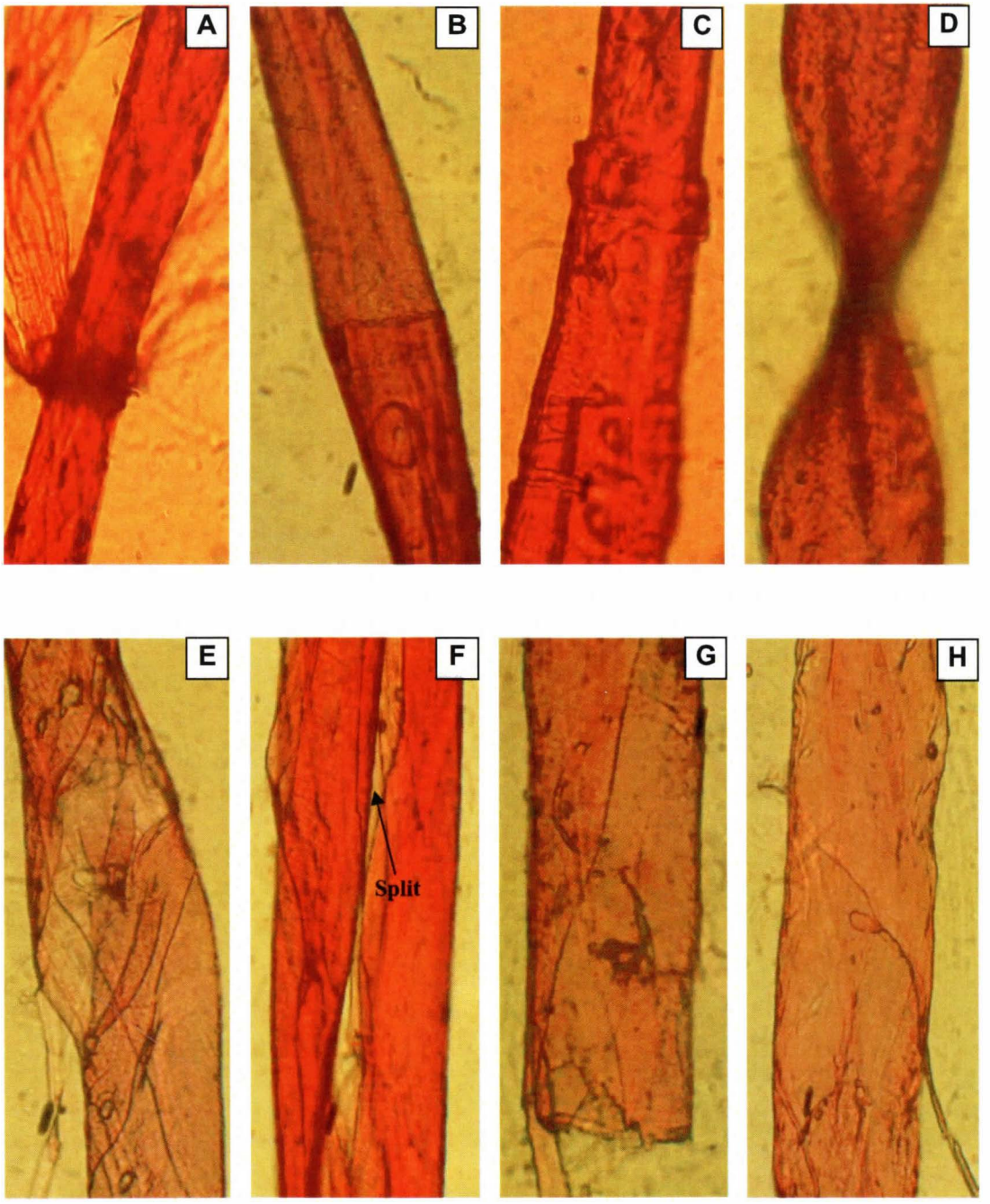

Figure 8.11 Micrographs demonstrating the refining effect on R28 fraction of LW and EW fibres (x400)

(A):"Sleeve rolling effect" and fibrillation of S2 on LW fibre;
(B, C) Partly peeled LW fibres;
(D): A twist LW fibre;
(E): Wall fracture in EW fibre;
(F): A split EW fibre;
(G): An EW fibre end with a few fibrils on the surface;
(H): A partly fibrillated EW fibre.

The LW R48 fraction (Figure 8.12, A, C, D, E) consisted of well fibrillated and developed fibres. In contrast, the fibre elements of R48 fraction of EW were twisted, split and fractured, as shown in Figure 8.12 (B, E, F, and G). The fibrillation was rather limited in these fibres.

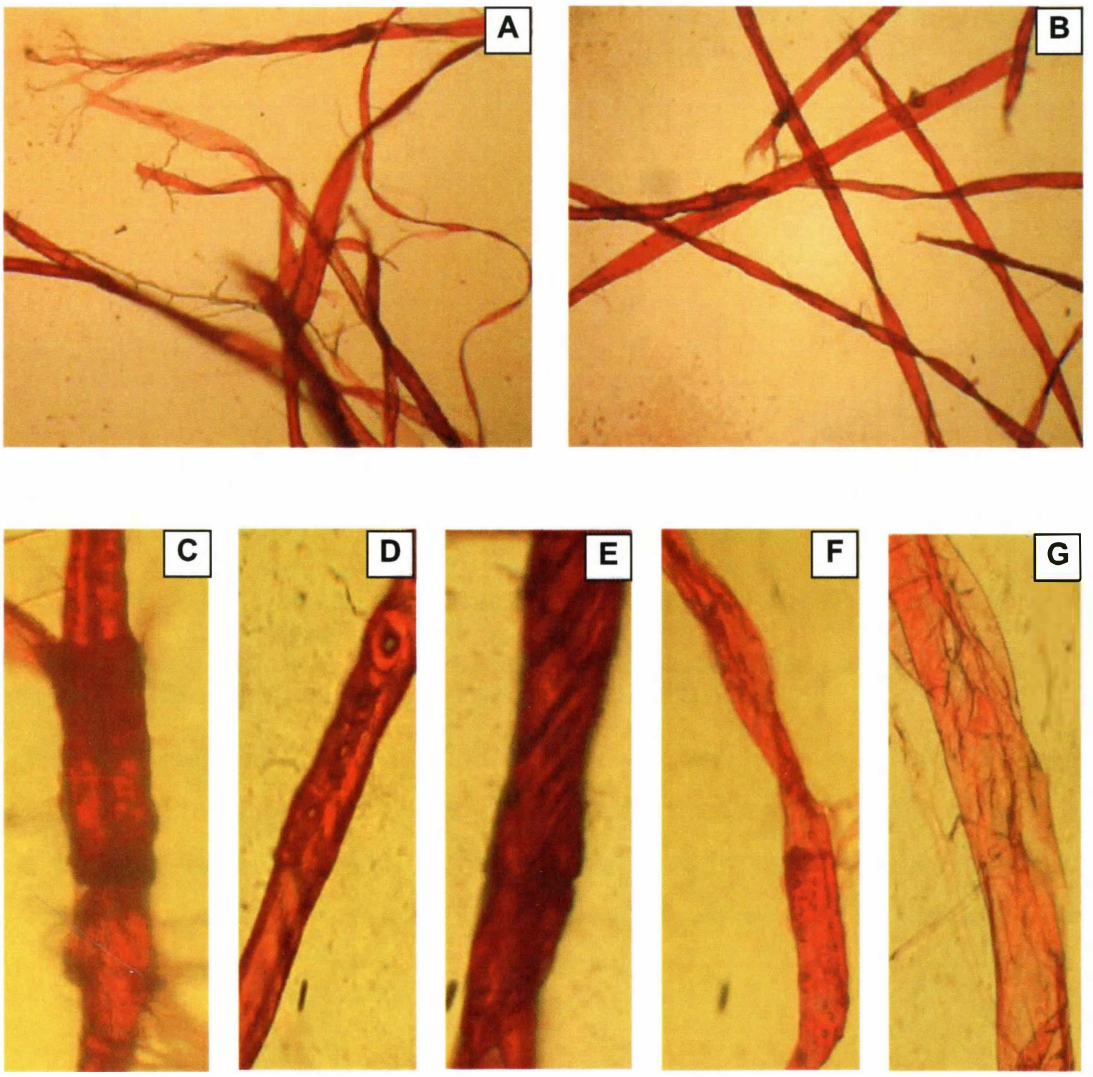

Figure 8.12 Micrographs showing the surface characteristics of fibres of LW and EW in R48 fractions

(A): LW R48 fibre (x40);
(B): EW R48 fibres (x40);
(C) "Sleeve rolling" effect on LW fibre (x160);
(D) Partly peeled LW fibre (x160);
(E): Fibrillated S2 layer in LW fibre, spiral cracks in fibre wall (x160);
(F): A twisted and fibrillated EW fibre (x160);
(G): Wall-fractured and split EW fibre (x160).

As seen in Figure 8.13 (A), the fibre elements of R100 fraction of LW showed fibrillated fibre ends and large bands of fibrils. In the R100 fraction of EW (Figure 8.13, B), there were many undeveloped and broken fibre segments. Almost all the fibrous elements were unfibrillated.

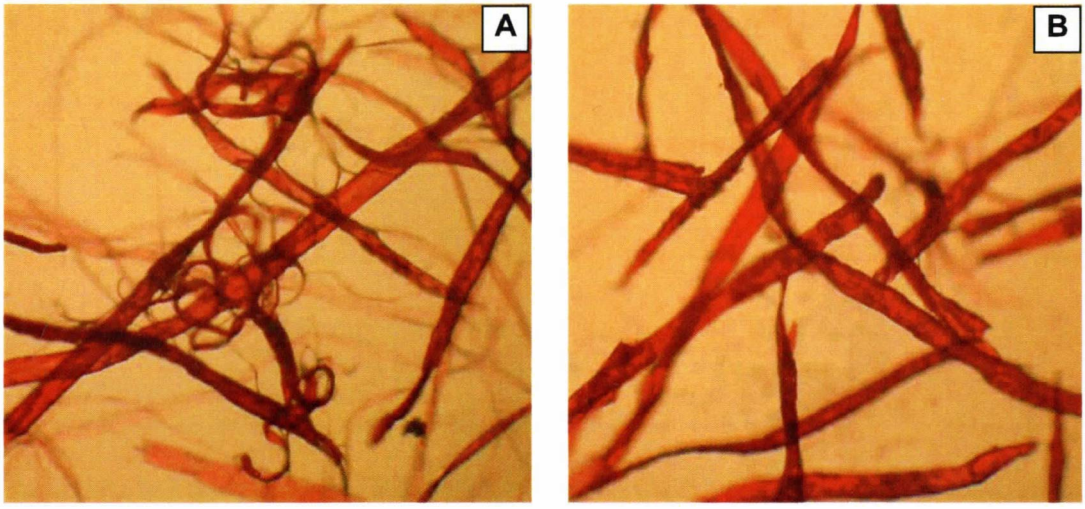

Figure 8.13 Micrographs showing the nature of the fibres of R100 fraction of LW and EW

(A): LW R100 fraction showing fibrillated fibre ends and large bands of fibrils (x40);
(B): EW R100 fraction showing broken fibre with little fibrils (x40).

The characteristics of fibres of the R200 fraction were very complex in terms of the size and shape of the elements. The fibres in the R200 fractions of LW (Figure 8.14, A) ranged from very fine bundles of fibrils, to large fragments of fibre wall while the elements of the R200 fraction of EW (Figure 8.14, B) were rather chunky, under-developed and broken.

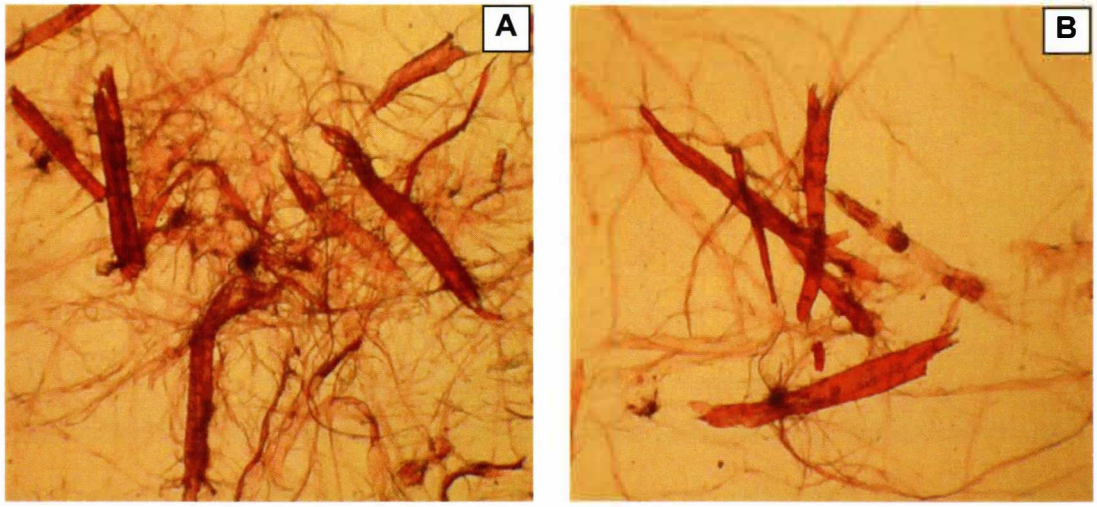

Figure 8.14 Micrographs showing the nature fibrous elements of LW and EW fibres in R200 fractions

(A): LW R200 fraction showing chunky developed fibre with bands of fibrils (x40);
(B): EW R200 fraction showing under-developed fibres and little fibrils (x40).

As shown in Figure 8.15, the P200 (fines) fraction was also complex in composition but with a different nature when compared with the R200. This fine fraction contained bundles of fibrils, flake-like fragments of various size and shape generated from the outer cell wall layer, and ray parenchyma cells. In the LW fines (Figure 8.15, A, B), the presence of annular fragments or broken bands of the outer layer indicated the delamination and separation of the outer layer around the secondary wall. Entanglement of fibrils during refining was also observed in this fraction. In addition, some pit borders are occasionally seen in LW fines. In EW fines (Figure 8.15, C, D), besides the annular fragments or broken bands of the outer layer, there were a lot of ring-shaped detached pit borders. In comparison with LW fines, EW fines showed little entanglements of fibrils.

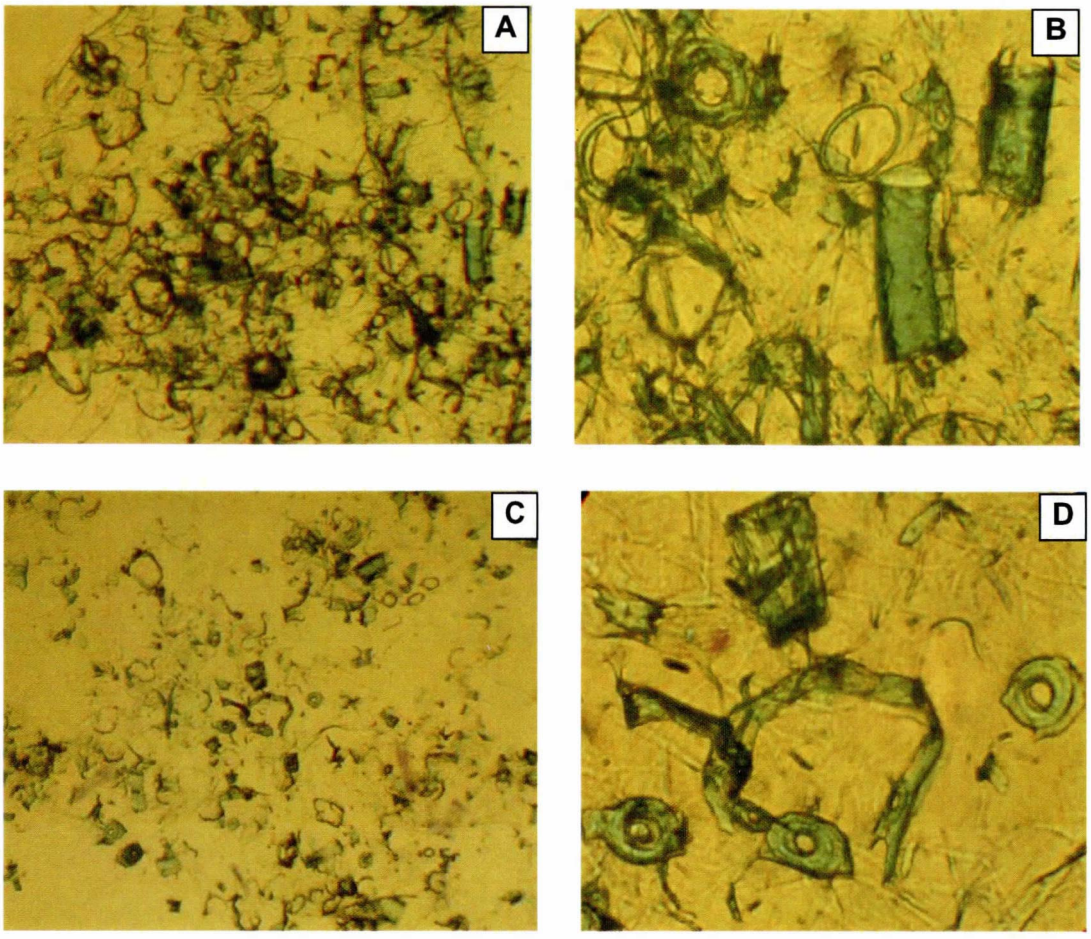

Figure 8.15 Micrographs showing the particles of P200 fraction (fines)

[A: LW fines (x40), B: LW fines (x400)]: LW fines show the presence of annular or straitened fragments of outer layer derived from delamination and separation around the secondary wall. The entanglement of fibrils is very noticeable. Some pit borders are occasionally seen in the LW fines;
[C: EW fines (x40), D: EW fines (x400)]: EW fines show some annular fragments of pit borders and their outer rings. There are little fibrils in EW fines.

8.4 Pulp characterization

8.4.1 Fibre length

As seen in Figure 8.16, with increasing specific energy, the fibre length decreased for all fibres of EW, LW and mixture of both. The adverse effect of high temperature (160°C) refining on fibre length was particularly evident when compared with refining at 120°C, which was more pronounced for the EW fibres. Interestingly, the fibre shortening effect caused by higher temperature was less evident with the mixture of EW and LW, which

may imply that the severe mechanical impact on EW was somewhat attenuated by the presence of LW fibres.

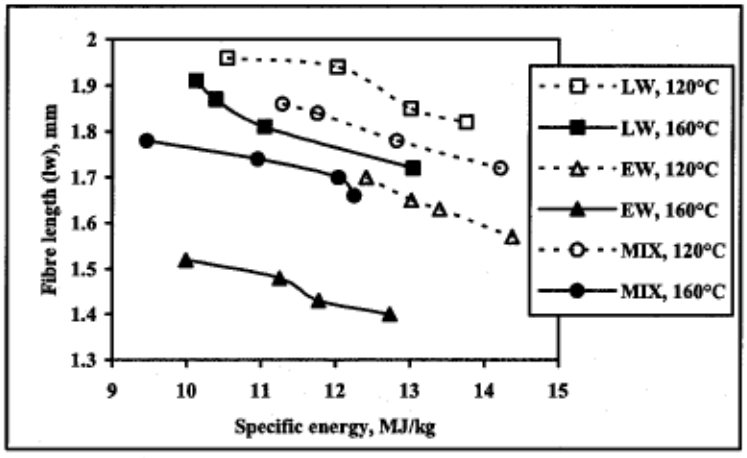

Figure 8.16 Fibre length as a function of specific energy

The fibre length reduction as a result of refining is presented in Figure 8.16; the comparison was made relative to the initial fibre length in wood. Since the thin-walled EW fibres are more flexible and collapsible than the thick-walled LW fibres, the refiner plate gap is slightly smaller for the EW refining than the LW refining as in the case of high temperature refining. Therefore, EW fibres experienced greater reduction in mean fibre length when compared to LW fibres. The phenomenon was observed at both refining temperatures; it was more significant at 160°C than at 120°C. This situation was due to the fact that narrower plate gap was used at 160°C, 0.40 mm vs. 1.20 mm.

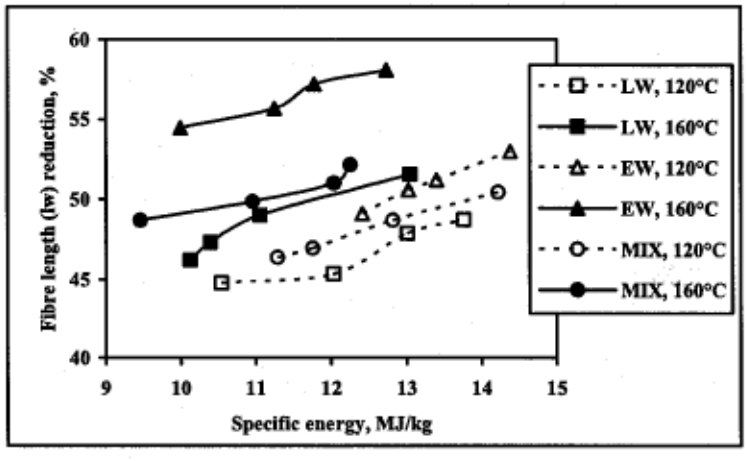

Figure 8.17 Fibre length reduction as a function of specific energy

8.4.2 Cell wall thickness

Cell wall thickness is an important morphological characteristic which affects the fibre stiffness and, as a result, influences inter-fibre bonding. The data on cell wall thickness provides us with the information on how the fibres response to the refining actions. As indicated in Figure 8.18, the cell wall thickness of fibres in R14 fraction, including EW, LW and mixture, decreased with increasing refining energy. The LW fibres showed the thickest fibre wall, while the EW counterparts the thinnest. The cell wall thickness of the fibres in the mixture lied between those of EW and LW. Moreover, the high temperature refining (160°C) seems to have slightly greater impact on thickness reduction when compared with low temperature refining (120°C). This is because refining at high temperature (160°C) with reduced plate clearance had greater shear impact on the fibres when compared with the low temperature refining. Similar trends were observed for fibres in R28 and R48 fractions (Figure 8.19 and 8.20).

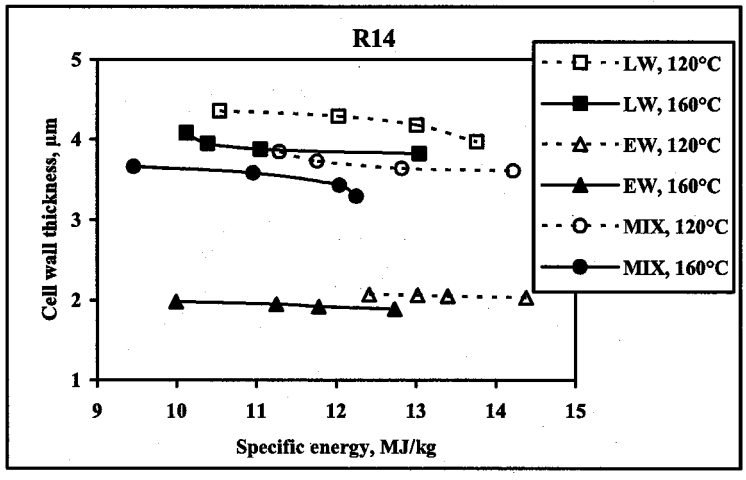

Figure 8.18 Cell wall thickness of R14 fraction as a function of specific energy

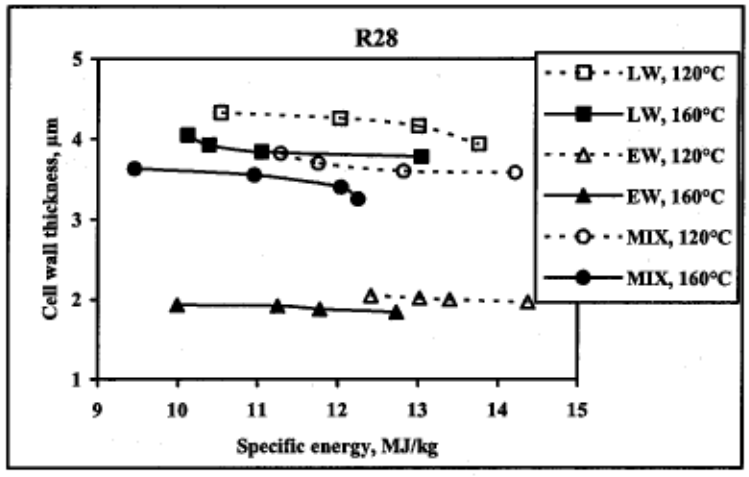

Figure 8.19 Cell wall thickness of R28 fraction as a function of specific energy

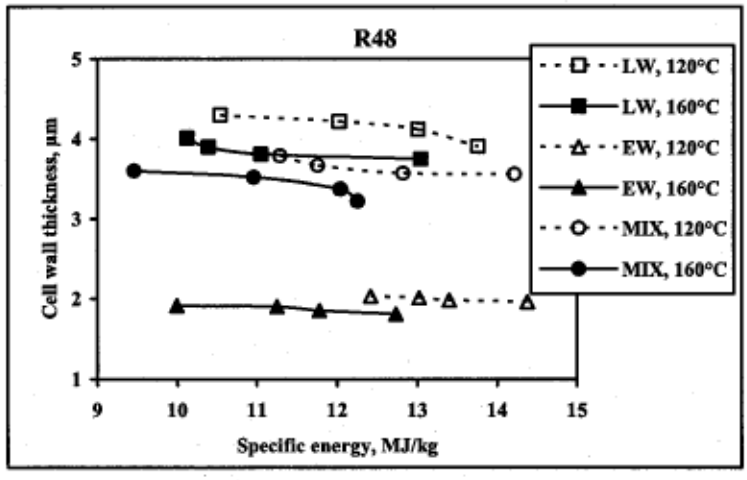

Figure 8.20 Cell wall thickness of R48 fraction as a function of specific energy

The images obtained with a Morfi cell wall analyzer (Figures 8.21 and 8.22) showed that the cell wall thickness diminishes gradually from R14 to R48 fractions, which is true for both EW and LW fibres. This means that fibre shortening and surface peeling occurred during refining, the longer fibres had thicker cell wall than the short fibre.

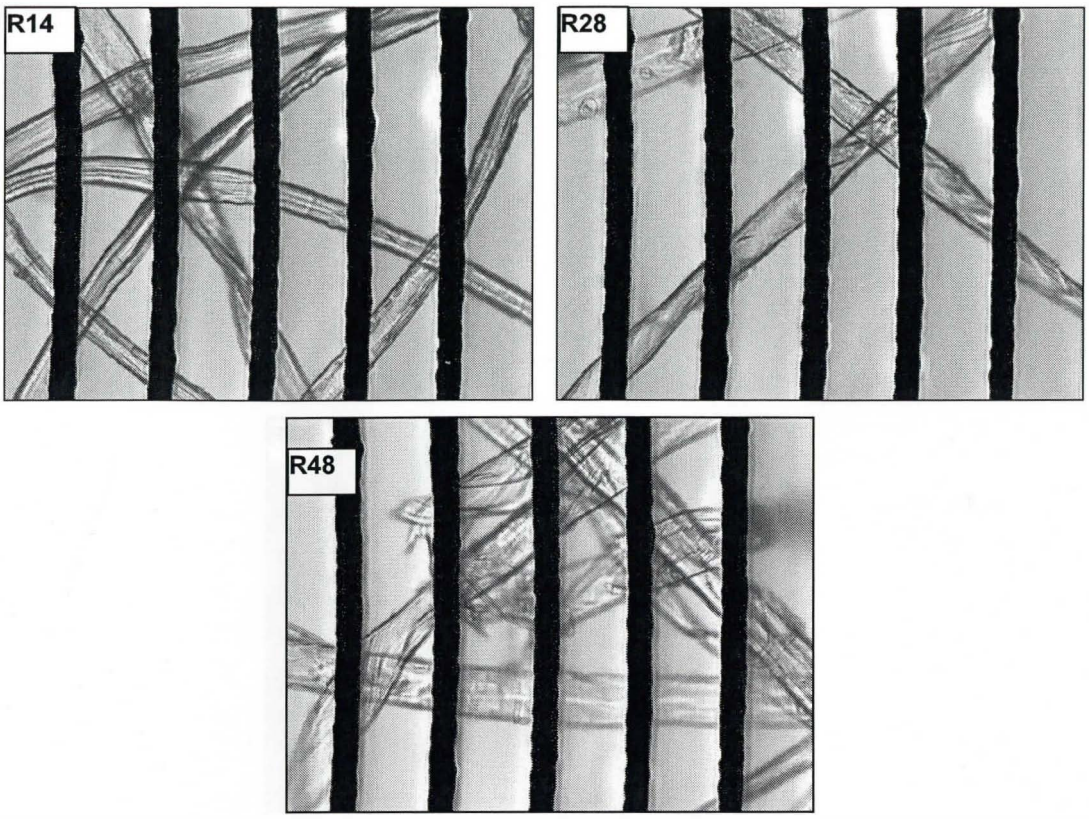

Figure 8.21 Image of cell wall thickness of LW fibres
(The fibres were fractionated from the LW pulp produced at 160 °C with a freeness of 150 mL)

The cell wall thickness reduction shown in Figures 8.23, 8.24 and 8.25 was based on the initial cell wall thickness in wood. These figures clearly show that the high temperature refining (160°C) operated with smaller refiner plate clearance had a greater effect on cell wall reduction as opposed to the lower temperature refining (120°C). These figures also indicated that the thick-walled LW fibres suffered greater reduction when compared to thin-walled EW fibres (Figure 8.23 and 8.24), which means that LW fibres exhibited more cell wall peeling and external fibrillation than those of EW fibres under the same refining conditions.

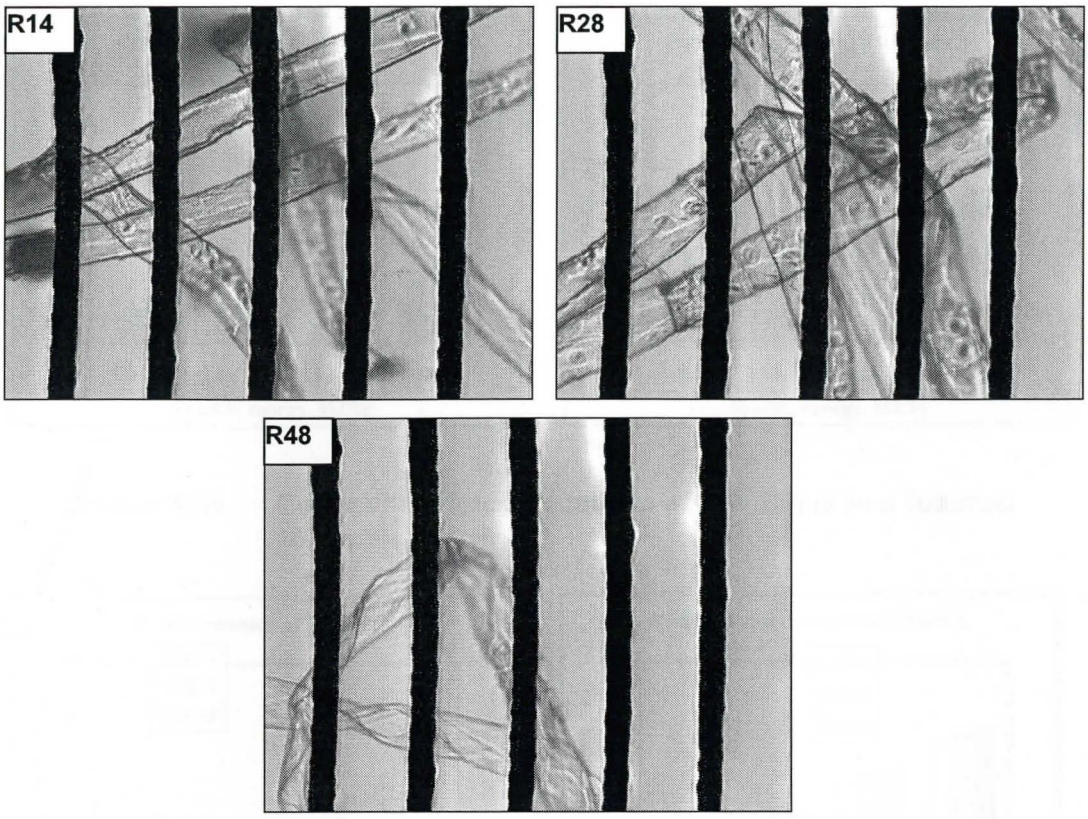

Figure 8.22 Image of cell wall thickness of EW fibres
(The fibres were fractionated from the EW pulp produced at 160 °C with a freeness of 150 mL)

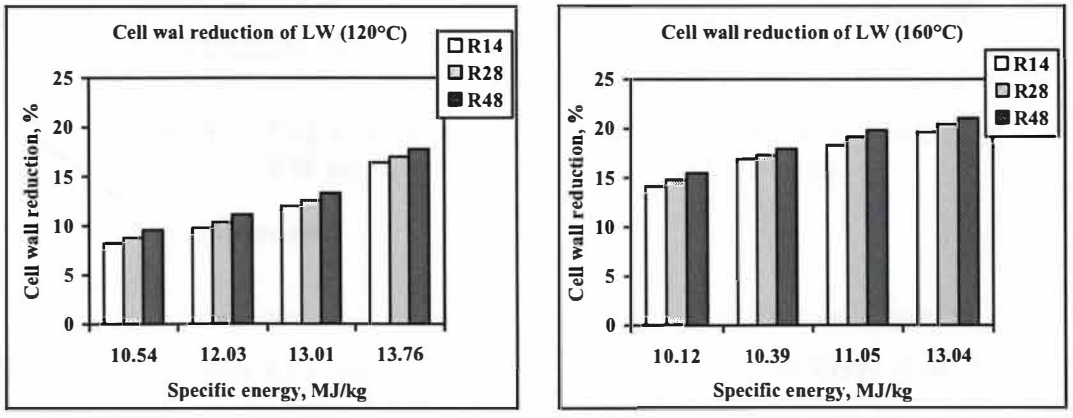

Figure 8.23 Cell wall thickness reduction of LW fibres as a function of specific energy

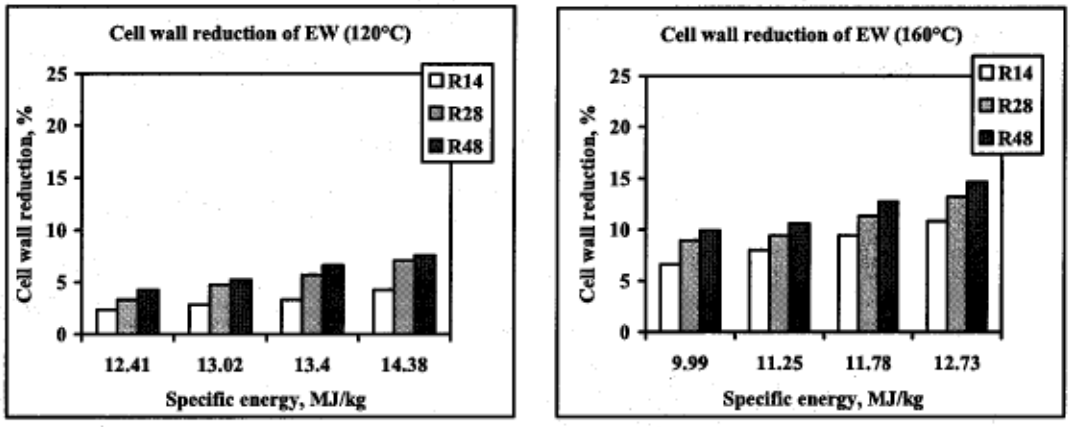

Figure 8.24 **Cell wall thickness reduction of EW fibres as a function of specific energy**

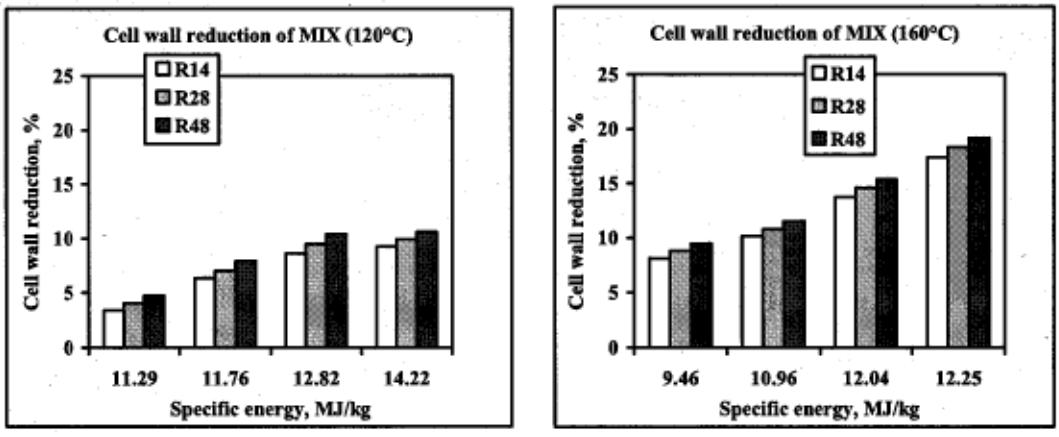

Figure 8.25 **Cell wall thickness reduction of fibres in the mixture of EW and LW as a function of specific energy**

8.4.3 Fibre coarseness

The change in fibre coarseness constitutes another feature of fibre development. As seen in Figures 8.26 and 8.27, the coarseness of the three types of fibres (LW, EW, and mixture) decreased with augmenting refining energy, the fibre wall being peeled off progressively as the refining action proceeded. Evidently, LW fibres were coarser than EW counterpart due to their initial thicker cell wall. Greater coarseness means fewer fibres per gram of pulp, and consequently fewer surfaces for fibre bonding [130].

Figures 8.26 and 8.27 also show that refining at 160°C had greater effect on the fibre cell wall peeling (cell wall reduction), resulting in lower fibre coarseness when compared with the refining at 120°C. This characteristic might be accounted for by reduced refiner plate gap at 160°C, favouring more fibrillation and cell wall peeling. In addition, the longer fibres tended to be coarser than the shorter ones, for example the R28 fraction (Figures 8.26) versus the R48 fraction (Figures 8.27)

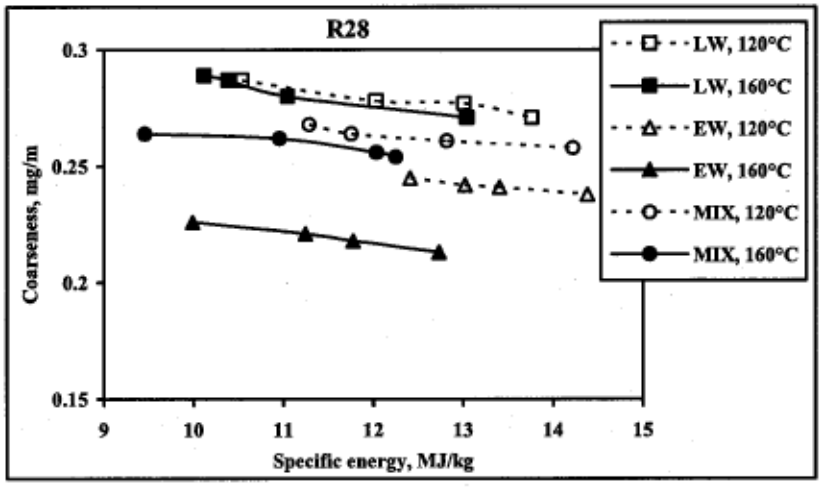

Figure 8.26 **Fibre coarseness of R28 fraction as a function of specific energy**

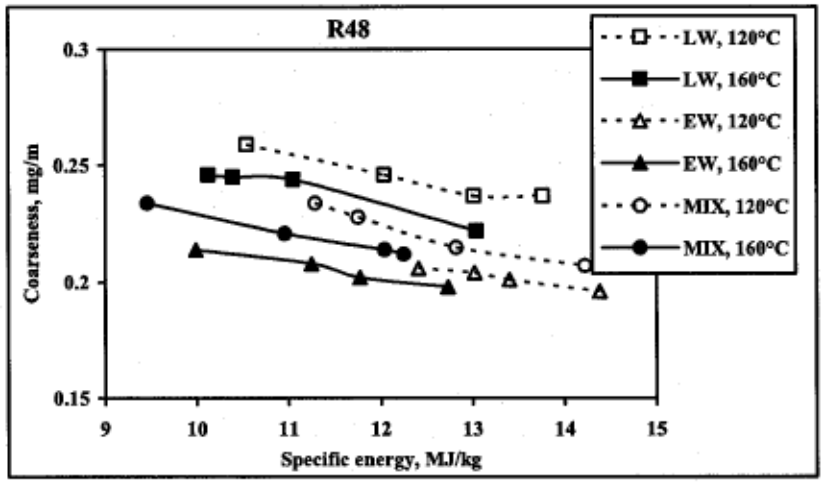

Figure 8.27 **Fibre coarseness of R48 fraction as a function of specific energy**

8.4.4 Curl index and kink index

Curls and kinks are introduced to the fibres during refining: under the moist atmosphere at high temperature which exists in refining, the fibres are subjected to cyclic deformations (compressive, tensile and shearing stresses). The stresses promote the flow of lignin and hemicelluloses, and the fibres under the influence of these stresses, are curled, twisted and kinked. This phenomenon is called "pulp latency" [172, 173]. The latency can be removed by means of hot water disintegration. When latency is removed from a pulp, curls and kinks are eliminated from the fibres and both the fibre flexibility and fibre straightness increases. Thus, the fibre contact surface increases resulting in the consolidation of fibre bonding. Consequently, the pulp freeness is reduced and the tensile strength of handsheets is significantly increased. Research also indicated that the hot water disintegration cannot completely remove the latency, there is a "residual" or "permanent" latency left in the pulp [174, 175]. A mechanism described by Page *et al.* [176] might explain the residual latency: under the high pressure and high temperature of refining, some paracrystalline regions in the cellulose fibrils are transformed to amorphous and viscoelastic regions. There are no stresses tending to restore it to its straight form. Therefore, the residual latency is left in the fibres.

In this study, a comparison was made to see the curl and kink changes between the "latent pulp" and "delatented pulp". The pulps were disintegrated with cold water at room temperature (about 20°C) were designated as "latent pulp". As a contrast, pulps disintegrated with hot water (about 90°C) were called "delatented pulp". The cold disintegration breaks the aggregates but removes little of the curl and kink from the fibres, while the hot disintegration disperses the aggregates and straightens the fibres by releasing the stress within the fibres. This comparison was made between the R28 and R48 fraction of EW, LW and mixture of both pulps. Since EW and LW fibres are morphologically different, they would behave dissimilarly and have different kink and curl indices after refining or after the latency removal process. Thus, it is necessary to compare these changes for EW and LW "latent pulp" and "delatented pulp".

Comparison between the latented (Figure 8.28) and delatented (Figure 8.29) pulps of R28 fraction reveals that the curl index was reduced by 87, 83 and 84% for EW, LW and

mixed pulps, respectively. It was observed that the curl index increased with increasing specific refining energy. It was also noted that thin-walled EW fibres had higher curl index than thick-walled LW fibres, indicating that the former is more curlable or more flexible than the latter. We found that the fibres produced at 160°C exhibited greater curl index in comparison with those prepared at 120°C. This characteristic might be due to the reduced plate gap at 160 °C, causing more shear friction between the fibre mass and refiner plates. We observed similar situation in the R48 fraction (Figures 8.30 and 8.31): EW fibres had higher curl index than that of LW; higher curl index at 160°C. Furthermore, comparison between the R28 and R48 fractions, the former showed higher curl index than the latter. This finding is in accordance with that of Law [130].

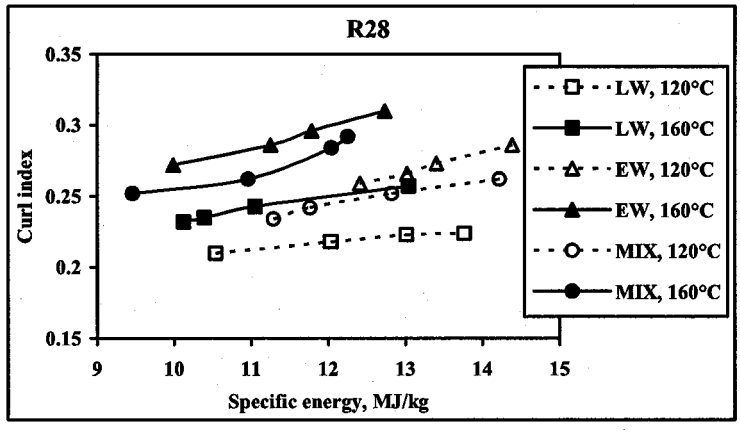

Figure 8.28 Curl index of latented fibres of R28 fraction as a function of specific energy

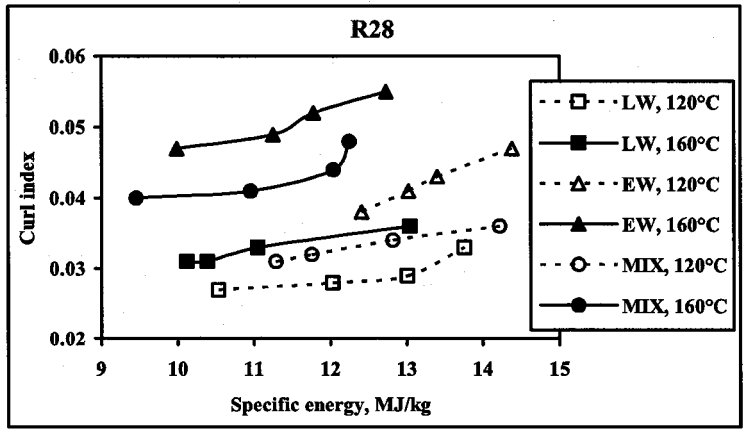

Figure 8.29 Curl index of delatented fibres of R28 fraction as a function of specific energy

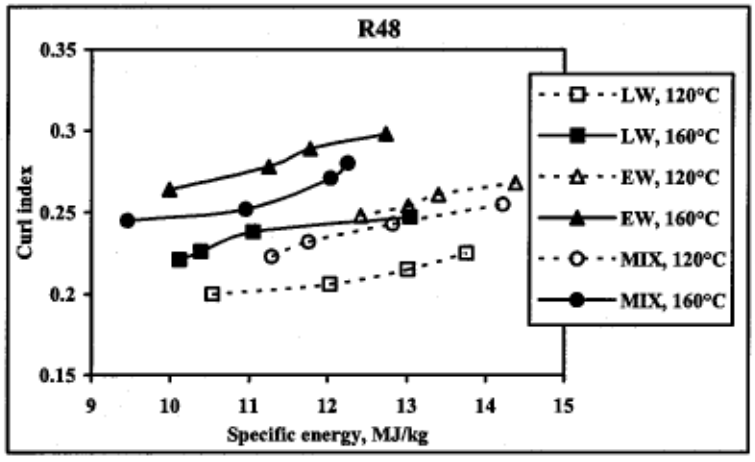

Figure 8.30 Curl index of latented fibres of R48 fraction as a function of specific energy

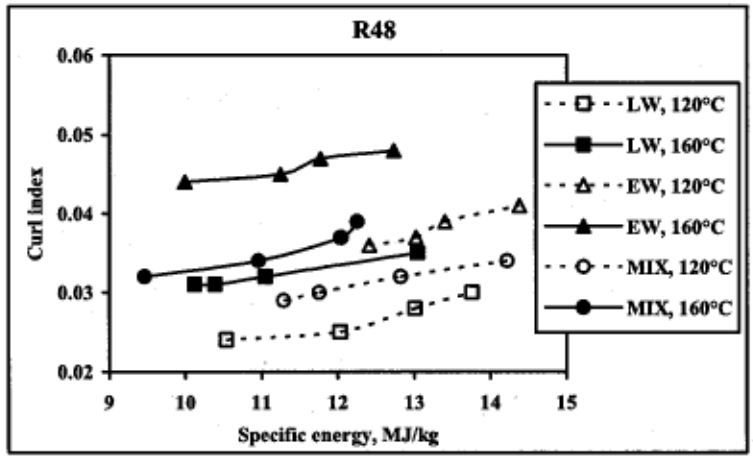

Figure 8.31 Curl index of delantented fibres of R48 fraction as a function of specific energy

Kink, which is the abrupt change in the fibre curvature, is another parameter used for characterizing fibre shape. The kink index indicates the fibres' behaviour during refining. As results of latency removal from the fibres of R28 fraction fibres (Figure 8.32 and 8.33), the kink index was substantially reduced, for example, by 73, 76, and 74% for EW and LW fibres and those in the mixture, respectively. We noted that the kink index increased for all the pulps as refining energy increased, and that thin-walled EW fibres had higher kink index than thick-walled LW fibres. This finding implies that LW fibres are more resistant to the mechanical actions of refiner bars. It was also observed that the increase in refining temperature from 120 °C to 160 °C provoked

higher kink index, which is probably due to the greater shear action caused by the reduced refiner plate gap. Similar tendencies were also observed for the fibres of the R48 fraction (Figures 8.34 and 8.35): EW fibres had higher kink index than LW fibres; high refining temperature incurred more kink fibres. In comparison with the fibres of R48 fraction, those of R28 fraction had greater kink index, i.e. the longer fibre (R28) exhibited higher kink index than the shorter ones (R48), which is similar to the tendencies described for the curl index.

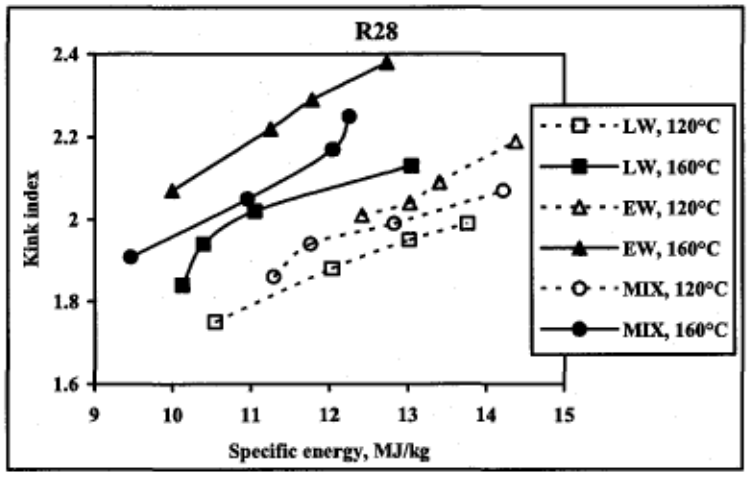

Figure 8.32 Kink index of latented fibres of R28 fraction as a function of specific energy

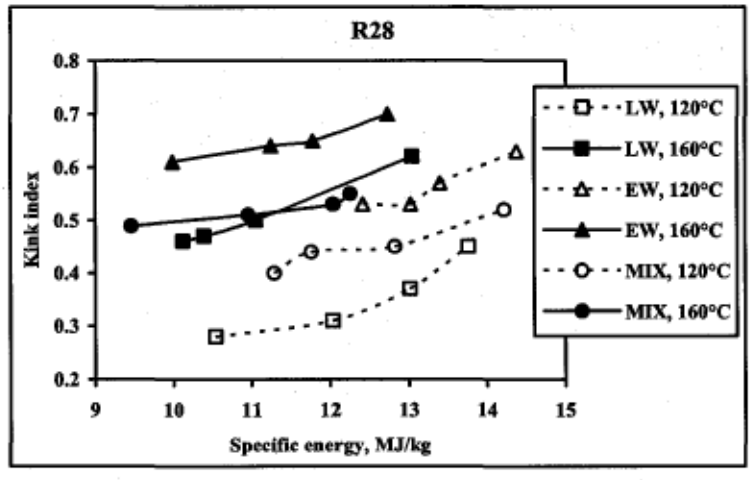

Figure 8.33 Kink index of delatented fibres of R28 fraction as a function specific energy

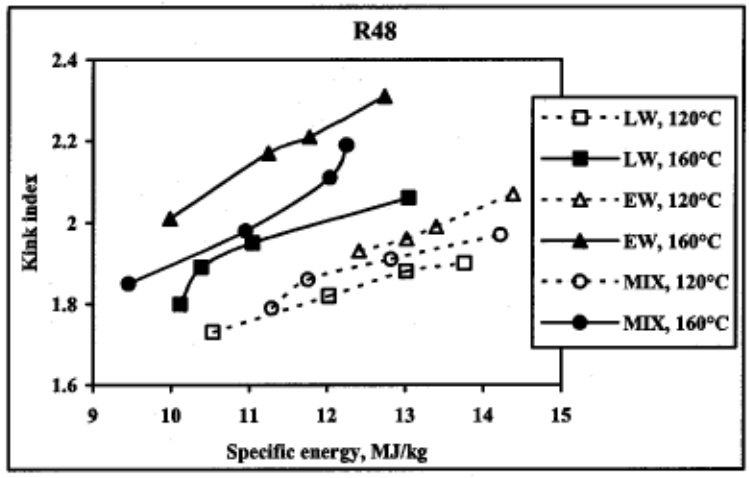

Figure 8.34 Kink index of latented fibres of R48 fraction as a function of specific energy

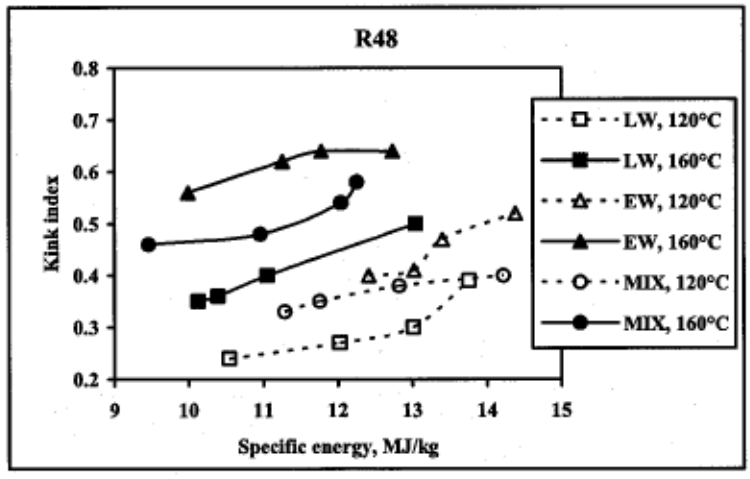

Figure 8.35 Kink index of delatented fibres of R48 fraction as a function of specific energy

8.4.5 Rejects

Light microscopic observation revealed that the rejects were composed of fibre bundles (shives) and aggregates composing of fibrils and separated individual fibres, as shown in Figure 8.36. The former are the non-separated fibres while the latter consists of partially or wholly separated fibres which were entangled together by means of fibrils. The presence of aggregates would increase the rejects rates in screening. As seen in Table 8.1, the shives accounted for more than 70% (numeric percentage) of EW rejects and less than 30% of LW rejects. On the other hand, LW rejects had more aggregates (about

70 %) than the EW rejects did (less than 30%). This finding suggests that LW fibres are more readily separated and fibrillated compared with the EW counterparts due to the difference in the cell wall thickness. Thick-walled LW fibres are more resistant to the refining actions and experience more surface peeling effect. In contrast, the thinned-walled EW fibres are more fragile and yield readily to the mechanical forces and break down more likely into unfibrillated fragments. As for the mixed furnish the percentage of shives and aggregates are nearly equal.

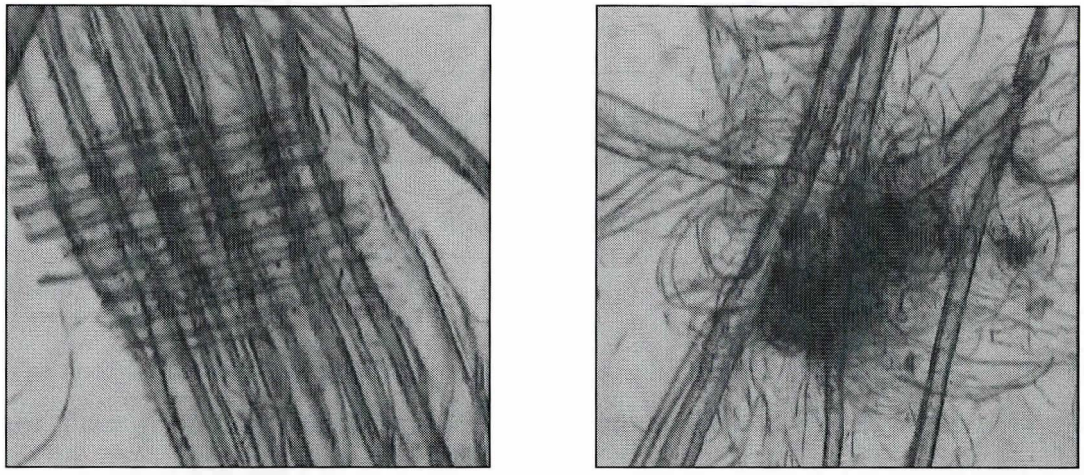

Figure 8.36 Basic elements in rejects
[A: EW bundles (shives); B: LW aggregates]

Table 8.1 Composition of the rejects of TMP

Sample*	Shives**, %	Aggregates**, %
EW	~70	~30
LW	~30	~70
MIX	~50	~50

*All the pulps were produced at 160 °C with a freeness of 150 mL;
**Numeric percentage.

Moreover, the high temperature (160°C) refining operated with reduced plate gap increased significantly the rejects of LW, but had no significant influence on that of EW (Figure 8.37). Due to the reduction of refiner plate gap, LW fibres are fibrillated more readily, generating more fibril elements and as a result more aggregates in the rejects.

However, the high temperature refining had little influence on the rejects production in EW pulps since the fibrillation is mostly limited to thick-walled LW fibres.

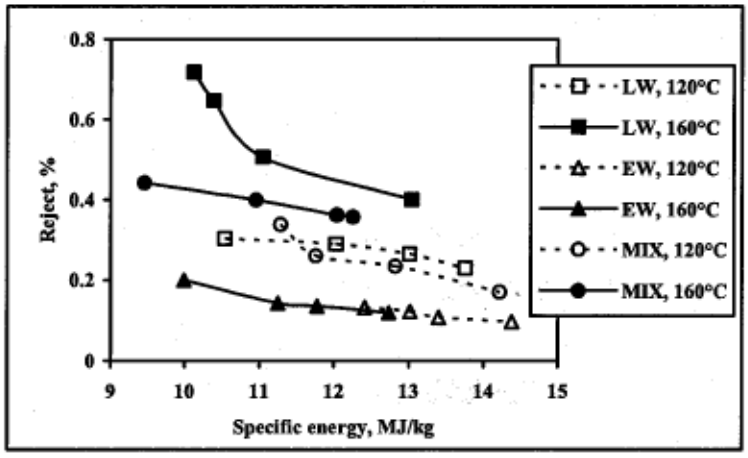

Figure 8.37 Rejects as a function of specific energy

8.4.6 Specific volume

The specific volume (SV) measurements of fines, which indicates the physical nature of the fines fraction provides us with useful information on the mechanism of the breakdown of EW and LW in refining. As discussed in chapter 6, the mechanical pulp fines consist of three components: fibrils, flake-like particles, and ray cells. The fibrils are cellulose-rich material with a high specific surface area, while the flakes are lignin-rich outer layer of cell wall (e.g. the compound middle lamella) with low surface area [134]. The ray cells are brick-like thin-walled parenchyma cells which usually remain intact during refining.

Figure 8.38 shows that the SV of fines increased with increasing refining energy because more fibrils are generated with increased refining energy. The increase in fibril component in the fines led to an increase in SV. As mentioned earlier (Figure 8.15), refining thick-walled LW fibres produced more fibril elements when compared to the refining of thin-walled EW counterpart. As a consequence, LW fines had higher SV as opposed to EW fines.

In addition, the fines produced at 160°C had higher SV values than those generated at a lower temperature, 120°C, which is attributed to the increased fibrillation effect at 160°C due to the reduced refiner plate gap.

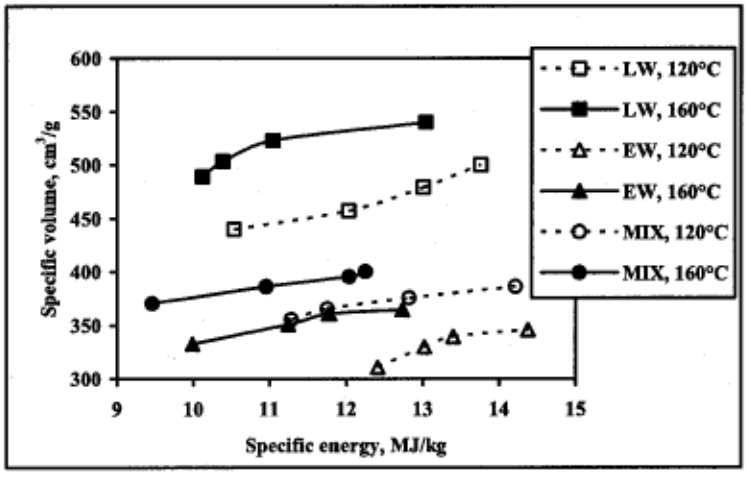

Figure 8.38 Specific volume as a function of specific energy

8.4.7 Water retention value

Water retention value (WRV) is a useful test for evaluating the water holding capacity of the fibres or the wetness of fibres [177]. The WRV of pulp is related to the external fibrillation as well as the internal fibrillation or delamination of cell wall. Splitting and delamination of cell wall under refining action facilitate water absorption, increasing the water holding capacity of fibres. Therefore, the WRV reflects the refining response of fibres: external fibrillation, cell wall split and internal fibrillation (delamination). All the effects ameliorate the swellability of fibres and inter-fibre bonding. [130].

As indicated in Figure 8.39, the WRV of R14 fraction fibres augmented slightly as the refining energy increased.

Figure 8.39 also shows that EW fibres have higher WRV than those of LW and those in the mixed furnish. According to Law [125], the internal fibrillation is mostly influenced by compression force in refining. Under a force, the thin-walled and flexible EW fibres collapsed and their cell wall fractured and cell corners became separated. These changes would improve the fibres' capability of absorbing of water. On the other hand, the thick-walled LW fibres being more rigid and resistant to the compression forces, they tend to

retain their form by separating in inter-fibre mode and exhibit little internal fibrillation [178]. Consequently, it is understandable that EW fibres have higher WRV than those of LW and those in the mixed furnish.

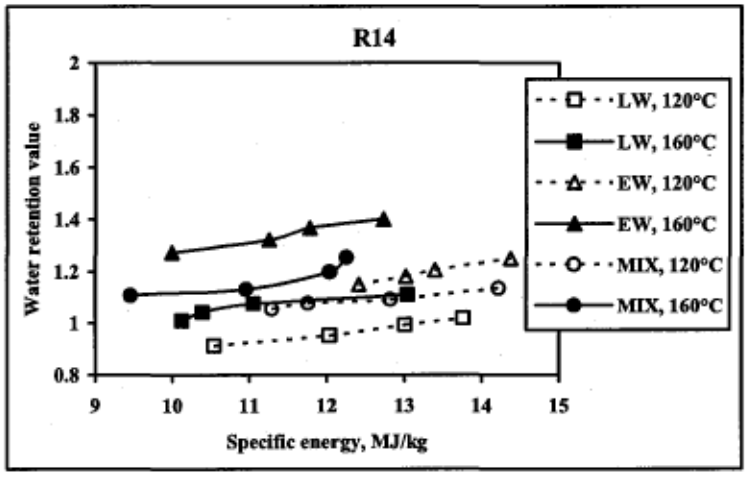

Figure 8.39 Water retention value of R14 fraction as a function of specific energy

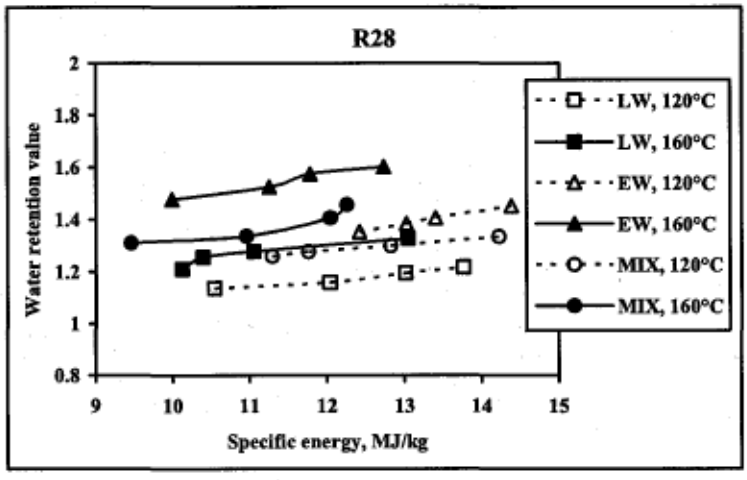

Figure 8.40 Water retention value of R28 fraction as a function of specific energy

The high temperature refining, thanks to the reduced refiner plate gap, had a greater influence on the WRV as compared with the low temperature operation at 120°C. At 160°C, the reduced plate clearance exerted more compression action against the fibres and produced greater amount of physical changes to the fibre structure. These changes improve the WRV of fibres.

Similar variation in WRV of other shorter fibre fractions (R28, R48 and R100) were also observed (Figures 8.40, 8.41 and 8.42): the WRV increased with increasing refining energy; EW fibres had higher WRV than LW fibres and those in the mixed furnish lay in between; higher temperature gave better WRV than the low temperature operation; the WRV increase with decreasing fibre length (shorter Bauer-McNett fractions).

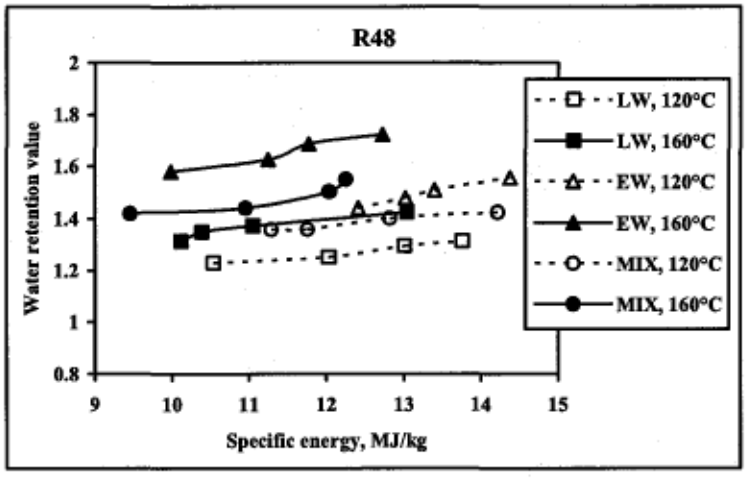

Figure 8.41 Water retention value of R48 fraction as a function of specific energy

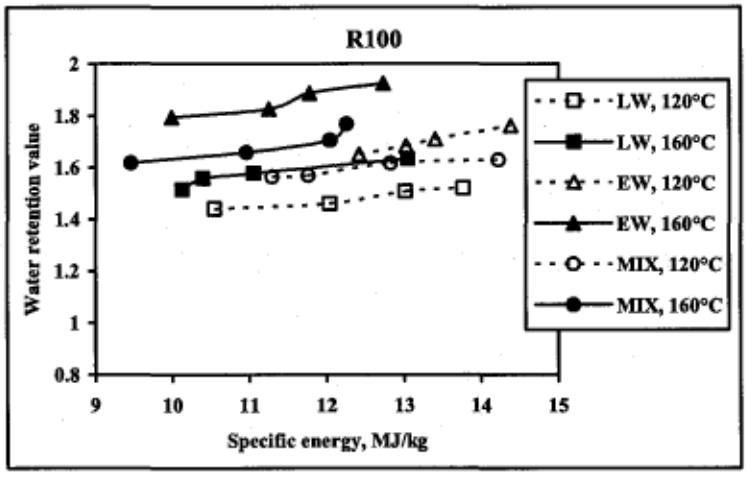

Figure 8.42 Water retention value of R100 fraction as a function of specific energy

8.5 X-ray photoelectron spectroscopy (XPS) analysis

X-ray photoelectron spectroscopy (XPS) analysis (or Electron Spectroscopy for Chemical Analysis, ESCA) is a useful and efficient means for characterizing the surface

chemical nature of fibres. The XPS data provides us with some supplementary information about fibre separation mode (for example, middle lamella/P, P/S separation, etc.) during refining. The principle of this analysis is based on the fact that the lignin concentration decreases gradually across the fibre wall, the highest concentration being in the middle lamella. Meanwhile the concentration of cellulose, which is practically absent in the middle lamella, increases from the primary wall towards the inner S layer [77].

In this study, we conducted XPS analyses on two types of pulp samples including the whole pulp without fines (fines-free pulp) and fines. Since the fines include flake-like particles and fibrils, which are generated from the fibre surface and S layer respectively, the analyses of fines and fines-free pulp permit us to understand the mode of fines formation. In addition, the refining temperature (or refiner plate gap) also influences the nature of fibre surface, so we took into account the effects of two refining temperatures. The pulp samples used had a freeness of 150 mL. Comparisons were made between EW, LW and MIX pulps.

Table 8.2 shows the results of the concentration of oxygen (O) and carbon (C) and the O/C atomic ratio. The surface coverage of lignin, $\varnothing_{lignin}$, was calculated from the O/C atomic ratio, as indicated in Eq.6.12 [155]. Figure 8.43 presents the distribution of surface lignin coverage in different samples.

As shown in the Figure 8.43: the surface lignin coverage of the fines is always higher than in the fines-free pulp for EW, LW or the mixed furnish. This is not surprising since the fines are the elements detached from the outer layer of cell wall. Since the lignin-rich middle lamella is the outmost layer of the cell wall, it is always the first to be peeled from the cell wall. As a result, in the fines, the surface lignin coverage is higher than in the fines-free pulp.

Table 8.2 The concentration of O and C and O/C atomic ratio of different samples

Sample	Atomic concentration, %		Atomic ratio
	O	C	
LW (WP-P200)*, 120°C	31.36	68.64	0.46
LW (WP-P200), 160°C	30.56	69.44	0.44
MIX (WP-P200), 120°C	30.95	69.05	0.45
MIX (WP-P200), 160°C	30.25	69.75	0.43
EW (WP-P200), 120°C	30.53	69.47	0.44
EW (WP-P200), 160°C	29.58	70.42	0.42
LW fines (P200), 120°C	30.38	69.62	0.44
LW fines (P200), 160°C	29.01	70.99	0.41
MIX fines (P200), 120°C	29.81	70.19	0.42
MIX fines (P200), 160°C	28.17	71.83	0.39
EW fines (P200), 120°C	29.32	70.68	0.41
EW fines (P200), 160°C	27.03	72.97	0.37

*WP-P200: fines-free pulp

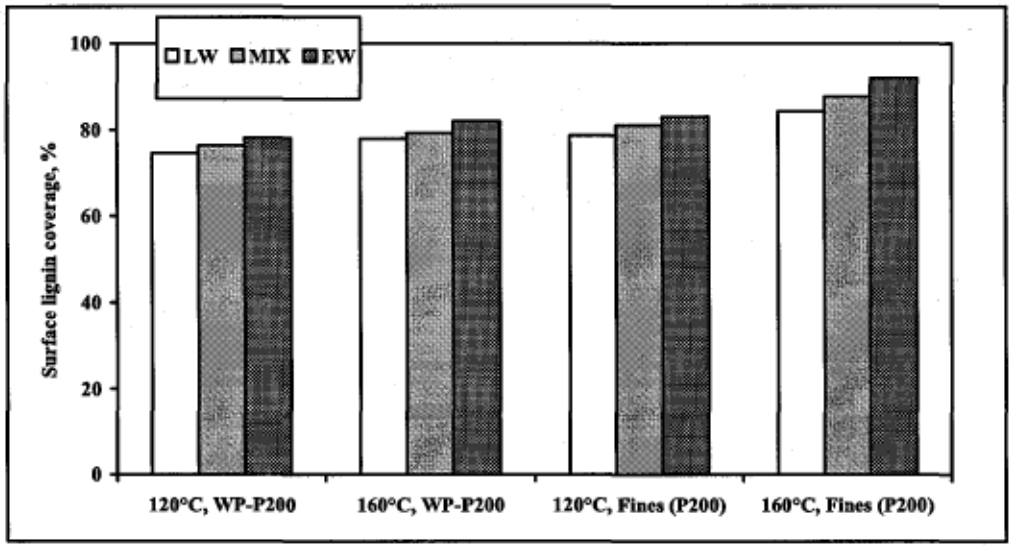

Figure 8.43 Surface lignin coverage of different samples

Among EW, LW and the mixed pulp, the surface lignin coverage of EW is the highest in both fines-free pulps and fines, and mixed furnish lies in between. This finding seems to support that the thin-walled EW fibres break down more readily with less surface fibrillation. The reduced fibrillation means that the surface of fibres maintains more lignin when compared to those which have suffered more fibrillation such as LW fibres. On the other hand, EW fibres have greater surface perimeter and hence produce more lignin-rich flakes in terms of surface. This might be another reason that EW fines have higher lignin content that those of LW and mixed furnish.

It was also observed that the higher temperature operation (160°C) yielded higher lignin coverage contents in the fines-free pulp and the fines when compared with low temperature process (120°C). As indicated in Table 8.3, at a given freeness, the fines contents are lower in the higher temperature refining (160°C), which means less cellulose-rich fibrils produced when compared lower temperature refining (120°C). For a given quantity of fines, the reduction in fibrils means higher concentration of lignin-rich flakes. Therefore, the surface lignin coverage increases as the refining temperature was increased. In addition, as indicated in Table 8.3, for a given freeness (150mL), EW pulps yields the highest fines content than those of LW and the mixed furnish. During refining, the fragile and thin-walled EW fibres tend to be cut more easily than thick-walled LW fibres, thus they produced more fines than the latter fibres.

Table 8.3 Fines content (% by weight) of pulps with freeness 150 mL

	LW	MIX	EW
120°C	28.2	29.5	30.8
160°C	27.8	28.3	28.6

8.6 Microscopic study

In this work optical microscopic evaluation was conducted to reveal the external fibrillation and cell wall failure (i.e. intra-wall and trans-wall failure) of EW and LW while scanning electronic microscopy (SEM) observation was used to quantify fibre cross-sectional deformation (Form circle and Form Shape), and qualify the internal fibrillation (delamination) and the mode of inter-fibre separation (e.g. middle lamella, S

layer separation, etc.). For each analysis, we examined at least 300 fibres per sample to render the data to be statistically significant. Due to the tedious sample preparation and time consuming observation, we studied only fibres of R28 fraction of the pulps having a similar freeness about 150 mL.

8.6.1 Light microscopy study

8.6.1.1 External fibrillation

As discussed in chapter 6, the external fibrillation is calculated by the comparison of fibre outer perimeter between the fibres with and without fibrils (Figure 6.9). The fibres having higher external fibrillation will show greater outer perimeter than the fibres less fibrillated. The degree of external fibrillation is calculated as in Eq. 6.9. The results are presented in Table 8.4, which shows that the LW fibres had higher degree of external fibrillation when compared to EW fibres and those in the mixed furnish. This finding is in line with the evidence provided by the microscopic observation, as illustrated in Figure 8.11. Under the refining forces, thick-walled LW fibres tend to fibrillate more readily than thin-walled EW fibres. Thus the so-called "sleeve rolling" effect occurred principally in the LW fibres, rarely in EW fibres.

Table 8.4 Quantitative data of degree of external fibrillation (%)

	120°C	160°C
EW	24.5	31.7
LW	75.2	88.6
MIX	58.4	62.1

The influences of temperature on fibre external fibrillation were noticeable, as shown in Table 8.4: high temperature refining with reduced plate gap produced more external fibrillation.

8.6.1.2 Fibre wall failure

As shown in Figure 8.44, the fibre failure could be of two types: trans-wall and intra-wall failure. In this study, the numeric percentage of each types of failure was assessed using a light microscope.

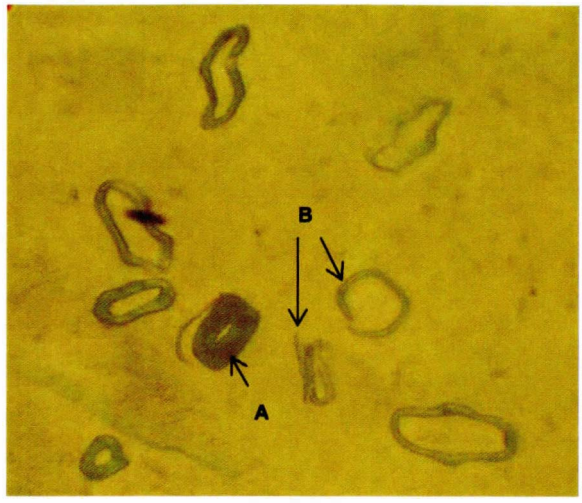

Figure 8.44 Cell wall failure of EW and LW fibres in refining
(A: Intra-wall failure in LW fibre; B: Trans-wall failure in EW fibre)

As seen in Table 8.5, most of the trans-wall failures occurred in EW fibres, while the intra-wall failures took place uniquely in LW fibres. The mixed furnish contains the two types of failure. This means that the cell wall failures are strongly influenced by the original morphological characteristics of fibre. Under the shear and compression actions, the thin-walled and flexible EW fibres tend to collapse (as shown in Figure 8.44) and split, resulting in the trans-wall failure. On the contrary, the thick-walled and rigid LW fibres, which are more resistant to the mechanical forces, retain their form and yield intra-wall failure.

Table 8.5 Numeric percentage of fibre wall failures in refining

	120°C		160°C	
	Intra-wall failure	Trans-wall failure	Intra-wall failure	Trans-wall failure
EW	0.3	26.3	0.3	30.6
LW	15.3	0.6	18.3	0.6
MIX	8.0	15.3	10.3	19.6

Table 8.5 also reveals that refining at high temperature with reduced refiner plate gap, thin-walled EW fibres experienced more trans-wall failure and thick-walled LW fibres showed more intra-walled failures. As explained earlier at high temperature refining (160°C) the plate gap had to be reduced to decrease the pulp freeness, resulting in increased shear force. As such, the mechanically weaker EW fibres split more easily and yielded higher trans-wall failure. However, the stronger LW fibres resist to the splitting action and tend to be fibrillated more and showed intra-wall failure.

8.6.2 Scanning electronic microscopy (SEM) study

8.6.2.1 Internal fibrillation (delamination)

Figure 8.45 shows the internal fibrillation of an EW fibre where the inner layer presumably the S3 layer of the secondary cell wall, was completely detached from the S2 layer. Also, some minor cracks are visible, indicating the presence of delamination. Interestingly, the nature of internal fibrillation is quite different for LW fibres where the S3 layer remained attached to the S2 layer (Figure 8.46). However, the bulk of S2 layer was fractured to a great extent. The distribution of cracks (delamination) in the S2 layer was uneven. Theses findings reveal that thin-walled EW fibres are much more fragile under the mechanical forces than the rigid and thick-walled LW. Therefore, EW fibres produce greater delaminations than LW fibres.

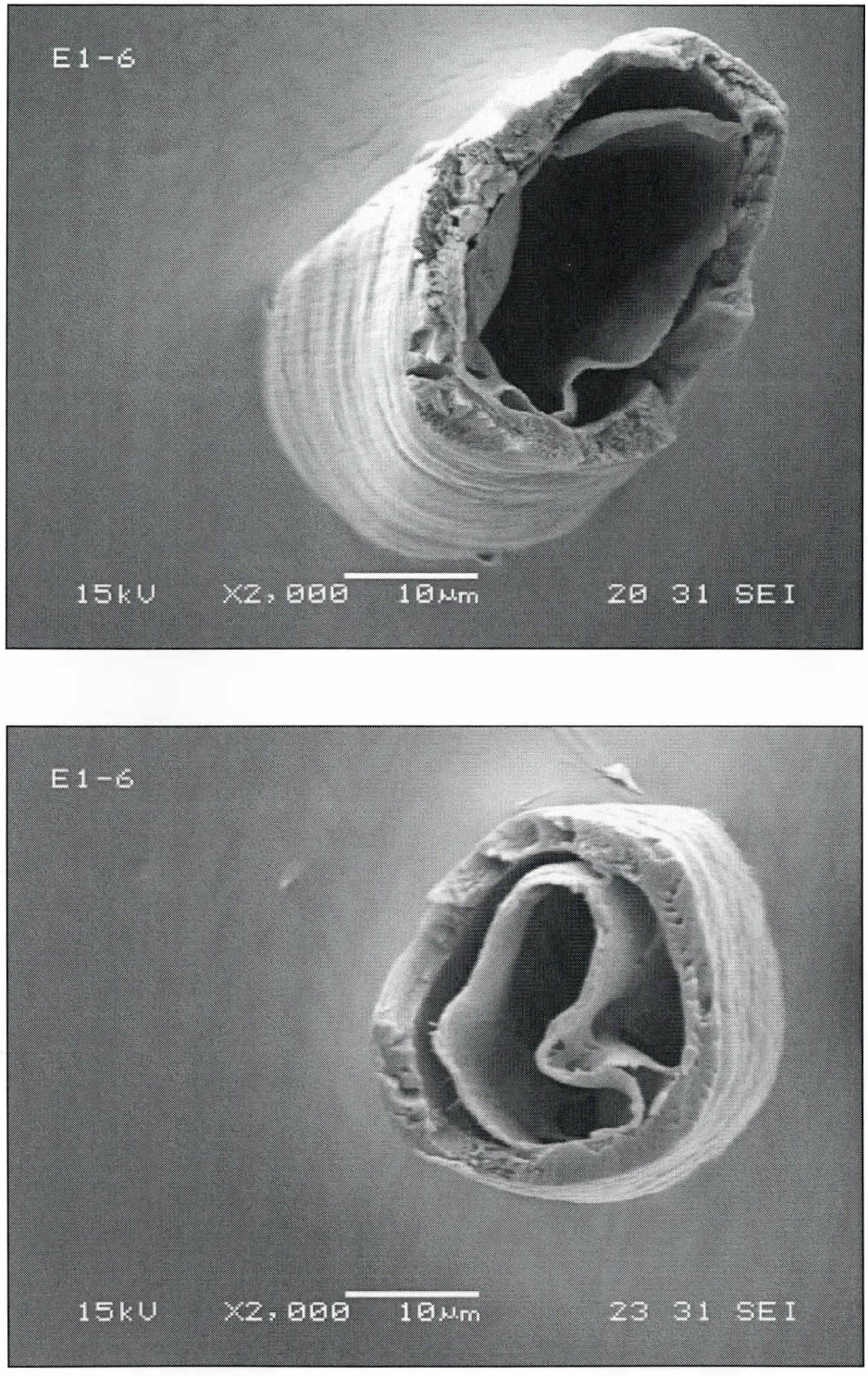

Figure 8.45 Internal fibrillation in EW fibres
(R28 fraction of EW pulp produced at 120°C with freeness 150 mL)

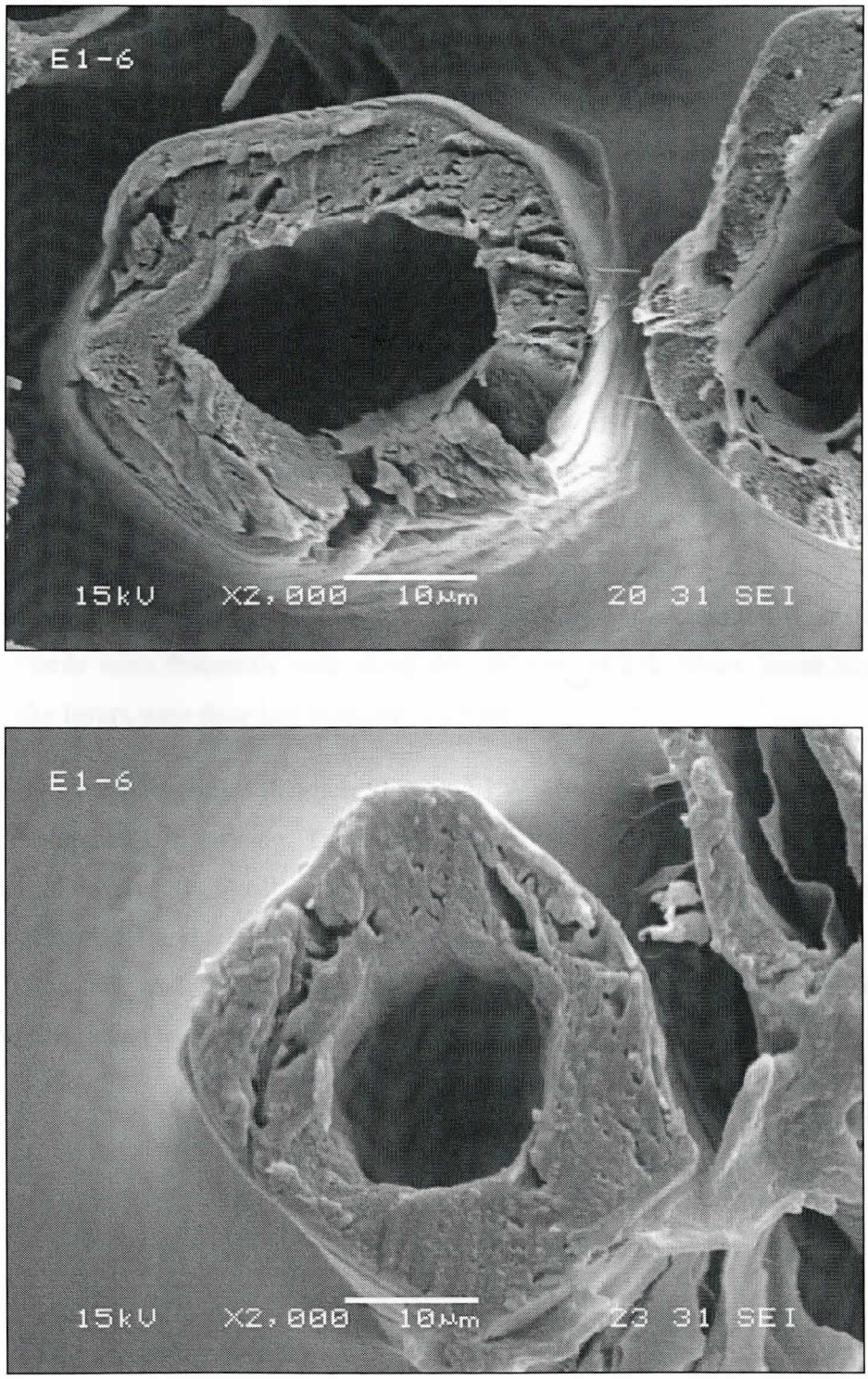

Figure 8.46 Internal fibrillation in LW fibres
(R28 fraction of LW pulp produced at 120°C with freeness 150 mL)

8.6.2.2 Fibre surface

After refining, EW fibres are completely collapsed, diminishing their lumens' volume, as shown in Figure 8.47. In fact, the compression and shear forces render EW fibres split and twisted (top micrograph Figure 8.47). The majority of the failures took place at the P/S1 interface, especially around the pits (bottom micrograph Figure 8.47). Failures at the S1/S2 interface were also occasionally noticeable (top micrograph Figure 8.47). Fibre external fibrillations associated with the shear force are not evident in EW fibres: a few fibrils were occasionally observed.

For LW fibres, the compression force has limited effects on the change in lumen dimension since the collapsed fibres are rarely observed (Figure 8.48). Most of the LW fibres showed various natures of the exposed surfaces such as the P, the S1, and the S2 (top micrograph Figure 8.48). However, the S1/S2 separation is commonly seen in LW fibres. Fibrils were frequently seen along the cell wall of LW fibres. Some long and ribbon-like layers were detached from the cell wall.

Due to the morphological differences, EW and LW fibres exhibited different nature of fibre developments in refining. The thin-walled EW separated principally at the P/S1 interface, showing little fibrillation. On the contrary, thick-walled LW fibres separated mainly at the P/S1 and S1/S2 interface with considerable amounts of fibrillation along the exposed cell wall. These findings are in agreement with the observation on the external fibrillation using an optical microscope.

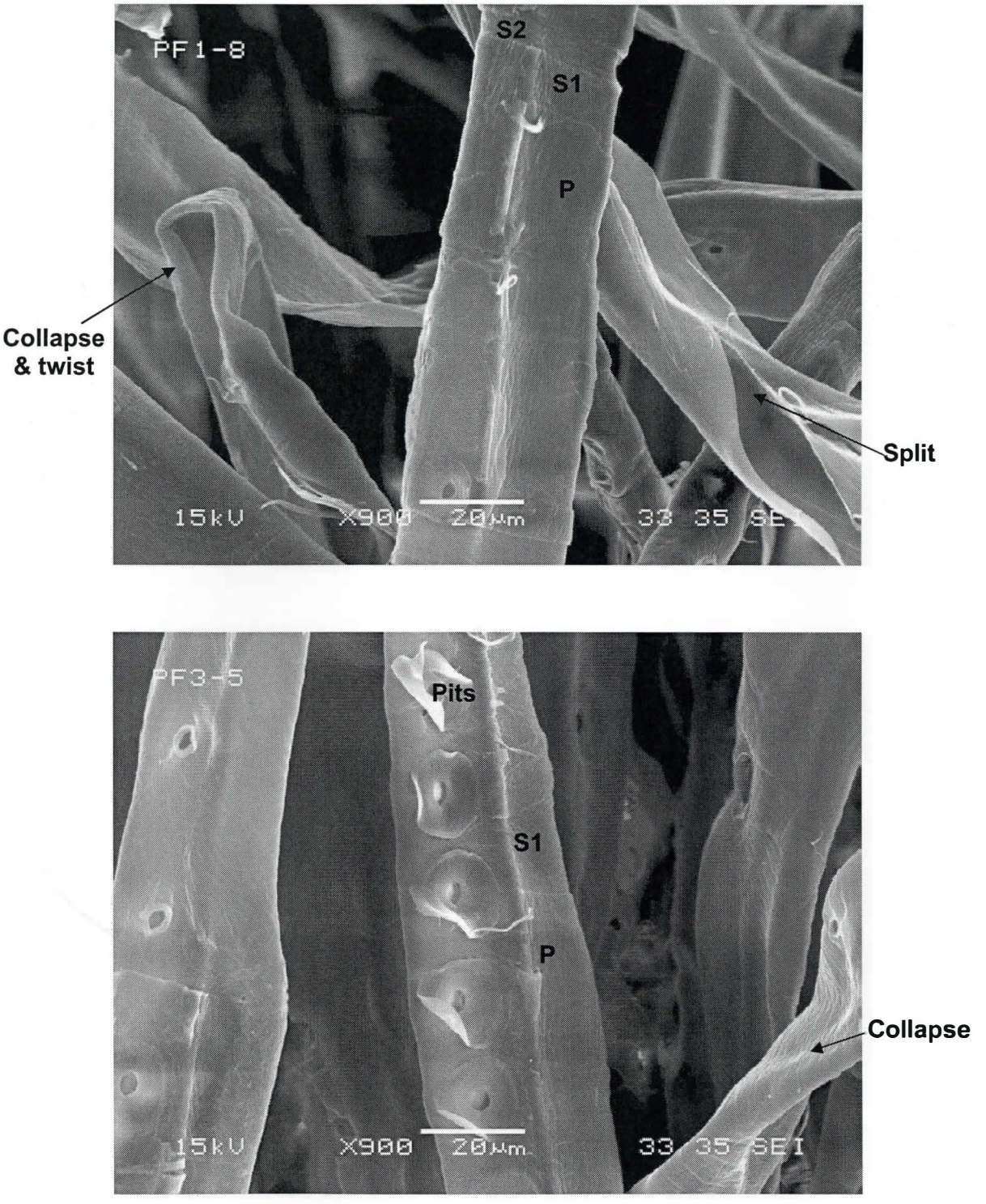

Figure 8.47 Surface nature of EW fibres
(R28 fraction of EW pulp produced at 120°C with 150 mL freeness)

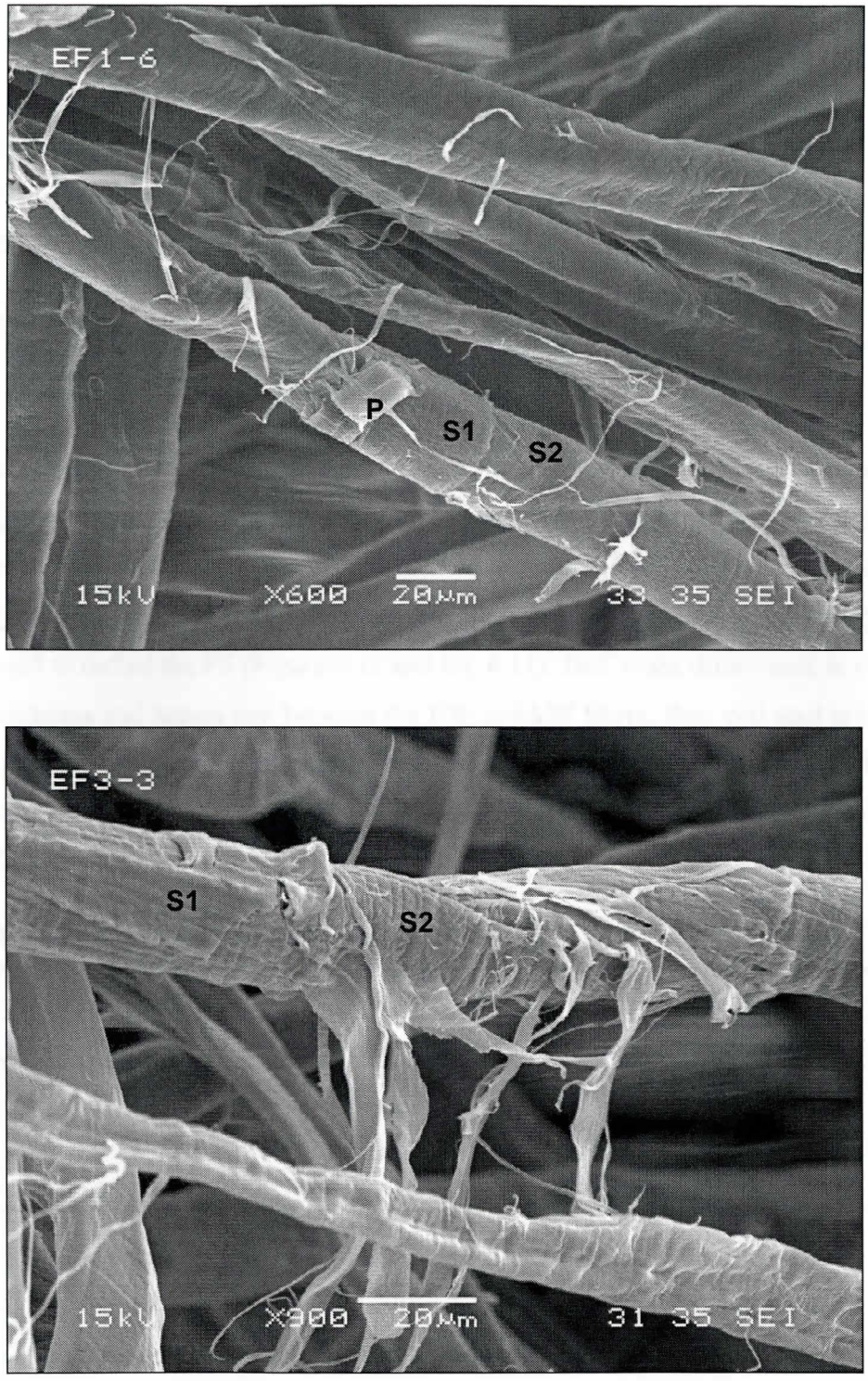

Figure 8.48 Surface nature of LW fibre
(R28 fraction of LW pulp produced at 120°C with 150 mL freeness)

8.6.2.3 Cross-section deformation

There exist great differences in cross-section characteristics between EW and LW fibres. The knowledge on the cross-section deformations of fibres is useful for assessing the overall quality of fibres. Such information would assist us to judge if special treatment should be given to some fibres to ameliorate papermaking properties and to predict the effect of processing variables on the fibres. The cross-sectional parameters, such as Form Circle (FC) and Form Shape (FS), reflect the fibre flexibility and collapsibility which are essential for the paper consolidation [105, 179, 180].

As discussed in previous chapter, both FC and FS are useful parameters to evaluate the fibre collapsibility. The cell wall area, lumen area and the outer perimeter of fibre are used in defining the FC equation (Eq.6.10). On the other hand, only the minimum and maximum distances measured between parallel tangents to the cross section are employed to define the FS (Figure 6.12 and Eq. 6.11). Due to the differences in the cell wall thickness and lumen size between the EW and LW fibres, their cell wall areas and outer perimeters are quite different. Therefore, when compared with the FS, the FC reflects more precisely the cross-section deformation. However, in this study, both of FS and FC are measured for the cross-section analysis.

Table 8.6 Cross-section deformation of EW and LW fibres

	120°C		160°C	
	Form Circle (FC)	Form Shape (FS)	Form Circle (FC)	Form Shape (FS)
EW	0.58	0.42	0.55	0.40
LW	0.74	0.68	0.71	0.62
MIX	0.73	0.65	0.70	0.60

Table 8.6 shows that the EW fibres had lower FC and FS values than LW fibres, which means that the former has higher collapsibility. Under the compression forces in refining, the thin-wall EW fibres are readily deformed and collapsed. While thick-walled LW fibres are rigid and resistant to the mechanical forces, thus they tend to retain their forms after refining. The SEM observations reveal that EW fibres are usually collapsed (Figure 8.49) while LW fibres are rarely noticeably deformed (Figure 8.50).

Table 8.6 also shows the influences of refining temperature on the cross-section deformations of fibres. Refining at 160°C yielded lower FC and FS values as compared to the refining at 120°C. This might be explained by the reduced plate gap at high temperature operation. As reported by Law [125], the compression force is mainly responsible for the fibre collapse in refining. The reduced refiner plate gap increases the compression action on the fibres, which produces more collapsed fibres and resulting in lower FC and FS values.

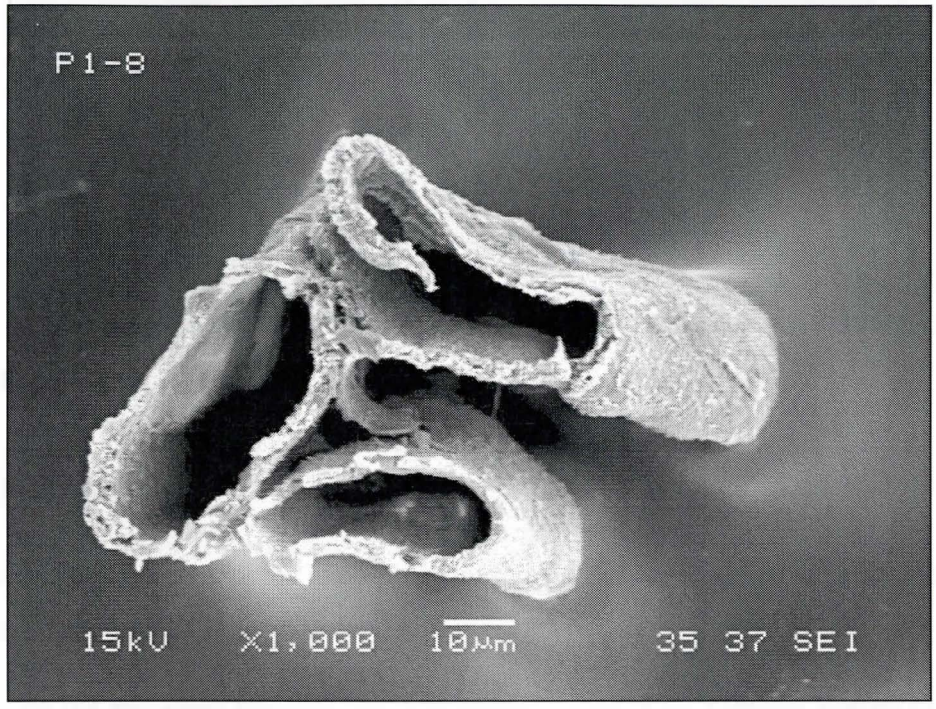

Figure 8.49 Collapsed EW fibres after refining
(R28 fraction of EW pulp produced at 120°C with 150 mL freeness)

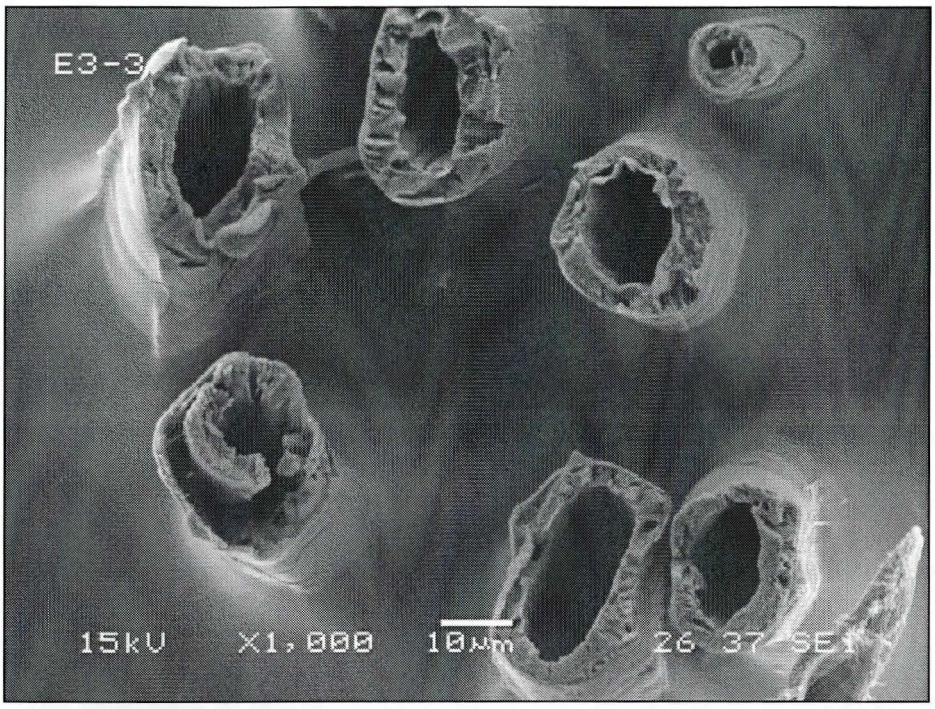

Figure 8.50 Uncollapsed LW fibres after refining
(R28 fraction of LW pulp produced at 120°C with 150 mL freeness)

8.7 Summary

The followings summarize the main points of this part of the study on the characteristics of pulp fibres produced in pressurized refining:

1. To get the same freeness, the EW pulps require more refining energy than the LW pulps;

2. In TMP thick-walled LW fibres exhibit mostly intra-wall failures and considerable external fibrillation while thin-walled EW counterpart tend to fail in trans-wall mode and show little external fibrillation. As a result, the former yield higher fibre length when compared with the latter;

3. In pressurized refining, LW yields more rejects than EW does. LW rejects have more fibrillar elements when compared with EW rejects, the latter contain more bundle-like shives;

4. Thick-walled LW fibres show lower curl and kink indices in comparison with thin-walled EW counterparts;

5. LW fines, which contain more fibrillar component than EW fines, give higher specific volume by sedimentation (SV);

6. EW pulps have higher water retention value (WRV) than LW pulps;

7. Refining at 160°C, which operates with reduced plate gap, produces more rejects and generates more fibrillation from LW than from EW when compared with refining at 120 °C;

8. EW fines-free pulps or fines has higher surface lignin coverage than those from LW;

9. The separation of thin-walled EW fibres take place mainly in the P/S1 interface while that of LW fibres occur commonly in the P/S1, S1/S2 regions;

10. In TMP, EW fibres show higher collapsibility and have lower FC and FS values than LW fibres.

Chapter 9 - Results and discussions (3) — handsheet characterization

Due to their morphological discrepancies, the properties of EW and LW fibres are developed differently in thermomechanical refining. These properties include fines formation, external and internal fibrillation of cell wall, and collapsibility of fibre, etc. Doubtlessly, the differences in these characteristics between EW and LW fibres would have dissimilar impacts on handsheet properties.

In this chapter, we firstly examine the physical and optical properties of handsheets prepared with EW and LW pulps, and the mixed furnish. Secondly, we discuss the characteristics of paper made with individual Bauer-McNett fraction or in combinations of 2-3 fractions, relative to EW and LW pulps. Thirdly, we also compare the properties of handsheets made from mixed furnish which was produced by co-refining both EW and LW chips with those prepared from EW and LW pulps produced in separated refining. With the information we hope to establish some possible interrelations between the fibre characteristics and paper properties.

9.1 Handsheet properties of whole pulp

The physical and optical properties of handsheets including density, roughness, porosity, tensile index, burst index, tear index, internal strength (Scott bond), brightness, opacity, light scattering and light absorption coefficient are discussed in detail in this part.

9.1.1 Physical properties

9.1.1.1 Density

Due to the superior collapsibility and conformability of EW fibres as compared to LW counterparts, the handsheets prepared from EW pulps were denser than those made of LW pulp, as Figure 9.1 shows. The refining temperature influenced also the handsheet density, which is particularly apparent when the sheet density is plotted against freeness. We learn that refining at high temperature (160°C) with reduced plate gap favoured

peeling of the outer-layer of cell wall and fibrillation of the exposed fibre surface, reducing cell wall thickness or coarseness of fibres. These changes brought about denser sheets. On the other hand, in the low temperature refining at 120°C (with a larger refiner plate clearance) the peeling and fibrillation were less evident, resulting in handsheets with higher bulk or lower sheet density.

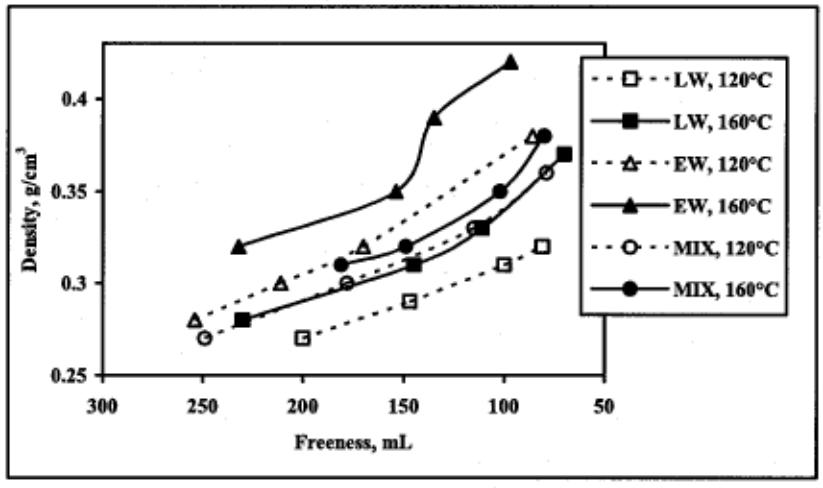

Figure 9.1 Handsheet density as a function of freeness

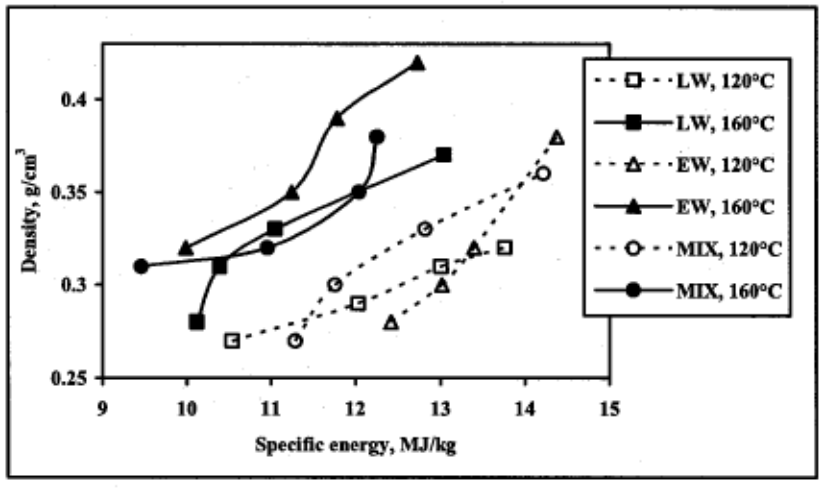

Figure 9.2 Handsheet density as a function of specific refining energy

When the sheet density is plotted against specific refining energy (Figure 9.2), the influence of high temperature refining on sheet density is still evident when compared with that of the low temperature refining, 120 °C. However, the trends of the curves are not as clear as those shown in Figure 9.1 where the sheet density is expressed as a

function of freeness. There are deviations of some curves, as shown in Figure 9.2. That will be the fact that the pulp freeness is not linearly with the refining specific energy, as indicated in the Figure 8.1.

9.1.1.2 Roughness

Surface smoothness is an important property for printing paper. Jack pine is known to produce sheet with high roughness due to its thick-walled fibres. For the same reason, thick-walled LW fibres produce handsheets with high roughness as compared to those of thin-walled EW fibres (Figure 9.3). Since the mixed furnish contains both LW and EW fibres, it makes handsheets with roughness inferior to that of LW sheet but higher than that for EW sheet. As mentioned earlier, the high temperature refining 160°C produced fibres with increased fibre flexibility and decreased fibre coarseness, and, as a result, made handsheets with increased density. These effects improve the surface quality of handsheets by reducing the surface roughness (Figures 9.3 and 9.4).

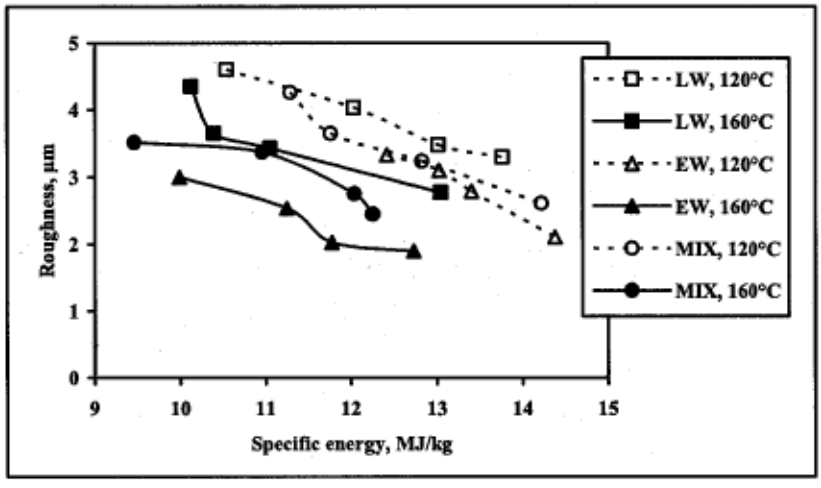

Figure 9.3 Roughness as a function of specific refining energy

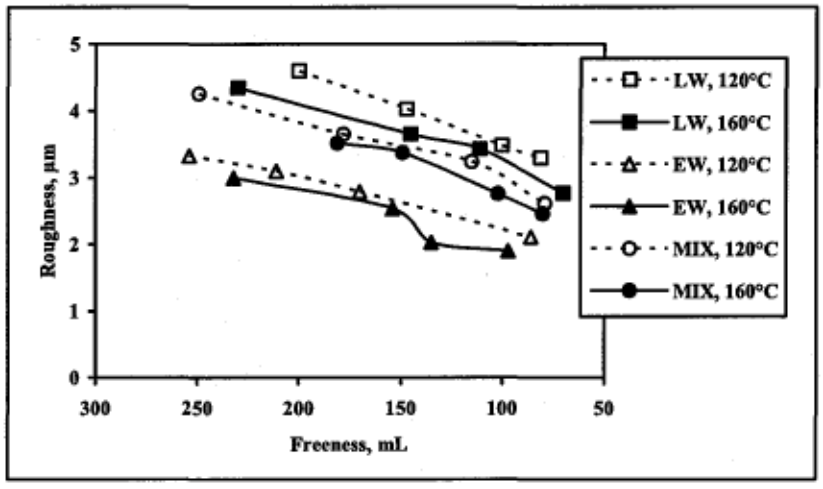

Figure 9.4 Roughness as a function of freeness

9.1.1.3 Porosity

The porosity of handsheets is mainly influenced by collapsibility of individual fibres within the sheet structure or the sheet density. The fines content in the pulp also plays an important role in determining the handsheet porosity. As indicated in Figure 9.5, the LW sheet has higher porosity than that of EW, which is due to the higher collapsibility and conformability of the EW fibres. As expected, the high temperature refining which was operated with reduced plate gap also reduced the porosity because more fines were generated at 160°C than at 120°C.

Similar tendencies can be observed when the sheet porosity is expressed as a function of freeness, as shown in Figure 9.6: LW produces higher porosity sheet than those of EW and the mixed furnish. However, the effect of higher temperature refining was inverted as compared with that shown in Figure 9.5. This difference might be attributed to the fact that high temperature refining requires less energy for a given freeness and consequently fewer fines were generated, which brings about higher porosity (Table 9.1). In high temperature refining (160°C) with reduced refiner plate gap more fibre cuttings and fibrillations occurred when compared with the lower temperature refining (120°C). In addition, the softening of lignin renders the fibres more easily separated in high temperature refining (160°C). As a result, for a given freeness, the high temperature operation consumed less energy than the low temperature refining.

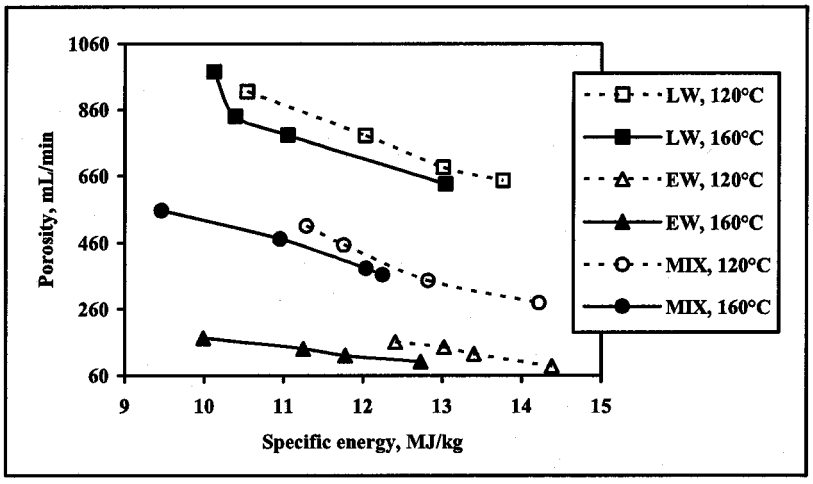

Figure 9.5 Porosity as a function of specific energy

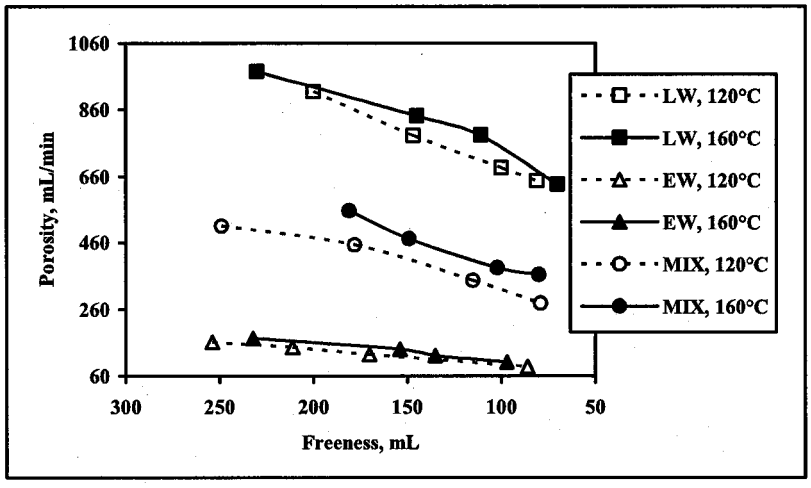

Figure 9.6 Porosity as a function of Freeness

Table 9.1 indicated that, for a given freeness, EW chips required more refining energy, relative to LW and the mixed furnish. During the refining, the thin-walled fibres which are more flexible than thick-walled LW fibres absorb more readily the refining energy and collapse more easily when compared to LW fibres. As for LW fibres, due to their thick cell wall, they tend to be fibrillated, causing more rapid drop in freeness when compared with EW fibres. As a result, they consumed less energy than EW fibres to reach a given level of freeness. In contrast, thin-walled EW fibres which are more fragile tend to break up more easily and yield less fibrils, resulting in giving more fines at a given freeness than LW fibres.

Table 9.1 Effect of refining temperature on fines formation at 150 mL freeness

	120°C	160°C
Freeness, mL	150	150
*SEC(EW), MJ/kg	13.40	11.25
SEC(LW), MJ/kg	12.03	10.12
SEC(MIX), MJ/kg	11.76	10.96
Fines(EW), %	30.8	28.6
Fines(LW), %	28.2	27.8
Fines(MIX), %	29.5	28.3

*SEC: specific energy consumption

9.1.1.4 Internal bond strength (Scott bond)

The Scott bond test represents the internal inter-fibre bonding strength in the direction normal to the plan of the handsheets. As seen in Figure 9.7, the advantage of high temperature over the low temperature refining is also evident in terms of Scott bond for all types of pulps. As discussed in chapter 8, at a given specific energy, the refining at higher temperature produced more fines and more collapsed and fibrillated fibres, which are beneficial for fibre bonding.

On the other hand, the handsheets prepared from LW pulp had higher internal bonding strength than those made from EW, which is true for both refining temperatures. A recent research work indicated that the fibril-like fines of TMP are responsible for good fibre bonding; they improve fibre bonding in three ways [132]:

- Increased specific bond strength (like glue)
- Increased bonded area at fibre crossing (accumulation of fines on bond borders);
- Increased numbers of bonds between fibres (pulling fibres into close contact).

Consequently, since LW pulps have more fibrils-like fines than EW counterparts, they showed higher Scott bond than EW pulps (Figure 9.7).

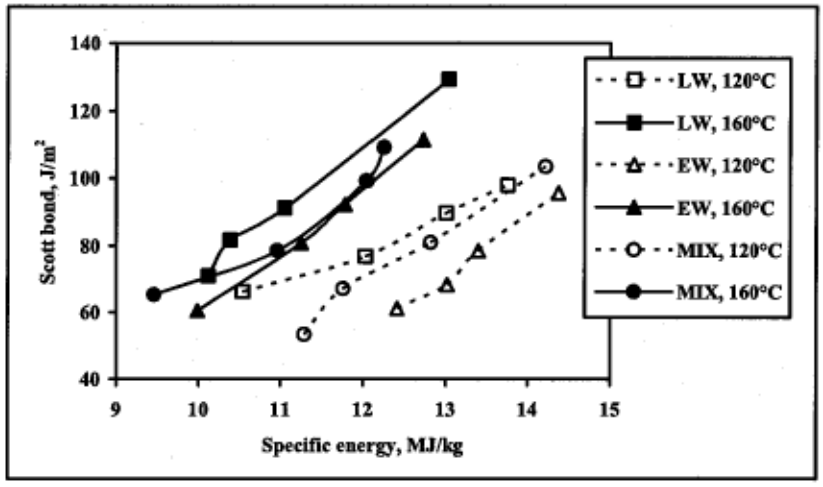

Figure 9.7 Internal bonding strength as a function of specific refining energy

The advantages of high temperature refining are evident only when the Scott bond of pulp is expressed as a function of refining energy; they are not apparent when the Scott bond is expressed in terms of freeness (Figure 9.8), which reveals the inadequacy of freeness for characterizing this refining effect.

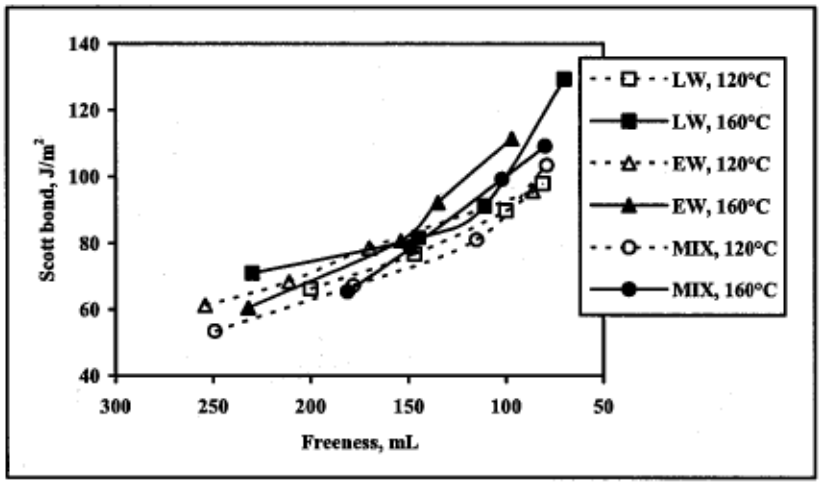

Figure 9.8 Internal bonding strength as a function of freeness

9.1.1.5 Tensile and burst indices

As a consequence of the increase in specific volume of fines (Figure 8.38) and sheet density (Figure 9.2) in high temperature refining at 160°C, the tensile (Figure 9.9) and burst (Figure 9.10) indices are significantly improved in comparison with the low

temperature refining. The increase in fines content (Figure 8.8) is also beneficial for bonding strength improvement. Similar tendency can also be seen in the Scott bond test.

For a given refining energy, the handsheets made from LW fibres exhibited a higher tensile and burst strength when compared to those prepared from EW and mixed furnish. This characteristic is evident for both refining temperatures. As discussed in 9.1.1.4, at a given refining energy, LW produce more fibril-like fines in the pulps, which favours the fibre bonding in the fibre network (paper). This might be the reason why LW pulps had better tensile and burst indices than EW as well as the mixed pulps, at a given refining energy.

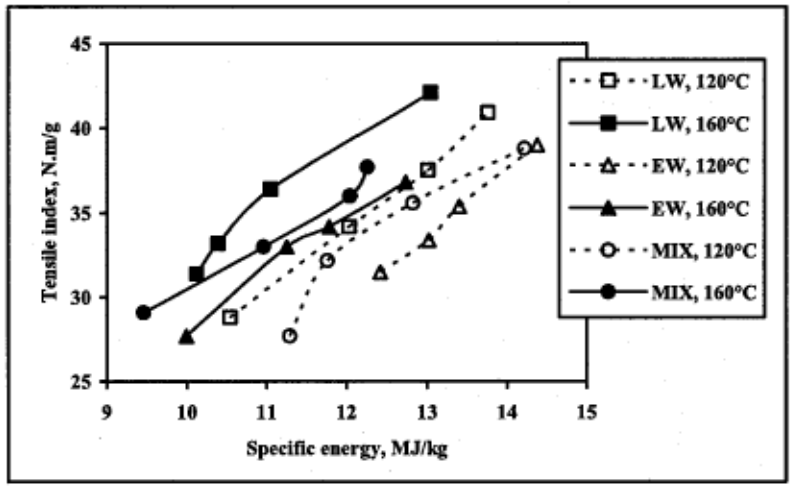

Figure 9.9 Tensile index as a function of specific energy

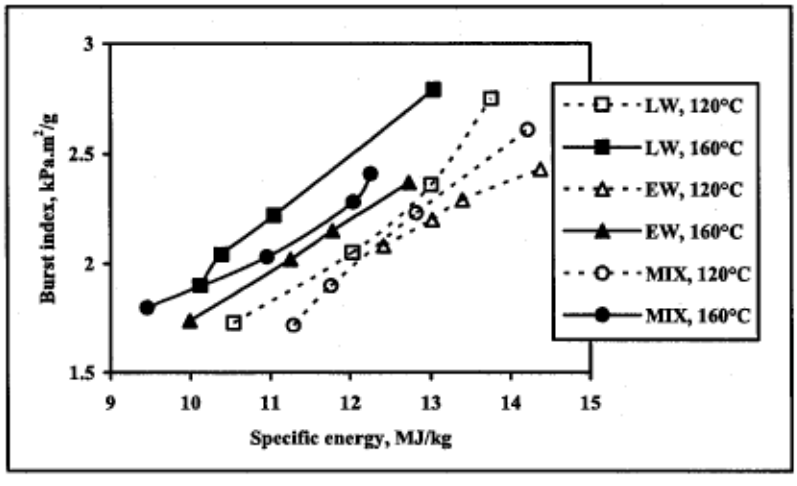

Figure 9.10 Burst index as a function of specific energy

However, when the tensile index (Figure 9.11) and burst index (Figure 9.12) are plotted against freeness, the influences of high temperature refining disappeared, implying the deficiency of freeness as a parameter for characterizing the refining properties. As discussed earlier, the high temperature refining requires less energy than the low temperature refining (120°C) to reach a given degree of freeness. The particularly good tensile index and burst index of EW pulps produced at 120°C is due to the higher energy consumption of EW fibres (Table 9.1).

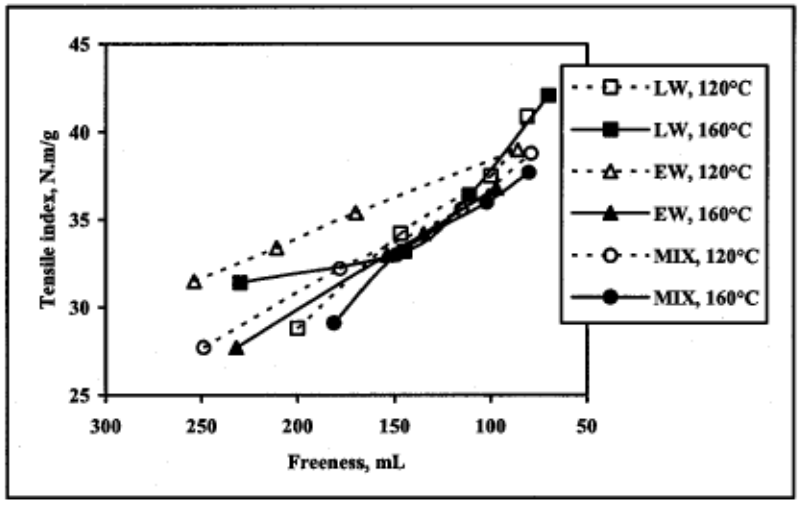

Figure 9.11 Tensile index as a function of freeness

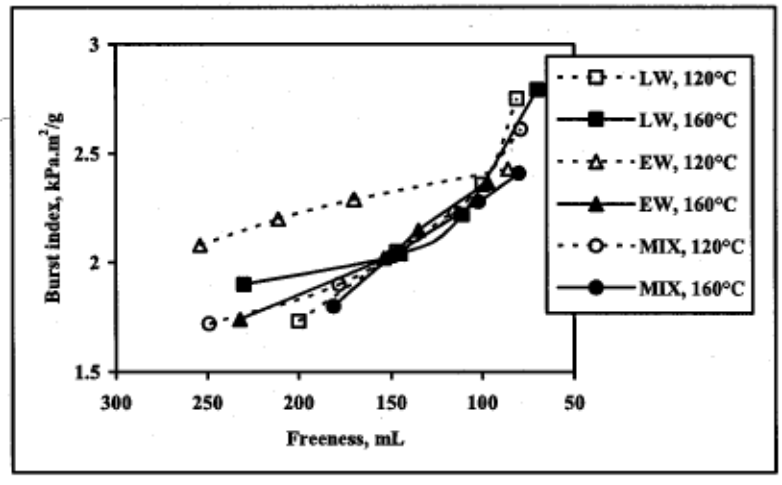

Figure 9.12 Burst index as a function of freeness

9.1.1.6 Tear index

Albeit the high temperature refining had more fibre cutting when compared to the low temperature operation it did not, however, adversely affect the tear index of handsheets when the refining energy increased up to 12 MJ/kg (for LW at 120°C) where the freeness is approximately 100 mL (Figures 9.13 and 9.14). This phenomenon is not unexpected because tearing resistance depends not only on fibre length but also on fibre bonding [181]. The negative effect of fibre length reduction could be well compensated by the increasing in fibre bonding. The fall in tear index due to high temperature refining is particularly notable for EW fibres which suffered more severe shortening in comparison to those of LW and mixed furnish during refining (Figure 8.17).

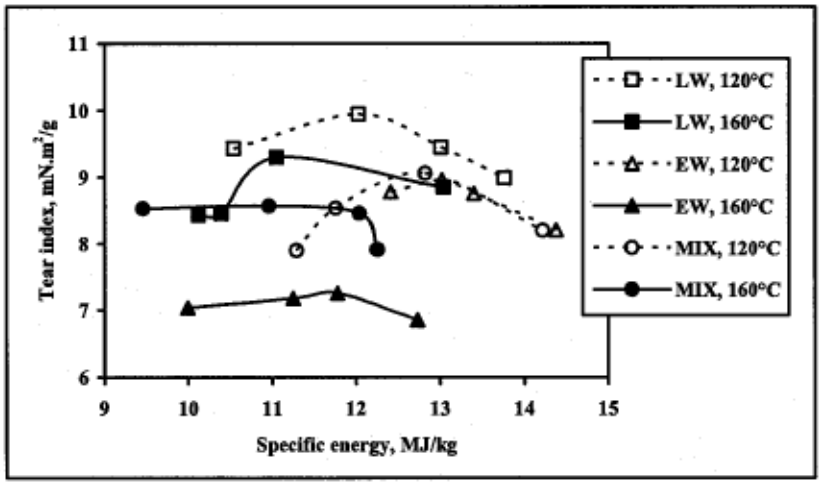

Figure 9.13 Tear index as a function of specific energy

The influence of high temperature refining is also noticeable in Figure 9.14 where the tear index is plotted as a function of freeness. Note that LW pulp always yields greater tear strength when compared to EW component since the former has longer fibres than the latter.

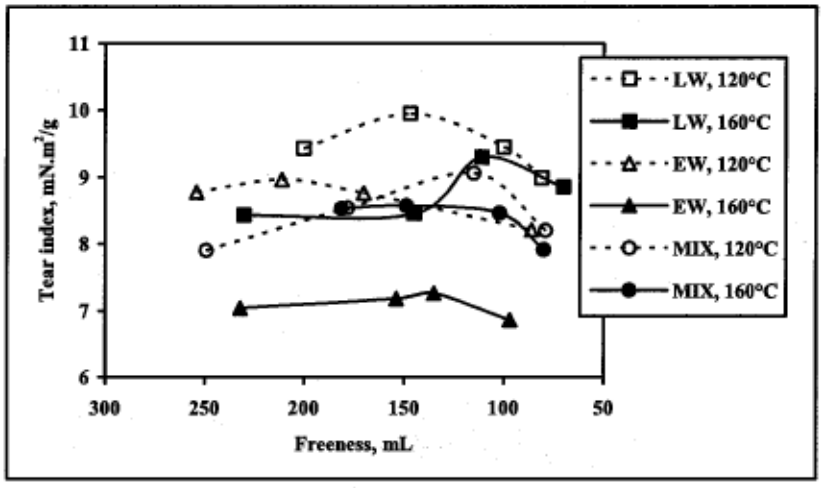

Figure 9.14 Tear index as a function of freeness

9.1.2 Optical properties

9.1.2.1 Brightness

The most substantial detrimental effect of high temperature refining was, perhaps, the remarkable darkening effect, resulting in a significant drop in brightness (Figure 9.15). But recent research [182, 183] indicated that the new RTS (Low Retention, High Temperature and High Speed) TMP pulping method developed by Andritz could produce pulp with less specific energy, improved strength properties and equivalent brightness compared to conventional TMP pulp. Due the equipment and operation restrictions, we couldn't realise this technique at our research centre. Thus the high temperature refining darkened the pulp. The decrease in initial pulp brightness would mean higher bleaching cost. On the other hand, EW always produces brighter pulp than LW, and the mixed furnish lies in between them. This is not surprising since the initial brightness of EW chips (Figure 7.1) is higher than those of LW, which results in higher pulp brightness.

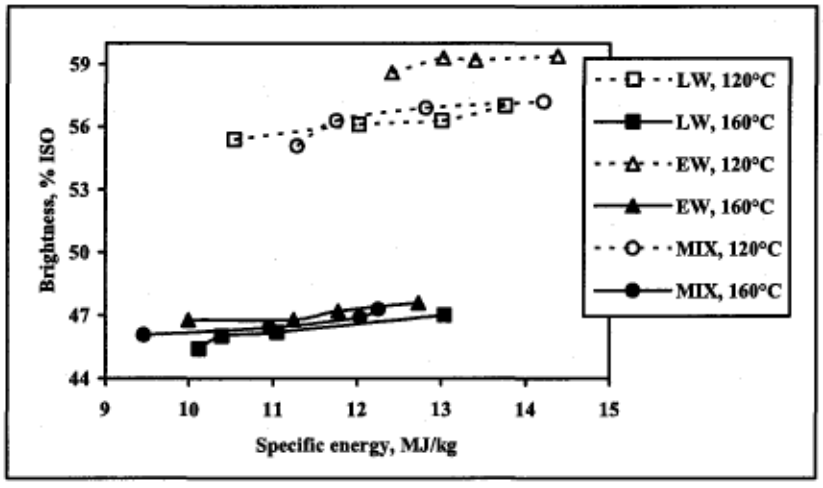

Figure 9.15 Brightness as a function of specific energy

Similar tendencies can also be seen in the brightness which is plotted either as a function of specific energy (Figure 9.15) or against freeness Figure 9.16.

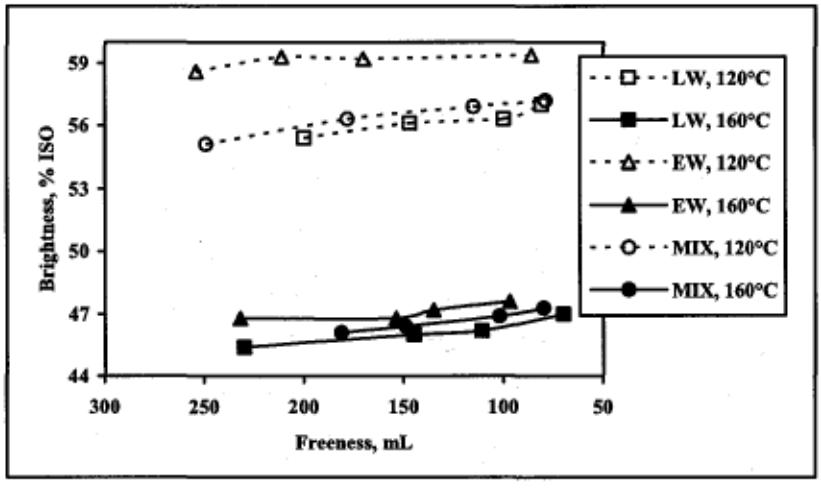

Figure 9.16 Brightness as a function of freeness

9.1.2.2 Light scattering coefficient

The light scattering coefficient of paper prepared from mechanical pulp is mostly influenced by the fines contents: higher fines contents yield greater light scattering coefficient [184]. Raising the refining temperature from 120 to 160°C also increases the light scattering coefficient (Figure 9.17) because of the increase in fines content at the high temperature refining which is operated at reduced plate gap.

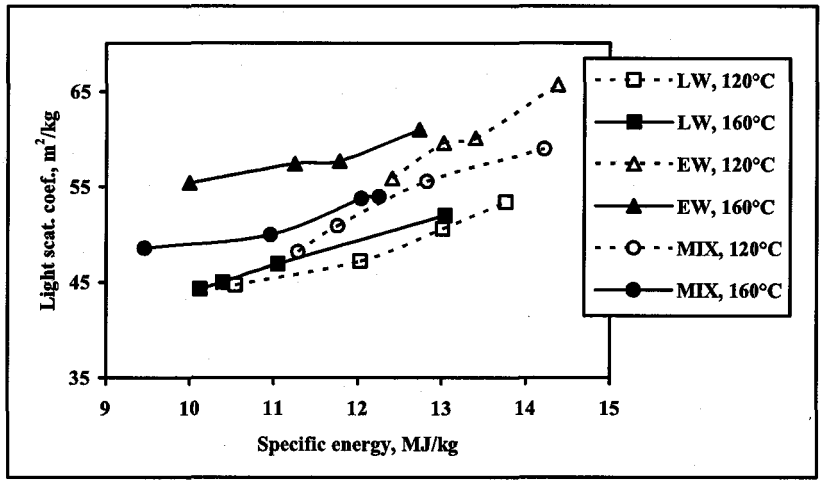

Figure 9.17 Light scattering coefficient as a function of specific refining energy

EW pulps always has higher light scattering coefficient than those of LW because they contained less fibrils in their fines, as discussed earlier. A recent research work indicated that the fibril-like fines have greater specific surfaces than those of flake-like components. The fibril components contribute to the increase in physical properties of paper, as indicated in Table 9.2 [132, 185, 186].

Table 9.2 Contribution of fibril and flake fines to paper properties

Attribution	Physical strength	Light scattering coefficient
Fibril-like fines	++++*	-**
Flake-like fines	+	++++

*: Positive effect
**: Negative effect

The fibril-like elements in the fines influence positively the physical strength of sheet (e.g. tensile and burst indices) and negatively the light scattering coefficient of paper. The flake-like fines particles, namely, ray cells and tiny pieces detached from the outer layers of cell wall may play a less important role in determining the paper strength, but have considerable influence on the optical properties of paper. Due to their extremely high conformability, the fibril-like fines could be bonded tightly to the fibre surface, having little contribution on the light scattering coefficient [132]. Therefore, the fibril-like fines have negative effect on the light scattering coefficient in contrast to the flake-

like fines. Since EW fibres produce more flake-like fines in the pulps, they give higher light scattering coefficient in relation to the LW pulps.

Note that when the light scattering coefficient is plotted as a function of freeness (Figure 9.18), the high temperature refining yields inferior light scattering coefficient. This is attributed to the fact that high temperature refining required much less energy to reach a given freeness and, consequently, fewer fines are generated, as indicated that in Table 9.1.

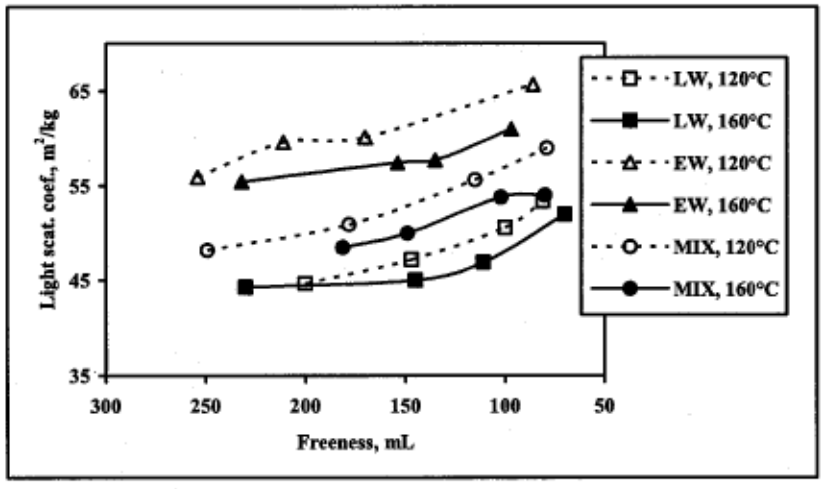

Figure 9.18 Light scattering coefficient as a function of freeness

9.1.2.3 Light absorption coefficient

The light absorption coefficient of paper is mainly influenced by the chemical nature of the wood components, such as cellulose, hemicelluloses, lignin and extractives. Cellulose and hemicelluloses are practically uncoloured. While the lignin is coloured since its chromophores absorb light and make the lignin coloured. The contribution of the chemical components of wood to the light absorption coefficient can be expressed by Equation 9.1 [35].

$$k_p = c_C \cdot k_C + c_L \cdot k_L + c_E \cdot k_E \qquad \text{Eq. 9.1}$$

where,

k_p = light absorption coefficient of the pulp;

c_C, c_L *and* c_E = relative amounts of carbohydrates, lignin, and extractives, respectively in the pulp;

k_C, k_L *and* k_E = light absorption coefficients of carbohydrates, lignin, and extractives.

Based on this equation, the contribution of the various chemical components on the light absorption coefficient can be estimated as shown in Table 9.3. The example shows that more than 90% of the coloured matter in Norway spruce (*Picea abies*) originates from the lignin. The light absorption coefficient of the extractives is also relatively high, but the contribution to the light absorption coefficient of the pulp is limited because of the low content of extractives in wood.

Table 9.3 Contribution of the various components of Norway spruce (*Picea abies*) to the light absorption coefficient [187]

	Relative amount, c_x	Light absorption coefficient, k_x, m^2/kg	Contribution to the light absorption coefficient of the pulp, $c_x \cdot k_x$, m^2/kg
Carbohydrates	0.70	0.35	0.25
Lignin	0.28	20	5.6
Extractives	0.02	7.5	0.15
Whole pulp	1.0		6.0

As discussed in Table 7.2, the Jack pine EW contains more lignin (28.30%) than the LW (27.09%). This might be the reason that EW sheet has higher light absorption coefficient than that of LW (Figure 9.19). Note that: the high temperature refining (160°C) produced pulps having higher light absorption coefficient than the low temperature refining (120°C). This is because of the darkening of high temperature refining, that is more chromophores are produced at higher temperature since they are sensitive to the heat. As a result, their light absorption coefficient increased with increasing temperature.

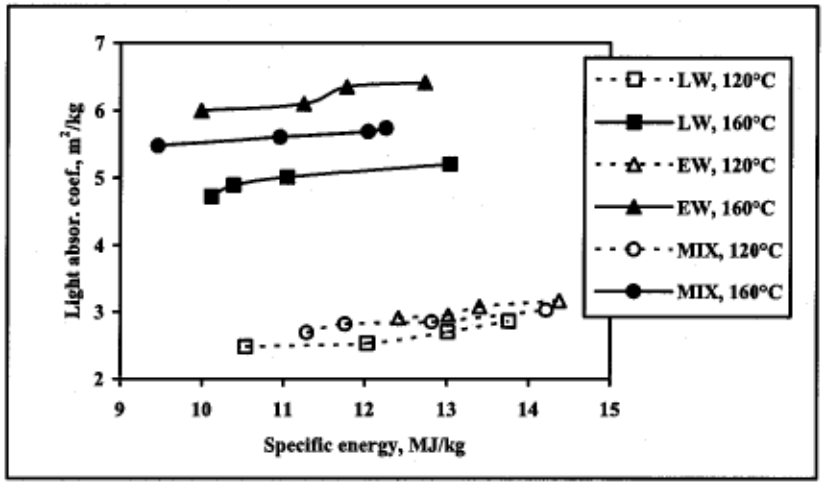

Figure 9.19 Light absorption coefficient as a function of specific refining energy

Similar tendencies can be seen in the Figure 9.20 where the light absorption coefficient is plotted against the freeness.

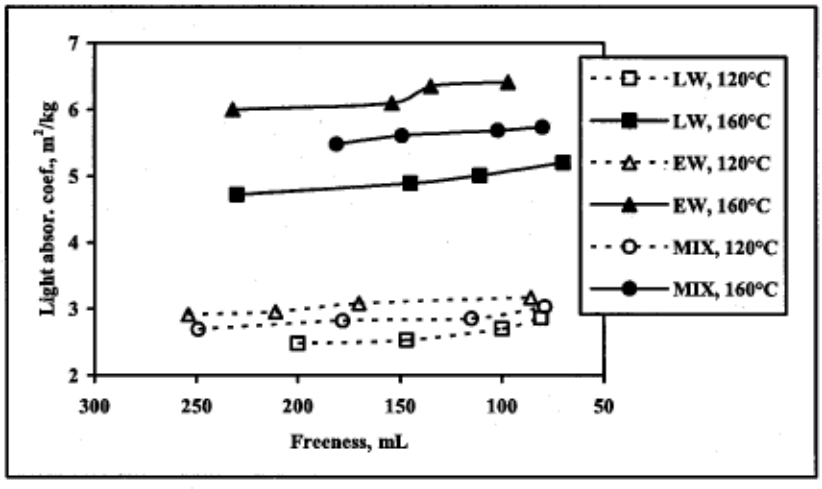

Figure 9.20 Light absorption coefficient as a function of freeness

9.1.2.4 Opacity

As a result of the decreased brightness in high temperature refining, the sheet opacity increased considerably (Figure 9.21). Another reason might be the increased light scattering and light absorption coefficient in the high temperature refining, which also increase the sheet opacity. It also owns this to its lower content of fibrils in the fines of

EW pulps, meaning a higher light scattering coefficient. Similar trends can also be seen in the Figure 9.22, the opacity is plotted against freeness (Figure 9.21).

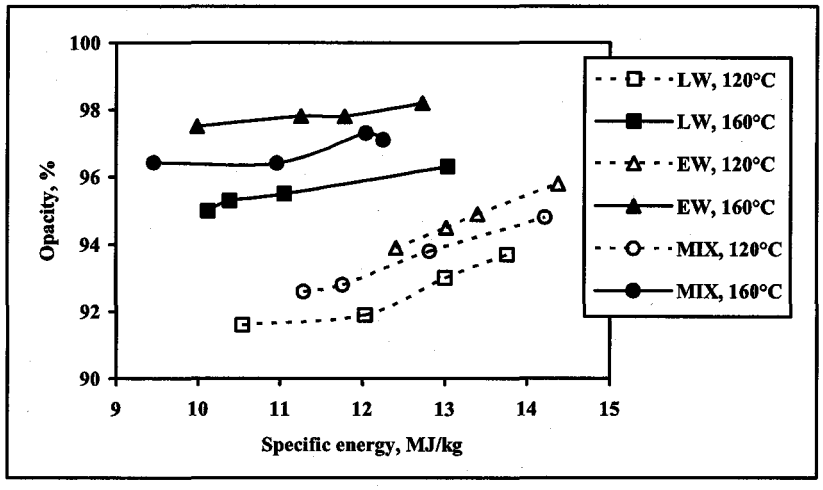

Figure 9.21 Opacity as a function of specific refining energy

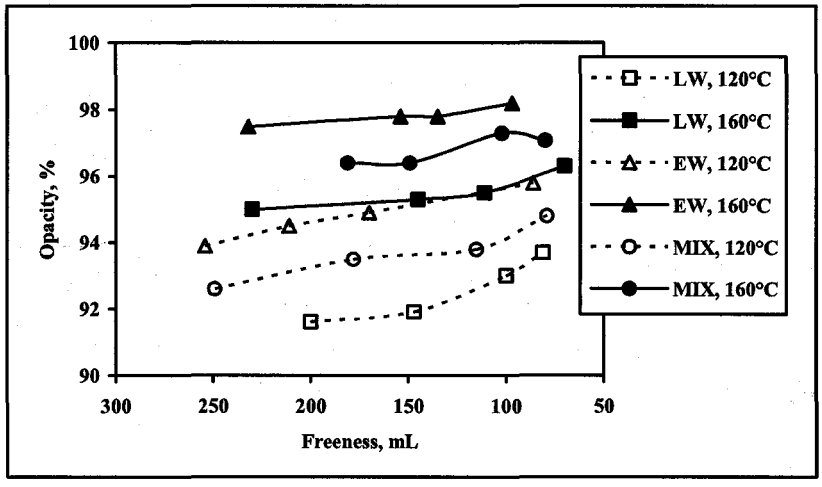

Figure 9.22 Opacity as a function of freeness

9.2 Characteristics of fibre fraction

Four Bauer-McNett fractions of EW and LW pulps, namely R14, R28, R48 and R100, are employed to make handsheets and studied their contribution on the sheet properties. EW and LW pulps having a freeness of 150 mL and produced at 160°C are used in this study.

9.2.1 Freeness

The measurement of pulp freeness could indicate the level of fibre flexibility and fibrillation of fibres in a particular fraction when the fines are removed from the pulp. Table 9.4 shows that the R14, R28 and R48 have particularly high freeness of about 700-650 mL, while the R100 had a slightly lower freeness of 450 mL. Interestingly, in spite of their reduced coarseness (Figure 9.23), the R28 and R48 still maintain a very high freeness, indicating that their cell wall is not sufficiently developed. On the other hand, the R100 being partially split and fibrillated with ribbon-like elements (Figure 8.13) showed a noticeable drop in freeness in comparison with the other three longer fractions.

Table 9.4 Freeness (mL) of pulp fractions

Sample	R14	R28	R48	R100
LW	700	690	670	450
EW	690	680	655	440

EW fractions including R14, R28, R48 and R100, always had a slightly lower freeness than those of LW. This might be accounted for by their higher water retention value and more significant internal fibrillation in comparison with the LW fibres.

9.2.2 Coarseness

As showed in Figure 9.23, the fibre coarseness decreases gradually from the R28 to R100 fractions or in order of fibre shortness, indicating the cell wall reduction progressed as the fibre length decreased. The figure also reveals that LW yields coarser fibres than that of EW, which is expected since LW fibres have much greater initial cell wall thickness than EW counterpart.

It should be noted that we did not measure the coarseness for R14 fraction because of a technical difficulty of the FQA which tends to be clogged with long fibres such as those in the R14 fraction. However, according the microscopic study and the curve tendency in Figure 9.23, we can still assume that the R14 fibres have highest coarseness among the four fibre fractions.

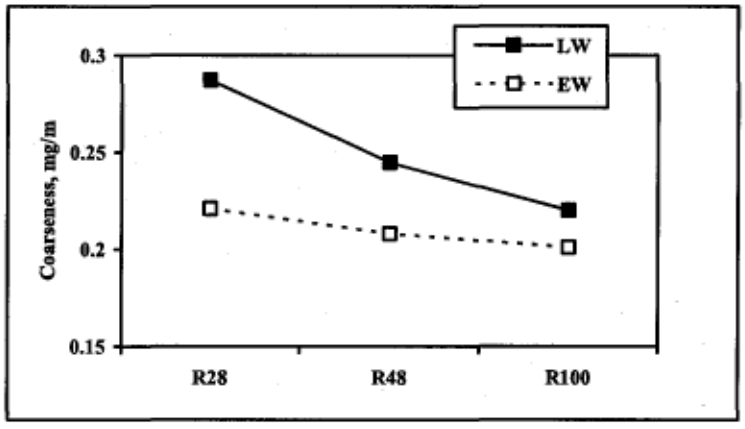

Figure 9.23 Coarseness of Bauer-McNett fractions

9.2.3 Sheet density

As showed in Figure 9.24, sheet density increases gradually from R14 to R100 fraction, indicating that the short fibres are developed to a greater degree than the long fibres. Apparently, the cell wall of the R100 fibre has to be substantially fibrillated to yield noticeable improvement in sheet density in comparison with the other three fractions.

Note that EW fibres produce higher sheet density than those of LW, which is apparently due to the fact that the former has much thinner cell wall than the latter. Another reason might be the fact that EW has a greater length of fibre in the sheet per unit weight so bond crossings are closer together. As a consequence, thin-walled EW fibres are more flexible and more conformable than thick-walled LW in the sheet consolidation.

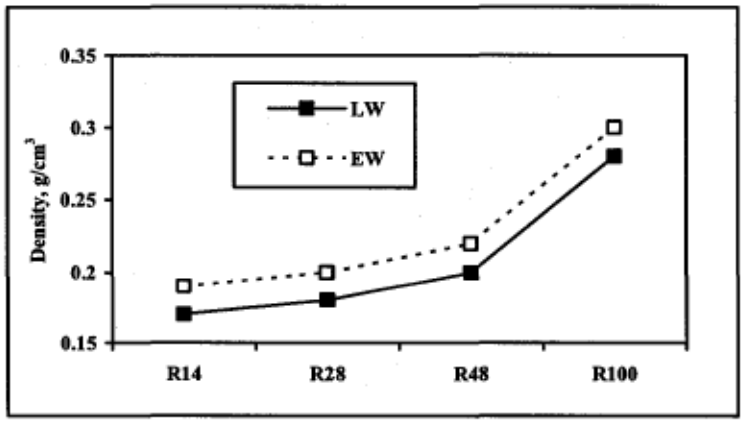

Figure 9.24 Sheet density of Bauer-McNett fractions

9.2.4 Physical properties

As discussed earlier, the fibre coarseness decreased in order of the length fraction, which is from R14 to R100 fraction, indicating that the shorter fibres were better developed than the longer ones. As a consequence, the strength properties such as Scott bond, tensile index and burst index tend to follow the same trends but with increasing magnitude (Figures 9.25, 9.26, and 9.27). The overall improvements in strength properties occurred in both EW and LW pulps.

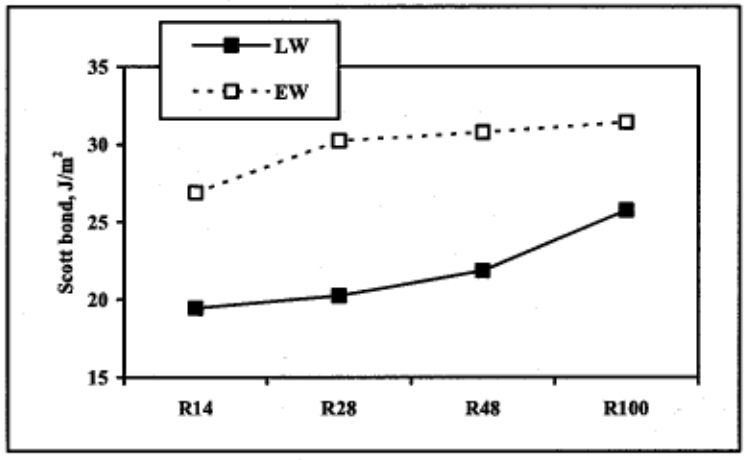

Figure 9.25 Scott bond of Bauer-McNett fractions

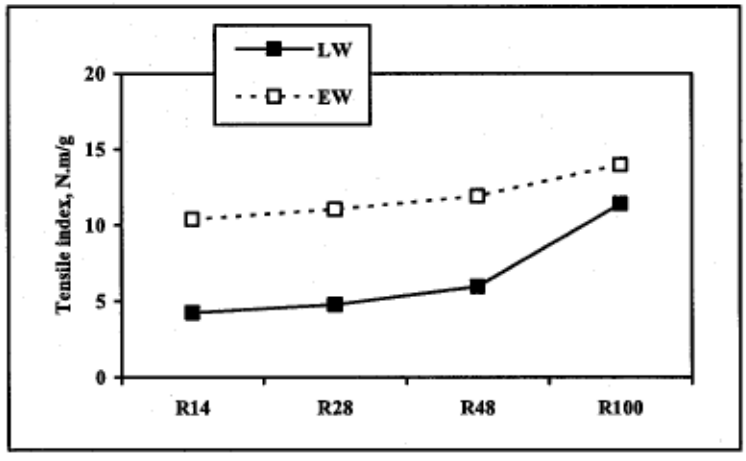

Figure 9.26 Tensile index of Bauer-McNett fractions

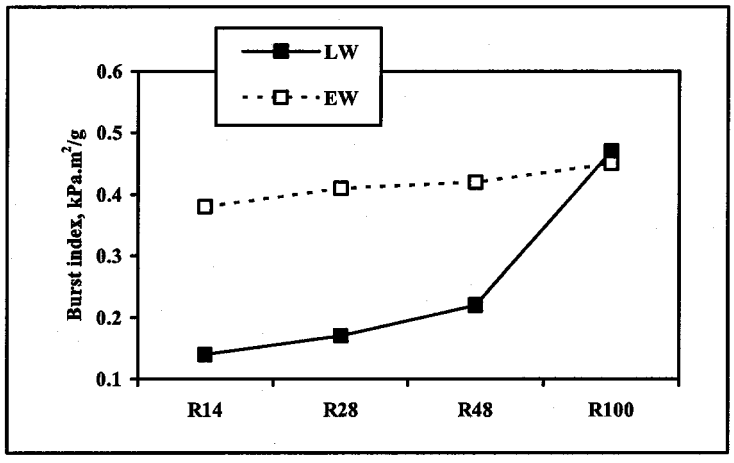

Figure 9.27 Burst index of Bauer-McNett fractions

In comparison with LW pulps, EW pulps always had higher values in Scott bond, tensile index, and burst index. The difference is especially apparent for the longer fibre fractions, i.e. R14, R28 and R48. However, this difference in strength properties diminished when the comparison was made with the R100 fraction. The fact that the EW pulps always had higher mechanical resistances in the sheets made from the relatively long fractions (R14, R28 and R48) might be accounted for the greater collapsibility and conformability of EW fibres. However, as discussed earlier, the thick-walled LW fibres have higher cell-wall thickness reductions and yield more fibrillated fibres during refining in comparison with EW fibres. Therefore, it is believed the short fibres of R100 of LW pulps had been well developed and yielded good strength properties which are comparable to those of the R100 fraction of EW. According to Heikkurinen [188], fibre fibrillation and collapsibility are both helpful for the fibre bonding and sheet formation. Based on this finding, the R100 fraction of both EW and LW fibres could have similar strength characteristics.

On the other hand, the tear index, which depends greatly on the average fibre length decreased gradually in descending order of magnitude of fibre length of each fraction size (Figure 9.28). Note that for a given fibre fraction (excepting the R100) EW fibres always had greater tear index than the LW ones. Although the tearing resistance of paper depends principally on fibre length, but it was found that inter-fibre bonding can also play an important role in determining the tearing strength [181]. However, as in the case

of Scott bond, tensile and burst indices, the difference in tear index between the EW and LW disappeared in the R100 fraction. This phenomenon indicates the important contribution of fibre bonding to this property. Mohlin reported similar findings [189, 190].

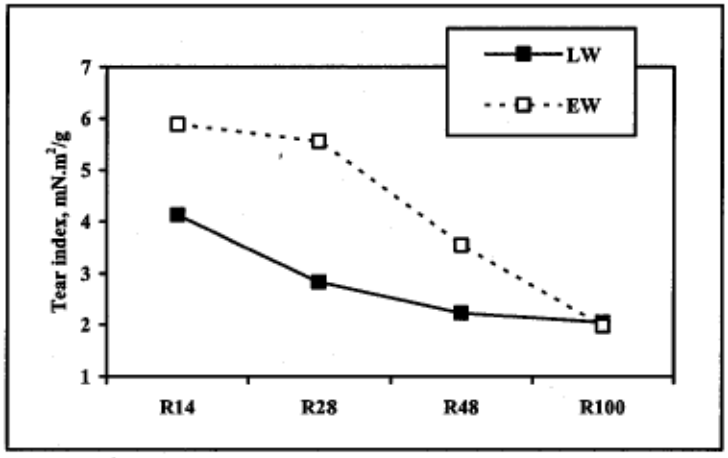

Figure 9.28 Tear index of Bauer-McNett fractions

9.3 Influence of short fibres and fines

The combination of the three longest fractions, that is R14+R28+R48, is referred to as the long fibre fraction while that of three shorter fraction, i.e. R100+R200+P200 as the short fibre fraction. In this study, the characteristics of handsheets made from short fibre and long fractions were compared with those of whole pulp (WP) as well as the whole pulp without fines (WP-P200). The EW and LW pulps produced at 160°C which has a similar freeness of about 150 mL are used in this study. The purpose here is to attempt to know the impact of the short fibres and fines on EW and LW sheet properties when these fibres are progressively removed from the whole pulp.

9.3.1 Freeness

As shown in Table 9.5, removing the fines (P200) from the whole pulp increases the freeness by 400% for both EW and LW pulps, indicating an extremely close correlation between freeness and fines in mechanical pulp. Further exclusion of the short fibre fractions (R100+R200+P200) from the whole pulp increased further the freeness, up to about 700 mL, an additional rise by 100%. This means that the long fibres were poorly

developed. The freeness of the short fibre fraction (R100+R200+P200) dropped by 400%, which indicates the contributions of these short fibres, especially the fines, to the pulp freeness.

Table 9.5 Freeness (mL) of pulp combinations.

Sample	WP*	WP-P200	R14+R28+R48	R100+R200+P200
LW	150	600	710	35
EW	150	590	700	25

*WP: whole pulp

Interestingly, if we exclude the whole pulps, in the comparison three other fibre combinations between LW and EW pulps, the former has slightly higher freeness than the latter. This might be LW fibres are stiffer and has less conformability than EW fibres. In addition, the higher water retention value of EW fibres might be another reason for their lower freeness, since the internal fibrillation is more evident in thin-walled EW fibres.

9.3.2 Sheet density

The elimination of fines (P200) from the whole pulp decreased the sheet density by about 30% while the removal of the short fraction (R100+R200+P200) caused an additional drop of only about 3% for both EW and LW pulps. This implies that the fibre fractions (R100+R200) had relatively less effect on sheet density. As for the short fibre combination, the sheet density was increased nearly by two folds when compared to the other combinations. This finding indicates the importance of the fines in sheet formation.

Short fibres of both EW and LW pulps produced handsheets having similar density. Comparatively, the other fractions of EW pulps showed slightly higher density than those of LW pulp, which might be attributed to their better fibre flexibility and conformability.

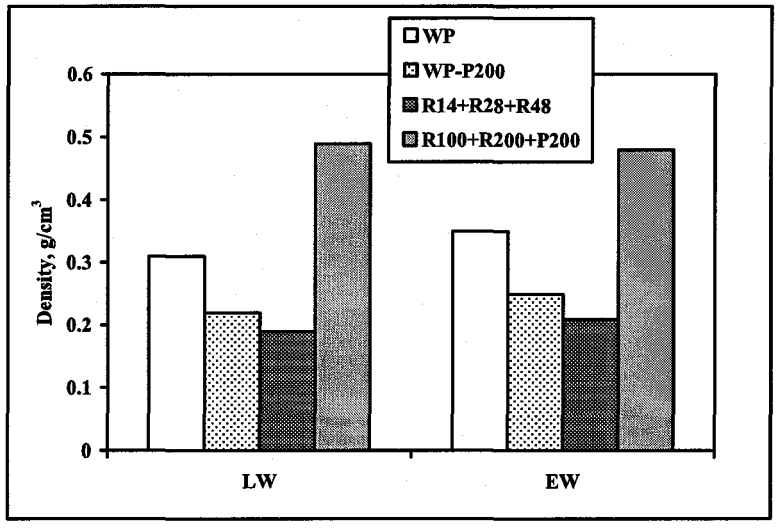

Figure 9.29 Density of handsheets made from different combinations of pulp fraction

9.3.3 Tensile index

Tensile index dropped significantly upon the removal of fines from the whole pulp, about 65% for LW and 50% for EW pulps (Figure 9.30). The difference in reduction in tensile index indicates the possible difference in characteristics or contributions of the fines in the two pulps (EW vs. LW). As seen earlier, LW fines have more fibrils than those of EW. The fibrils favour the fibre bonding in the handsheets [132].

Further exclusion of the R100+R200 fractions decreases the tensile strength, but less drastically. This could be due to the fact that these two fractions represent a relatively small proportion of the whole pulp, about 9 and 6% respectively (Figures 8.6 and 8.7). In addition, the R100 had a relatively high freeness of about 450 mL (Table 9.4), indicating that these fibres are poorly developed. As such, it would be a relatively less important role in affecting the tensile strength.

The tensile index for LW short fibre combination (R100+R200+P200) was nearly eight times that for the long fibre combination (R14+R28+R48). For EW pulp fibres the difference are about three times. Hence, the principal source of the fibre bonding in the whole pulp originates from the short fibres. On the other hand, LW short fibres give

higher tensile index than EW fibres, as shown in Figure 9.30, which might be due to the more abundant fibrils in LW fines, as discussed earlier.

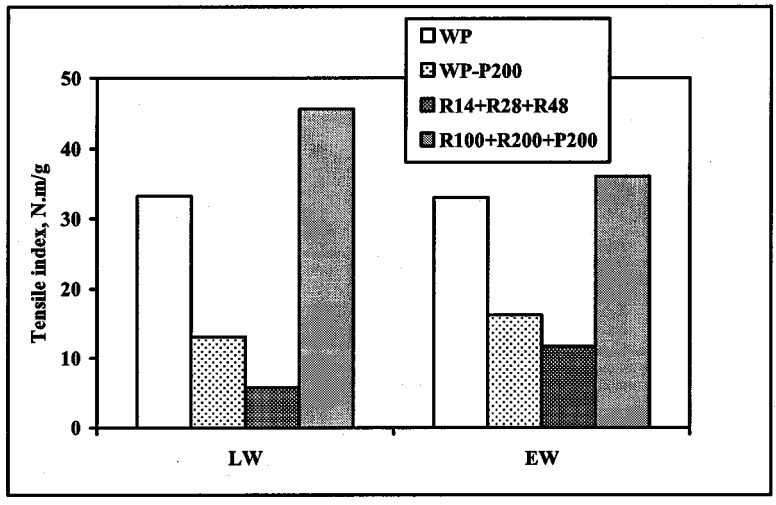

Figure 9.30 Tensile index of different pulp fraction combinations

9.3.4 Burst index

The burst resistance is closely associated with the tensile strength of handsheets. Very similar influences of various fibre fractions on the burst index of handsheets (Figure 9.31) was observed when compared to those in tensile index discussed previously (Figure 9.30). Eliminating the fines from the whole pulp reduces the burst index by about 250% for LW pulps and 170% for EW pulps. This indicates that LW fines had a better bonding potential than EW fines, since the former having more fibrillar elements than the latter. Further exclusion of the R100+R200 fractions reduced further the tensile strength, but to a less degree. LW short fibre combination (R100+R200+P200) had a tensile index which was nearly 8 times that of the long fibre combination (R14+R28+R48). For EW pulps it was only about 1.8 times. Again, this shows that LW fines have greater bonding capacity than those of EW pulp, the former containing more fibrils in the fines.

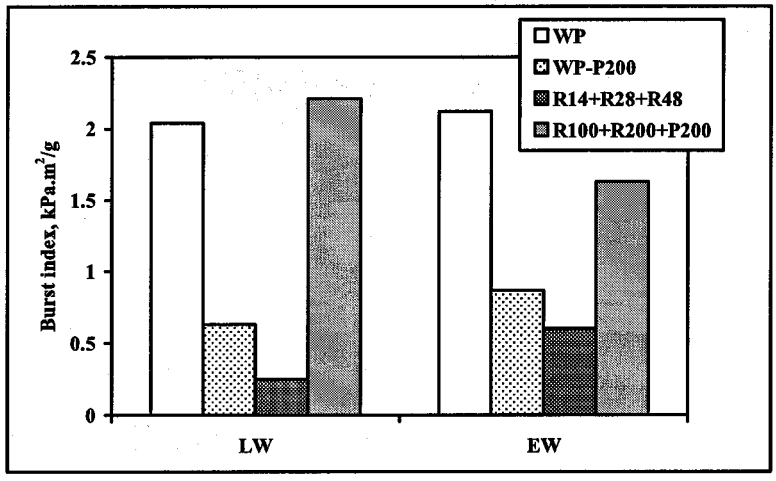

Figure 9.31 Burst index of different pulp fraction combinations

9.3.5 Tear index

Removing the short fibres including the fines from the whole pulp produced intriguing results in terms of tear strength (Figure 9.32). Firstly, the loss of fines, decreases the tear index by about 40% for LW pulp, and 25 % for EW pulp, substantiating the fact that the fines have an important role in influencing the tear resistance. This effect is more evident for LW pulps since they contain more reinforcement element, namely the fibrils, in the fines. Secondly, the long fibre combinations (R14+R28+R48) were only slightly superior in tear strength when compared with the short fibre combination (R100+R200+P200), which could be explained by the conventional thinking that the tear index increases with fibre length. The long fibre combination (R14+R28+R48) had, in fact, lower tear index compared with the whole pulp. This result underlines the fact that the increase in the fibre length alone does not necessarily improve the tearing resistance, implying an important role of fibre collapsibility and conformability. Note that even if LW fibres have higher fibrillations than EW fibres, EW short fibre combination still had higher tear strength than that of LW. Speculatively, this might be due to the fact that EW elements have better conformability and flexibility than LW counterpart, enhancing the bonding between the fibres.

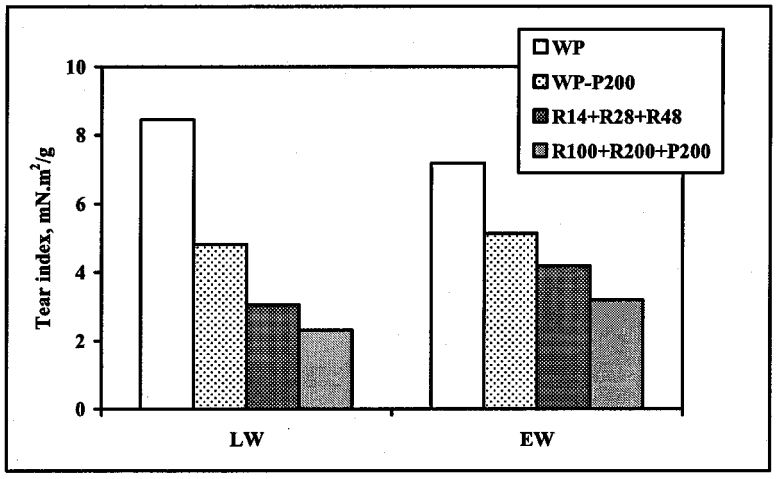

Figure 9.32 Tear index of different pulp fraction combinations

9.3.6 Porosity

As discussed earlier, the fines content had a great influence on the porosity of handsheets which can be clearly observed from the Figure 9.33: the magnitude of handsheet porosity of various fractions can be arranged in the following decreasing order:

(R14+R28+R48) > (WP-200) > WP > (R100+R200+P200)

According to this order, from the left to the right, the fibre length decreased gradually while the fines content increased progressively. This indicates the importance of the fines in affecting the sheet porosity. In fact, the fines fill the voids in the fibre network, reduce the sheet porosity. Note that the LW fines had a greater influence on the porosity than EW counterparts. This may be explained by the fact that LW fibres have thicker cell wall and are stiffer in comparison with thin-walled EW fibres, making the sheet structure more porous.

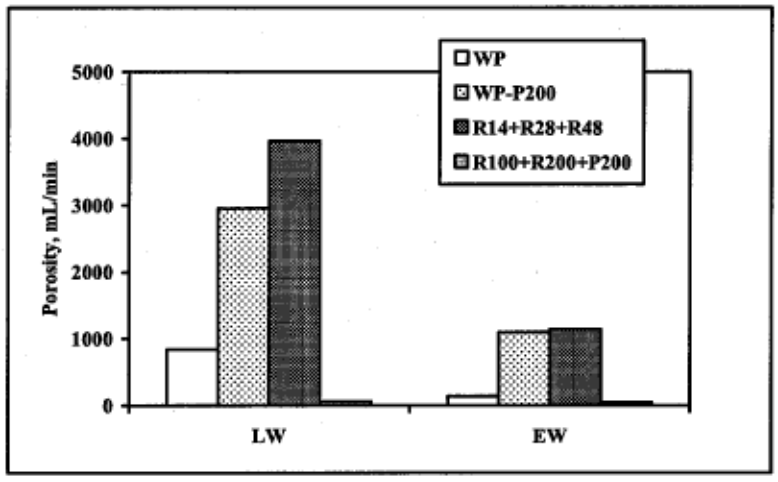

Figure 9.33 Porosity of different pulp fraction combinations

9.3.7 Roughness

The sheet roughness followed the same trends as the porosity, as shown in Figure 9.34. The roughness decreased progressively as following:

$$(R14+R28+R48) > (WP\text{-}200) > WP > (R100+R200+P200)$$

The magnitude order shows that the sheet roughness decreased as the fibre length of the fractions decreased. This observation suggests that the sheet surface furnish depends on the composition of various length fraction, increased proportion of short fibres and fines improves the surface quality. On the other hand, thin-walled EW fibres favour a smooth surface when compared to thick-walled LW fibres.

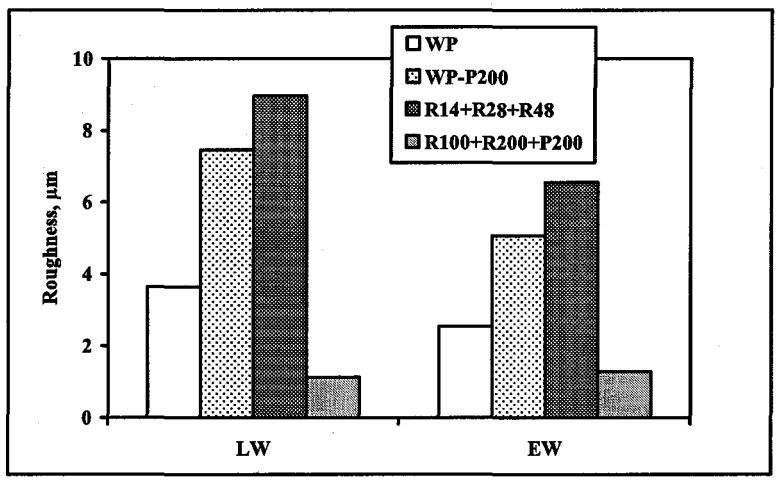

Figure 9.34 Roughness of different pulp fraction combinations

9.4 Influence of mixing processes

In this part of our study, we attempt to investigate the influence on pulp properties of co-refining of EW and LW chips and on the blending of EW and LW pulps which were separately refined. In the case of post-refining blending we used the same weight proportion of EW and LW as in the chip mixture for co-refining. We used the term "Chip Mixing" to signify the co-refining process, and "Pulp Mixing" to indicate the post-refining blending of EW and LW pulps after separate refining. For this study, we employed the pulps produced at 160°C and had freeness range of 50-250 mL.

9.4.1 Effect on fibre properties

9.4.1.1 Fibre length

Refining of mixture of EW and LW chips gave better fibre length than refining of EW and LW chips separately (Figure 9.35). As a result, this translates into less fibre length reduction in the co-refining process (Figure 9.36).

As indicated in 9.37, when compared with the fibre length reduction of EW and LW pulps in their individual refining, thin-walled EW fibres were more fragile and suffered severer fibre cutting (fibre length reduction) than thick-walled LW fibres.

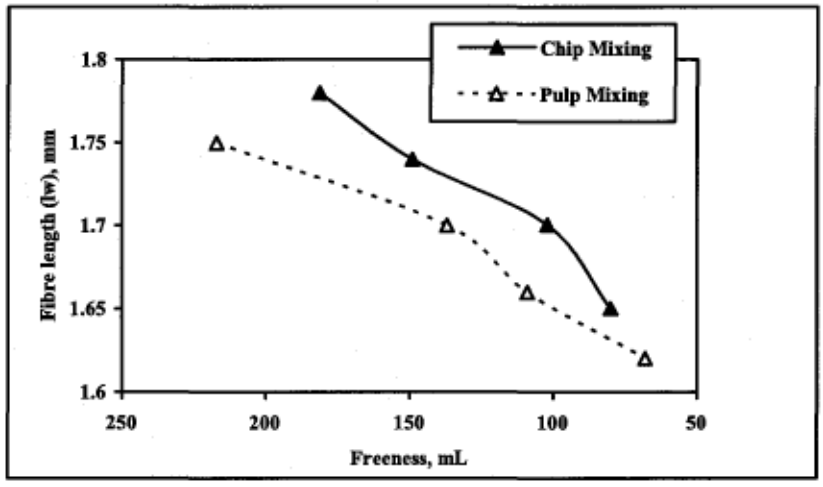

Figure 9.35 Effect of chip mixing and pulp mixing on fibre length (lw) as a function of freeness

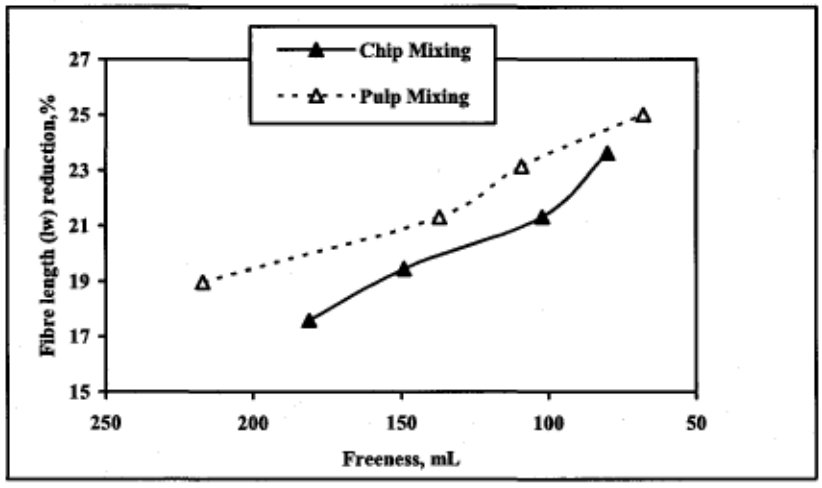

Figure 9.36 Effect of chip mixing and pulp mixing on fibre length (lw) reduction as a function of freeness

While in EW/LW chips co-refining (i.e. "Chip Mixing"), the curve of fibre length reduction lies between EW and LW refining alone, which means EW fibre suffers less severe fibre cutting when co-refined with LW fibres. That might be the fact that the relatively stiffer and thick-walled LW fibres could yield a "buffer effect" on the fragile and thin-walled EW fibres, rendering the latter less vulnerable to be shortened [191]. As a result, the "Chip Mixing" process yields pulps with greater fibre length (Figure 9.35) and less fibre length reduction (Figure 9.36) than in the "Pulp Mixing" process.

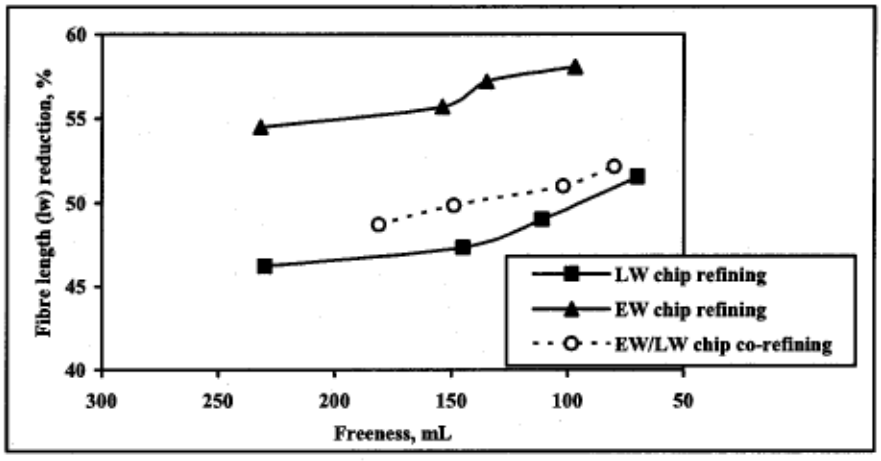

Figure 9.37 Fibre length reduction as a function of freeness with a refining temperature of 160°C

9.4.1.2 Fines content

The direct consequence of fibre cutting will bring about high fines content in refining. As discussed earlier, when compared with the individual refining and co-refining, EW fibres suffered severer fibre cutting in the former refining process. As a result, in the "Pulp Mixing", the weight-weighted fines content are higher in comparison with the "Chip refining" (Figure 9.38).

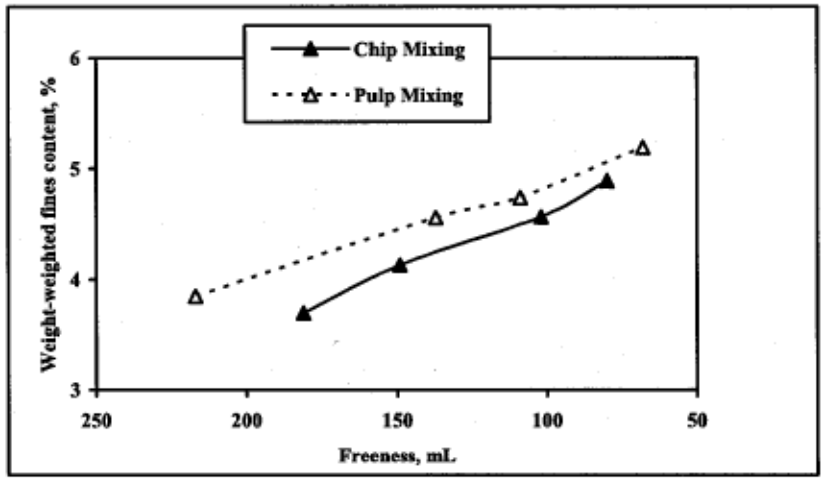

Figure 9.38 Effect of chip mixing and pulp mixing on weight-weighted fines contents as a function of freeness

9.4.1.3 Refining energy

Comparison of co-refining of EW and LW with separate refining indicates that the difference in energy consumption was relatively small between these two processes, as shown in Figure 9.39, despite the fact that the freeness for separate refining at the lowest energy of about 10.2 MJ/kg was particularly high. However, when this particular point was excluded it was observed that there existed some degrees of linearship between the co-refining and separate refining.

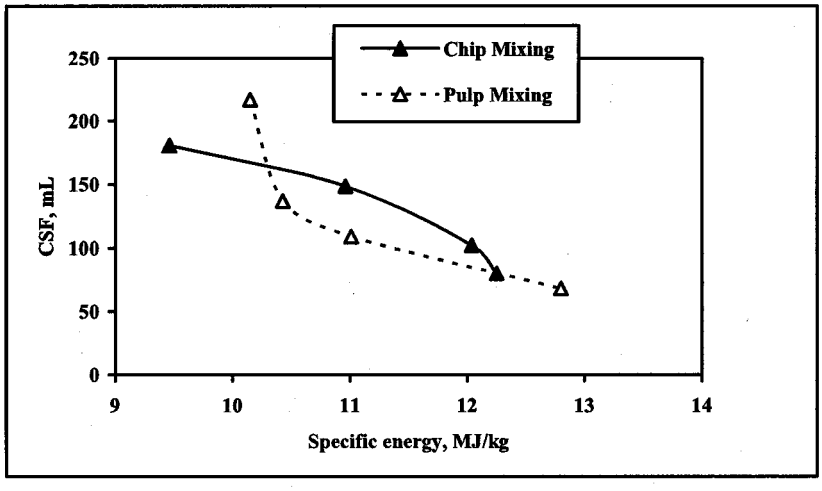

Figure 9.39 Effect of chip mixing and pulp mixing on freeness as a function of refining energy

9.4.2 Effect on sheet properties

9.4.2.1 Density

When EW chips were refined alone severe cutting of thin-walled EW fibre occurred to a greater extent than in the process when they were co-refined with the LW chips (Figure 9.37). As a consequence, from the "Chip Mixing" process to the "Pulp Mixing" process, the average fibre length was reduced (Figure 9.35) and the fines contents increased (Figure 9.38). In co-refining thin-walled EW fibres were expected to be "protected" in some way by thick-walled LW fibres, reducing the generation of fines and the overall average fibre length. Consequently, the handsheets prepared from "Pulp Mixing" showed higher sheet density when compared with the co-refined pulps ("Chip Mixing").

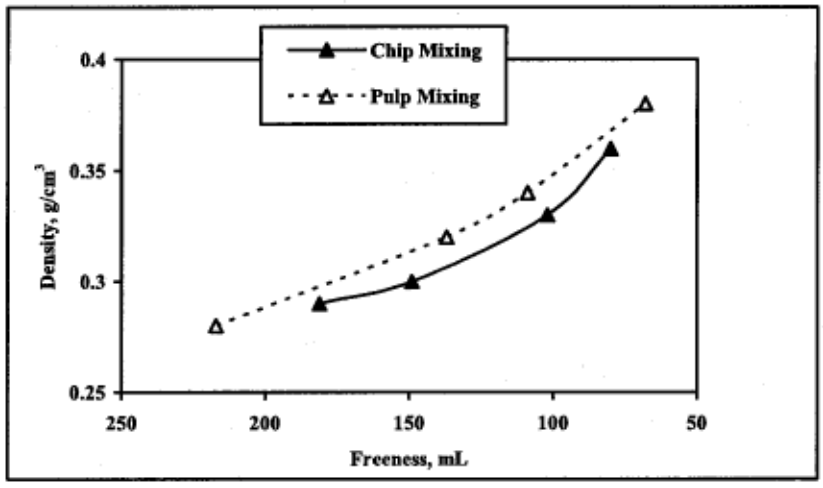

Figure 9.40 Effect of chip mixing and pulp mixing on density as a function of freeness

9.4.2.2 Physical properties

Blending of pulp after separate refining yielded better tensile and burst indices than the co-refining process Figures 9.41 and 9.42 show. These characteristics may be explained by the fact that separate refining of EW and LW generated more fines than the co-refining of EW and LW (Figure 9.38). The increase in fines content is beneficial for fibre bonding, improving the mechanical resistances of paper.

In contrast, the "Chip Refining", or co-refining gave higher tear index than the post-refining blending of pulps or "Pulp Mixing" (Figure 9.43) process. It is because co-refining had less fibre cutting effect than the separate refining, particularly for the thin-walled EW fibres. In co-refining, the less resistant EW fibres are "protected" by thick-walled LW fibres against the harsh mechanical action exerted by the refiner bars, a "buffer effect", thus minimizing the fibre shortening effect. The resulting increase in average fibre length helps improve the tear resistance of handsheets.

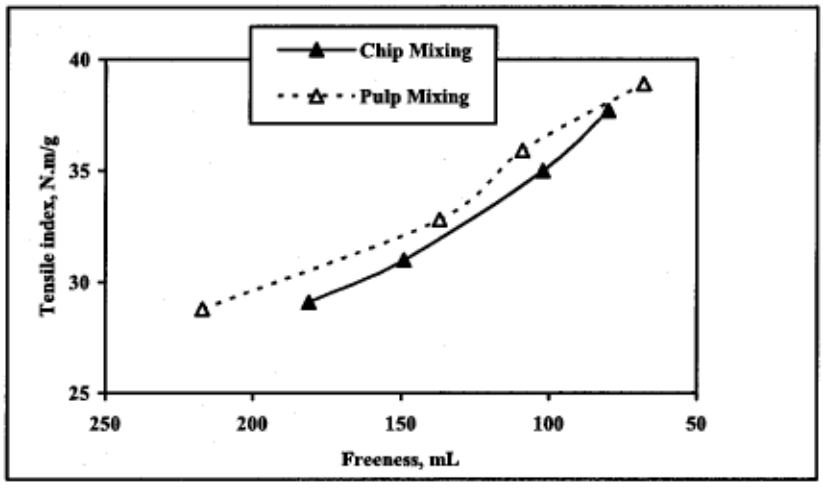

Figure 9.41 Effect of chip mixing and pulp mixing on tensile index as a function of freeness

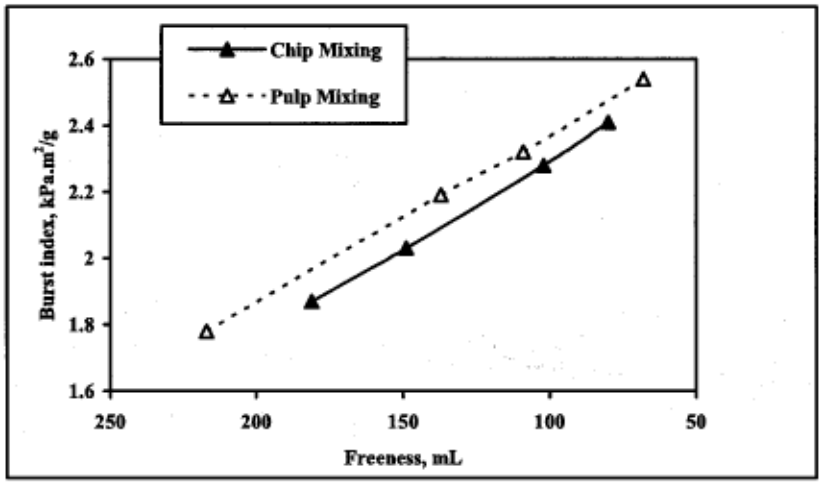

Figure 9.42 Effect of chip mixing and pulp mixing on burst index as a function of freeness

9.4.2.3 Optical properties

"Pulp Mixing" produced handsheets having higher opacity (Figure 9.44) and light scattering coefficient (Figure 9.45) in comparison to the "Chip Mixing" process. Since the pulps used in "Pulp Mixing" were refined separately and contained more fines than the co-refined pulps as in the case of "Pulp Mixing", it is comprehensible that the handsheets prepared from the "Pulp Mixing" process had higher opacity and light scattering coefficient. This is because these two properties are directly influenced by the quantity as well the quality of the fines in the pulps.

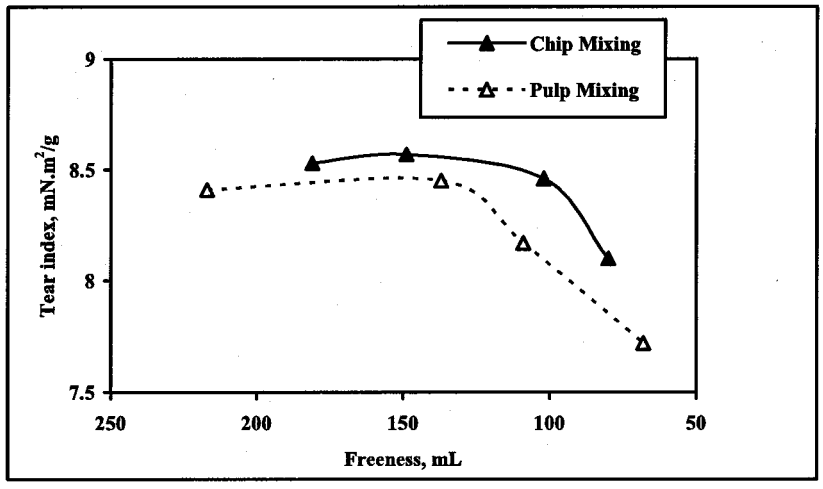

Figure 9.43 Effect of chip mixing and pulp mixing on tear index as a function of freeness

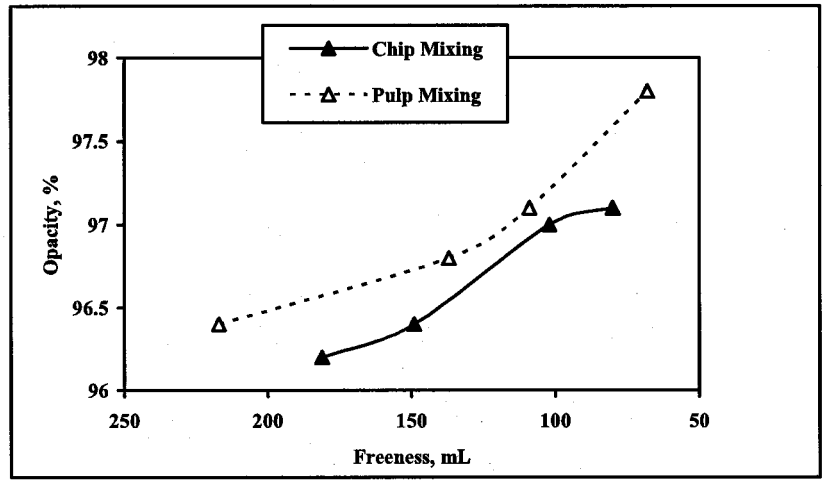

Figure 9.44 Effect of chip mixing and pulp mixing on opacity as a function of freeness

As for the light absorption coefficient (Figure 9.46), there is no difference between these two processes, "Chip Mixing" and "Pulp Mixing" processes. This implies that there are not significant chemical changes occurring between these two pulps, especially in lignin content. It is known that the light absorption coefficient of mechanical pulps is mainly dependent on the chemical components in the wood raw material (i.e. chemical components) [35].

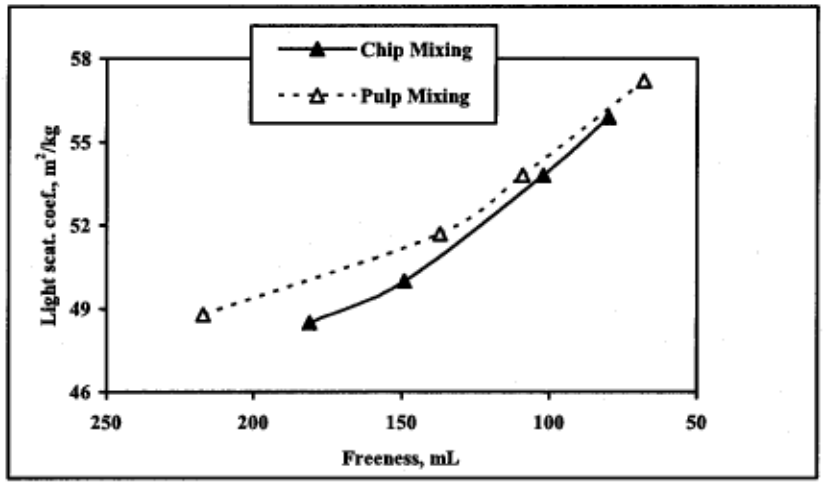

Figure 9.45 Effect of chip mixing and pulp mixing on light scattering coefficient as a function of freeness

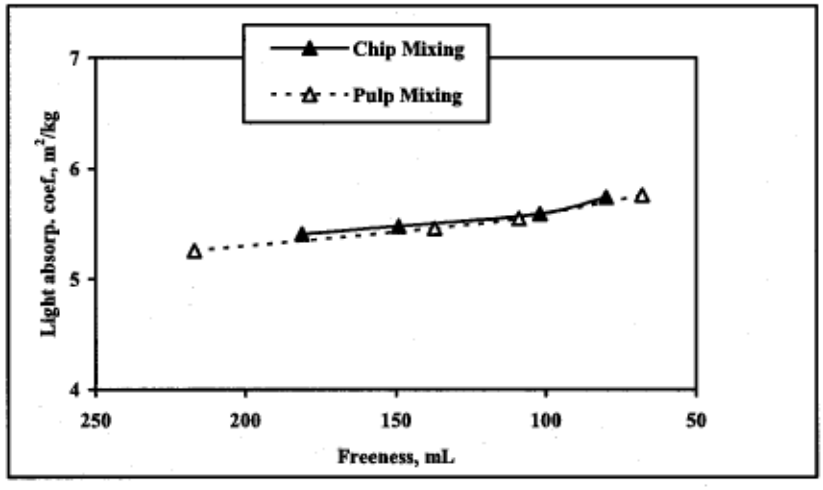

Figure 9.46 Effect of chip mixing and pulp mixing on light absorption coefficient as a function of freeness

Regarding the brightness of handsheets, the "Pulp Mixing" process yielded higher brightness than in the "Chip Mixing" (Figure 9.47).

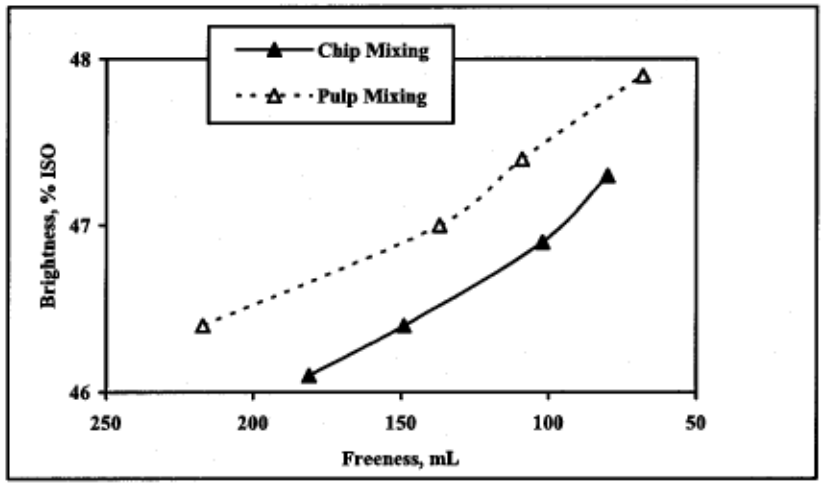

Figure 9.47 Effect of chip mixing and pulp mixing on brightness as a function of freeness

According to the Kubelka-Munk theory [192, 193], the pulp brightness is determined by the light absorption (k) and light scattering (s) abilities. The theory determines the relation between the properties in the following way (Eq. 9.1):

$$R_\infty = 1 + (k/s) - \sqrt{(k/s)^2 + 2(k/s)} \qquad \text{Eq. 9.2}$$

where,

R_∞ = brightness
k = light absorption coefficient
s = light scattering coefficient

The light absorption coefficient is a measure of the quantity of coloured matter in the pulp, while the light scattering coefficient is dependent on the pulping method [35]. For these two handsheets making processes, that is the "Chip Mixing" and "Pulp Mixing" processes, the latter yields higher sheet light scattering (s) coefficient than the former (Figure 9.45). For the light absorption coefficient (k), there is no evident difference between them. As shown in Eq. 9.1, in the comparison of "Chip Mixing" and "Pulp Mixing", the "k" value is constant for both, but the latter had higher "s" value. As a result, the latter ("Pulp Mixing") has a higher brightness (R_∞) in comparison to the "Chip Mixing".

9.5 Scanning electronic microscopic (SEM) study

The SEM can be used to examine the cross-section characteristics of fibres in handsheets including fibre collapsibility, fibre twist angle, and fibre bonding. The purpose of this study is to evaluate the response of EW and LW fibres to the pressurized refining and to establish possible relations between the fibre morphologies and sheet properties.

9.5.1 Collapsibility

The parameter of Form Circle "FC", which has been described in Chapter 6, is employed here to quantitatively measure the collapsibility of EW and LW fibres in handsheets. The whole pulps (WP) with a freeness of 150 mL were used in this study. In addition, the cross-section of handsheets made from R28 fractions were also qualitatively compared with those of whole pulp.

Due to their thin cell wall EW fibres, the handsheets made from the R28 (Figure 9.48) exhibited higher collapse than thick-walled LW (Figure 9.49) which were also from the R28 fraction. This suggests that under the same sheet formation condition, thin-walled EW fibres changed their forms more readily than LW fibres in the fibrous network. Improved collapsibility of EW fibres would lead to better conformability, resulting in higher sheet density in comparison to LW fibres (Figure 9.24). Since the R28 fraction fibres are relatively stiff and less developed, they make sheet with high bulk and poor fibre bonding. For the same reason, LW sheet had higher porosity than LW sheets (Figure 9.5).

In contrast to the sheets made from the R28 fibres, the voids in the handsheets prepared from the WP (Figures 9.50 and 9.51) were less noticeable because the fibrous network was filled with shorter and small fibrous elements. The thin-walled EW fibres in the WP sheet also showed higher collapsibility and conformability than those of LW counterpart.

The characteristics observed above are supported by the quantitative analysis presented in Table 9.6 which shows that the magnitude of FC for EW fibres is considerably lower than that for LW fibres. The mixed furnish containing both EW and LW fibres; the

values of FC were also higher than that for EW. These findings imply that EW fibres had higher collapsibility than LW fibres in the fibre network.

Note that the refining conditions also had an important effect on the FC. As indicated in Table 9.6, the FC values for the refining at 120 °C were somewhat greater than those for the 160°C operation. This suggests that the high temperature (160°C) refining, which was operated with reduced plate gap, improved fibre collapsibility. This is understandable since the reduced refiner plate gap increases the compression force and causes higher mechanical action on fibres, including more fibre collapses.

Table 9.6 Data on Form Circle (FC)

	WP sheet	
	120°C	160°C
EW	0.49	0.45
LW	0.69	0.63
MIX	0.63	0.56

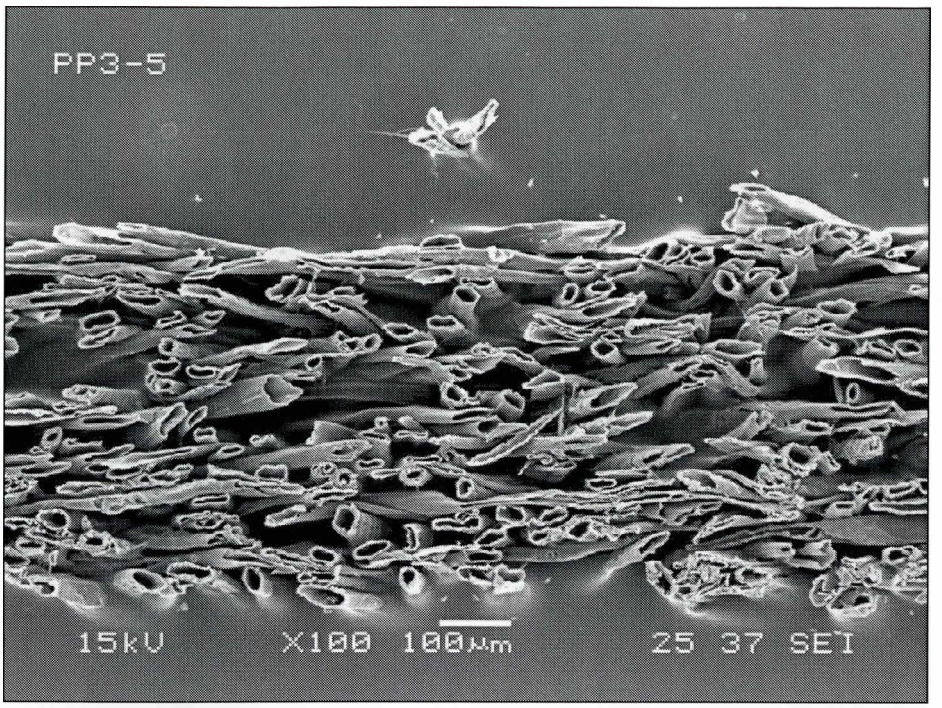

Figure 9.48 **SEM micrograph showing a cross-section of EW sheet made from R28 fraction (CSF: 680 mL, refined at 160°C)**

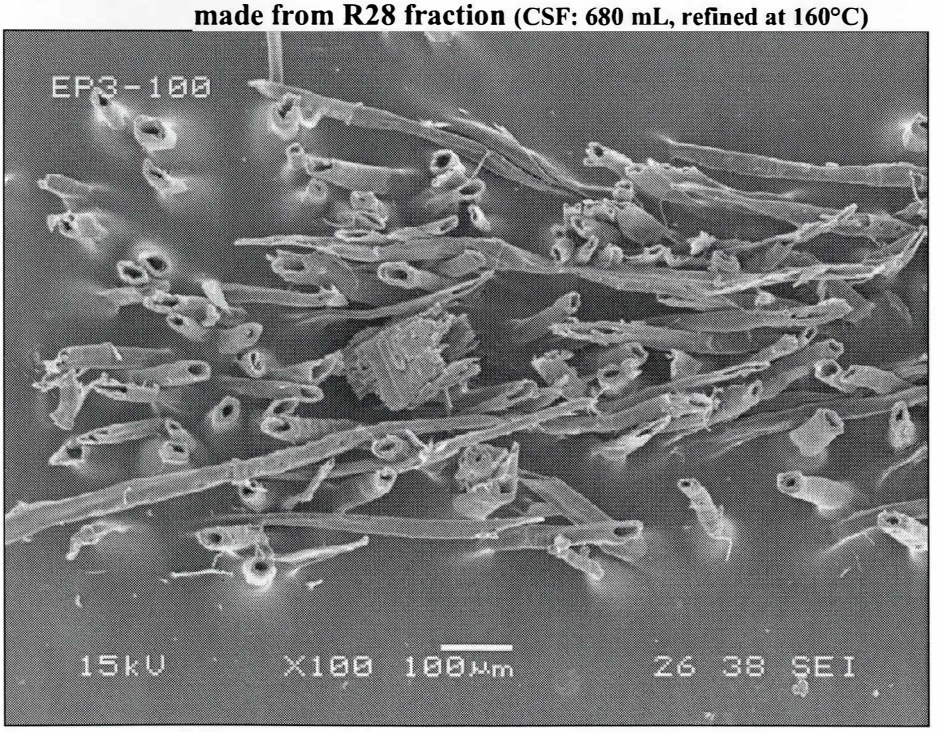

Figure 9.49 **SEM micrograph showing a cross-section of LW sheet made from R28 fraction (CSF: 690 mL, refined at 160°C)**

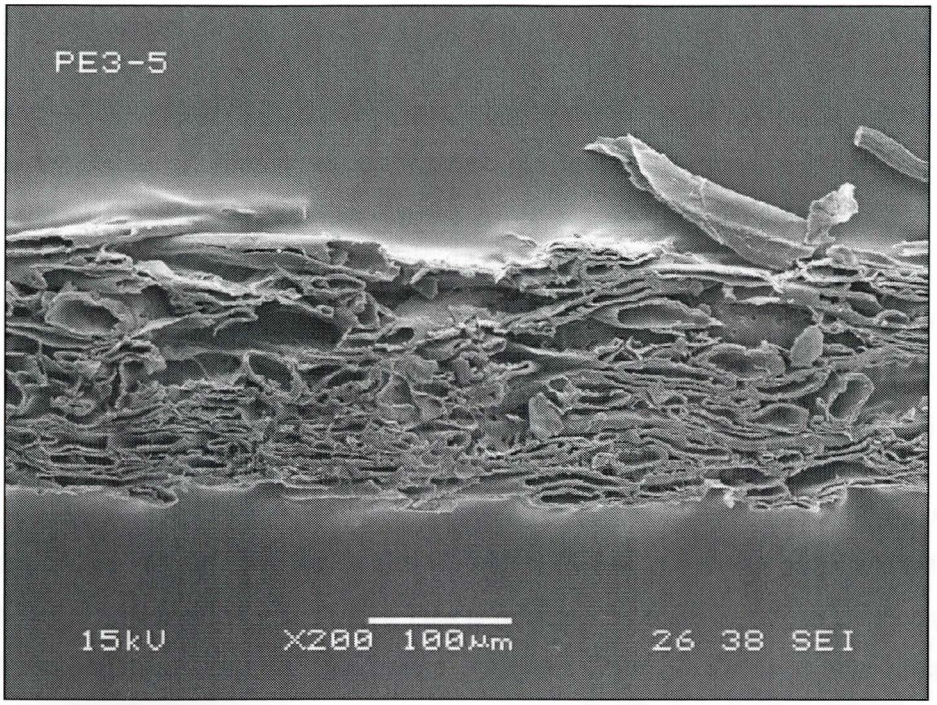

Figure 9.50 **SEM micrograph showing a cross-section of EW sheet made from whole pulp (CSF: 150 mL, refined at 160°C)**

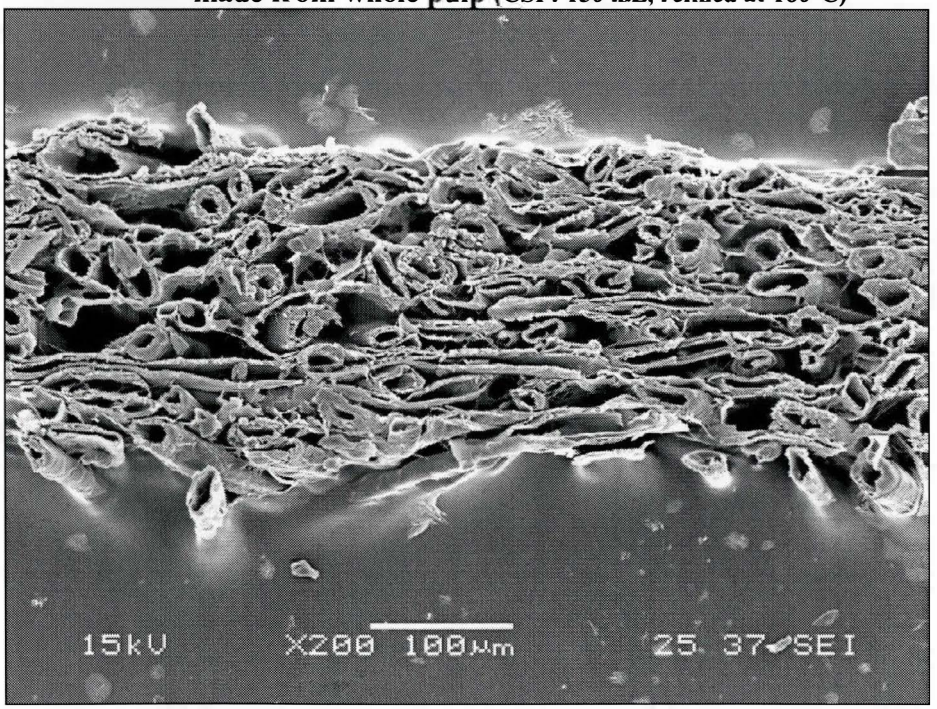

Figure 9.51 **SEM micrograph showing a cross-section of LW sheet made from whole pulp (CSF: 150 mL, refined at 160°C)**

9.5.2 Twist angle

As discussed in section 6.7.1.2, the twist angle was the angle measured between the longest dimension of fibre cross section and the horizontal frame of the image. The orientation of the fibre with respect to the paper plane (fibre twisting) could strongly affect the structure of the paper. The reduction in fibre twist would decrease the amount of space taken by the fibre in the paper structure, thereby reducing the void space and increasing the density of paper. The reduction in fibre twist could also increase the potential bonding surface area of the fibres, especially for the collapsed fibres [156].

Table 9.7 reveals several tendencies. Firstly, the high temperature refining (160°) produced fibres with lower twist angles in both R28 and WP sheet, when compared to the low temperature refining at 120°C. This is because the reduced refiner plate gap used at high temperature refining yielded more collapsed fibres, resulting in less fibre twist during the sheet formation. Secondly, when compared with the WP sheet, the R28 sheet has higher twist angles. This is mainly due to the fact that the R28 fibres were stiffer and less developed than the shorter and smaller elements in the WP pulps, resulting higher twists in the fibre network. Thirdly, the LW fibres always had the higher twist angle than those of EW. In the mixed furnish, which contained LW and EW fibres, the twist angle was greater than that for EW fibres but lower than that for LW fibres. This is because LW fibres are stiffer than thin-walled EW fibres, and have less conformability and more twists than the latter. Owing to their less twists, the EW pulps have higher sheet density than that of EW sheet, as indicated in Figure 9.1.

Table 9.7 Twist angles (°) in different handsheets

	R28 sheet		WP sheet	
	120°C	160°C	120°C	160°C
EW	16.1	10.2	8.5	3.9
LW	32.8	27.6	10.5	11.3
MIX	28.9	24.7	9.1	10.1

9.5.3 Fibre bonding

The measurements on fibre collapsibility and twist angle in the fibre network indicate that EW fibres always show better conformability than LW fibres. Thus, the former has better fibre bonding than the latter due to its higher potential in bonding surface area. But other research work also indicated that the physical strength of TMP pulps is mainly attributed to the fines, especially the fibril-like fines [132]. SEM micrographs of the fibres in the WP of EW (Figure 9.52) show that these fibres had less fibrils even though they have better conformability. But the fibres in the WP of LW (Figure 9.53), had many fibrils, which also favours the fibre bonding.

This finding reveals that EW and LW fibres have their own distinct advantage in relation to the inter-fibre bonding. Although EW fibres have less fibrillations, they have thinner cell wall and, therefore, higher collapsibility and conformability, which are also useful for fibre bonding. On the other hand, despite their thick cell-wall, LW fibres are more readily fibrillated or developed during refining, which also favours fibre bonding. The shortcoming of the EW fibre is their lower fibrillation while that for the LW fibre is its poor conformability. Therefore, for the sake of improving fibre bonding, it would recommendable to blend them together to compensate each one's defaults.

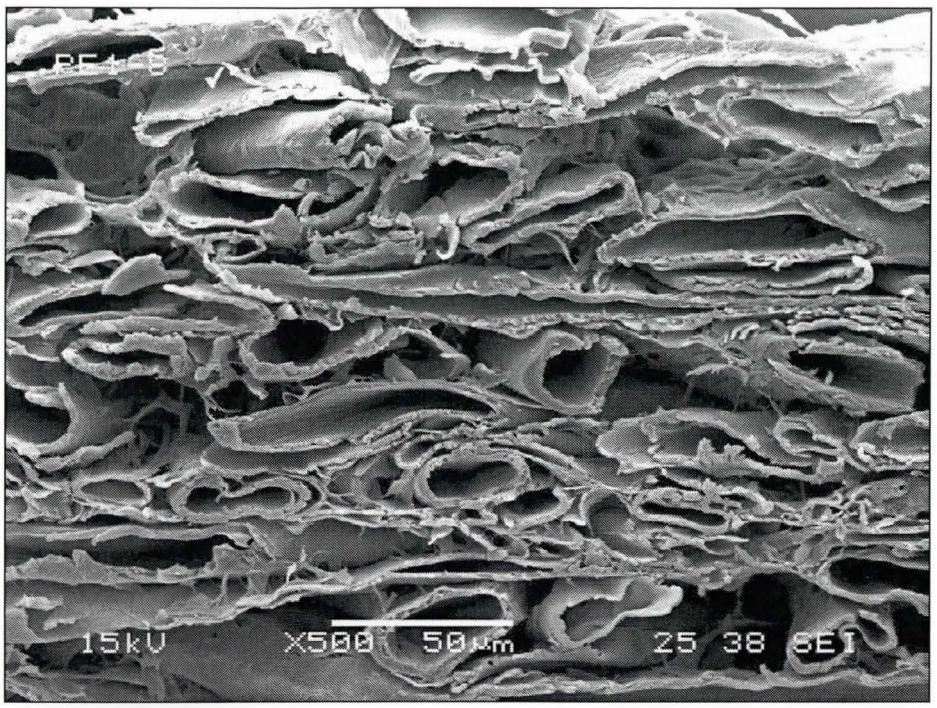

Figure 9.52 **SEM micrograph showing a cross-section of whole pulp sheet containing EW fibres (CSF: 150 mL, refined at 160°C)**

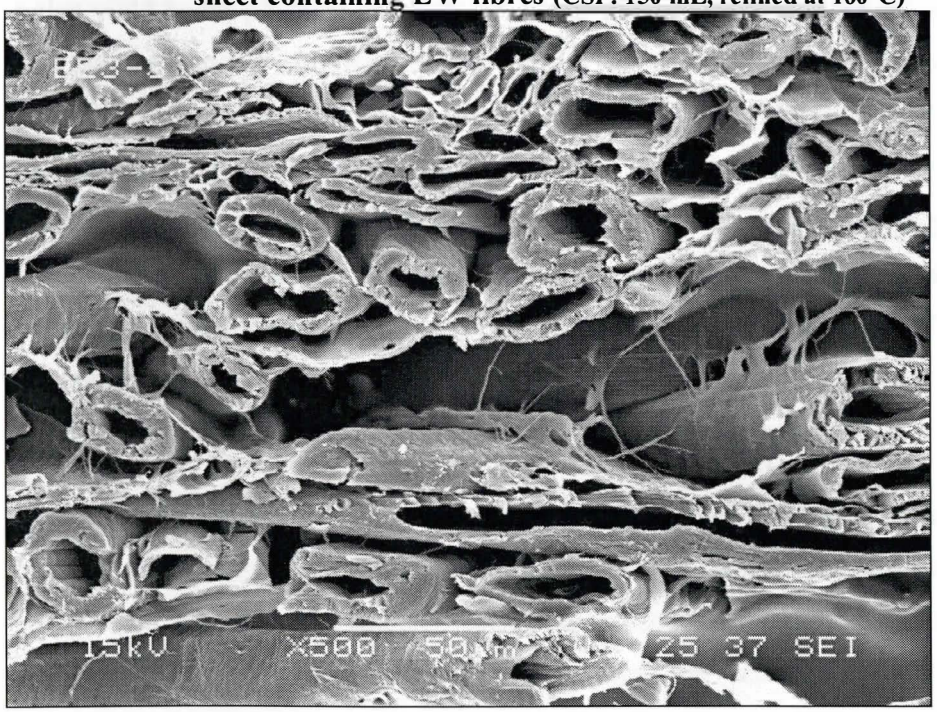

Figure 9.53 **SEM micrograph showing cross-section whole pulp sheet containing LW fibres (CSF: 150 mL, refined at 160°C)**

9.6 Summary

In this part of study we have evaluated the physical and optical characteristics of different pulps which were produced in separate refining of EW, LW and mixed furnish made in co-refining of both EW and LW. Other furnishes examined include whole pulp (WP), fines-free pulps (WP-P200), and pulps of different Bauer-McNett fractions or in combination. We also compared the differences in papermaking properties between "Chip Mixing" in co-refining of EW and LW chips and "Pulp Mixing" or post-refining mixing of separately refined EW and LW pulps. The principal points of findings are summarized as following:

1. For a given freeness, EW whole pulps have higher sheet density, lower roughness, porosity and opacity than LW pulps do because EW fibres have better fibre conformability;

2. When the comparison is made at a given specific refining energy, the LW whole pulps yield better physical strengths, in terms of Scott bond, tensile, burst, and tear indices than EW counterparts. Greater degrees of fibrillation of thick-walled LW fibres are responsible for the differences. In terms of brightness EW pulps exhibit higher values than LW pulps due to their higher initial brightness. On the other hand, EW pulps show higher light absorption coefficient because they have higher lignin content when compared to LW pulps. However, EW pulps have better light scattering coefficient than LW pulps because the fines in EW pulps containing more flake-like particles which increase the scattering of light in the sheet structure, in contrast, LW fines have more fibillar elements;

3. In comparison to the low temperature (120°C) refining, the high temperature (160°C) operation produced pulps having better sheet density, physical strength (Scott bond, tensile, burst, and tear resistances), sheet opacity and light scattering and absorption coefficient. These characteristics are accounted for by the increase in fines content and fibre collapsibility. On the other hand, the surface roughness, porosity, and brightness are negatively affected;

4. Among the Bauer-McNett fibre fractions, e.g. R14 to R100, the short fibres are more developed, resulting in lower freeness and coarseness, and they yield handsheets with better strength properties;

5. Despite their low degree of fibrillation the fibre fractions (R14 to R100), EW pulps always exhibit better physical properties than LW furnishes do. Even when the fines are removed from the pulps, EW pulp fractions still have higher physical properties than those of LW furnishes. This is because EW fibres posses greater conformability during sheet consolidation and, hence, better fibre bonding;

6. We have observed that the TMP fines play a very important role in determining the sheet density, tensile, burst and tear indices, surface roughness, sheet porosity and the optical characteristics. In addition, we also noted that LW fines have greater influences than EW fines in affecting the physical properties of handsheets because they contain a higher proportions of fibrils;

7. In co-refining or "EW/LW Chip Mixing" process, the fibres suffer less shortening or breakage, probably due to a possible "buffer effect" by which thin-walled EW fibres are "protected" by thick-walled LW counterpart from the harsh mechanical treatment. In contrast, in separate refining of EW and LW chips, EW fibres experience more cutting and produce more fines. Therefore, when the separate refining pulps (EW and LW) are recombined in handsheet making ("EW/LW Pulp Mixing") the resulting sheets have higher fines content. As results, the "Chip Mixing" process gives greater average fibre length and, hence, better tearing resistance as compared to the "Pulp Mixing" process. In contrast, the fibres in the "Pulp Mixing" process are more developed and have more fines, resulting in increased physical properties and light scattering coefficient;

8. In terms of sheet structure, thin-walled EW fibres have better collapsibility and conformability and thus make sheets with higher density while thick-walled LW fibres produce sheet with higher bulk and have greater fibre twists and fibrils in the network.

Chapter 10 - Conclusions

The main purpose of this study was to examine the physical responses of the earlywood (EW) and latewood (LW) of Jack pine in thermomechanical pulping (TMP). In this investigation we studied the changes in morphology of both EW and LW following two stages of refining (pressurized primary stage and atmospheric secondary stage). The pressurized stage was conducted at two temperatures: 120°C and 160°C.

We also evaluated the impacts of the fibre's morphological modifications on the papermaking characteristics of the resulting pulps.

The main findings of the investigation are summarized in the following paragraphs:

- Characteristics of raw material

Jack pine has a large proportion of LW fibres, 60% by mass, and a density of 0.49 g/cm^3 which is 1.6 times greater than that of EW (0.30 g/cm^3). Additionally, LW fibre of Jack pine has a cell wall thickness of 4.75 μm, which is approximately two times greater than that of EW fibre (2.12 μm), and a cell diameter (radial width) two times smaller than that of EW. All these differences in fibre characteristics between LW and EW fibres are responsible for the dissimilar refining behaviours of these two wood tissues. LW fibre is physically more resistant to the refining actions than the EW counterpart, resulting in different failure modes and dissimilar papermaking attribute.

- Characteristics of pulp, for a given specific refining energy

1. Due to their morphological differences, EW and LW fibres behave differently in thermomechanical pulping (TMP), namely fibre separation and fibre development. During refining, thin-walled EW fibres tend to separate in the P/S1 interface and show little external fibrillation. While the fibre separation of thick-walled LW fibres takes place commonly in the P/S1 and S1/S2 regions, generating considerable amounts of external fibrillation. In addition, the fragile and thin-walled EW fibres manifest more noticeable delamination (internal

fibrillation) and have higher water retention value (WRV) than the thick-walled LW fibres.

2. Thick-walled LW fibres are more resistant to the refining actions than EW counterpart. Under the harsh mechanical actions, LW fibres exhibit mostly intra-wall failure and lower curl and kink indices, while EW fibres tend to fail in trans-wall mode (splitting) and show higher curl and kink indices. As a result, the LW yields higher fibres length while EW suffers more fibre cutting mainly due to its thin cell wall;

3. Under the compression forces existing in refining, EW and LW fibres have different cross-section deformations. Thanks to their thinner cell wall and larger lumen, EW fibres show higher collapsibility and conformability than the LW counterparts. As a result, after refining, EW fibres have lower Form Circle (FC) and Form Shape (FS) values than those of LW.

4. The different physical response of EW and LW fibres in refining influences directly their fines and rejects productions. During refining, thick-walled LW fibres experience considerable surface peeling effect (external fibrillation) and produce more fibillar components in the fines. In contrast, the fragile thinned-walled EW fibres yield more unfibrillated and flake-like fragments in the fines. As a consequence, EW fines have higher surface lignin coverage while the LW fines give higher specific volume (SV). Mainly due to their differences in fibre development, the rejects of LW and EW are qualitatively and quantitatively different. LW rejects have more fibrillar elements when compared with EW rejects; the latter contains more bundle-like shives. The aggregates of fibrils give LW a higher rejects content when compared to EW counterpart;

5. The change of refining temperature from 120°C to 160°C also has different impacts on the behaviours of EW and LW in refining. High temperature refining at 160°C, which operates with reduced plate gap, produces more rejects and generates more fibrillation from LW but more collapsed fibres from EW when compared with the refining at 120 °C.

- Characteristics of handsheets

1. The morphological modifications of EW and LW in refining strongly influence the papermaking characteristics of the resulting pulps, namely the physical and optical properties. For a given freeness, thanks to their better fibre conformability, the fibres of EW whole pulps produce handsheets with higher sheet density, lower roughness, porosity and opacity than those of LW pulps. At a given specific energy, mainly due to highly fibrillated fibres and fibril-rich elements in the fines, the fibres of LW whole pulps yield handsheets with better physical strengths, such as Scott bond, tensile, burst, and tear indices when compared to the EW counterparts. In addition, the EW pulps are brighter than the LW pulps due to their higher initial brightness. On the other hand, the EW pulps show higher light absorption coefficient because they have higher lignin content when compared to the LW pulps. However, EW pulps have better light scattering coefficient than LW pulps because the fines in EW pulps contain more flake-like particles which increase the scattering of light in the sheet structure.

2. In comparison to the low temperature (120°C) refining, for both EW and LW, the high temperature (160°C) operation produced pulps having better sheet density, physical strength (Scott bond, tensile, burst, and tear resistances), sheet opacity and light scattering and absorption coefficient. These characteristics are accounted for by the increase in fines content and improved fibre collapsibility. On the other hand, the surface roughness, porosity, and brightness are negatively affected.

3. Despite of their low degree of fibrillation of the Bauer-McNett fibre fractions (R14 to R100), EW pulps always exhibit better physical properties than LW furnishes do. This is because EW fibres have greater conformability during sheet consolidation and, hence, better fibre bonding.

4. The TMP fines play a very important role in determining the sheet density, tensile, burst, tear indices, surface roughness, sheet porosity and the optical characteristics. Due to the differences in fines components, LW and EW fines

haves different contributions to the physical properties of final pulps. LW fines have greater influences than EW fines in affecting the physical properties of handsheets because they contain a higher proportions of fibrils which are more favourable for fibre bonding than the flake-rich elements in EW fines.

5. The interactions between EW and LW in co-refining of EW and LW chips or "Chip Mixing" process are different than those observations when blending of EW and LW pulps after separate refining or "Pulp Mixing" process, producing final pulps having different physical and optical characteristics. In separate refining of EW and LW chips, EW fibres experience more cutting and produce more fines. Therefore, when the separate refining pulps (EW and LW) are recombined in handsheet making (i.e. "Pulp Mixing" process) the resulting sheets have higher fines content. In the "Chip Mixing" process, probably due to a possible "buffer effect" by which the thin-walled EW fibres are "protected" by thick-walled LW counterparts from the harsh mechanical treatment, the fibres have greater average fibre length and, hence, better tearing resistance as compared to the "Pulp Mixing" process. In contrast, the fibres in the "Pulp Mixing" process are more developed and have more fines, resulting in increased physical properties and light scattering coefficient than the "Chip Mixing" process.

6. In terms of sheet structure, thin-walled EW fibres have better collapsibility and conformability and thus make sheets with higher density while thick-walled LW fibres produce sheet with higher bulk and have greater fibre twists and fibrils in the network.

Chapter 11 - Recommendations

The morphological and chemical differences between earlywood (EW) and latewood (LW) of Jack pine strongly influence their refining behaviours and the final sheet properties. When compared with the traditional pulping species – black spruce, Jack pine produces paper with inferior physical strength mainly due to its high LW proportion. In addition, the resin-rich EW is the origin of its "pitch problem" in papermaking. By means of this study, I would like to propose some recommendations to overcome these problems caused by the differences between EW and LW and improve the quality of Jack pine TMP.

1. During refining, thin-walled EW fibres tend to disintegrate into tiny fibrous elements and suffer more fibre cutting. While thick-walled LW fibres are more resistant to the mechanical refining forces, they are readily to retain their form and produced more long fibres. As a result, in the fibre classification, the majority of long fibre fractions (R14+R28+R48) are derived from LW. In addition, these long LW fibres are less developed and have higher coarseness and inferior inter-fibre bonding abilities. In order to improve the pulp qualities, it would be preferable to separate EW fibres by fractionation from the long fibres before adding more energy to develop these thick-walled LW fibres. It is desirable to refine these long fibres or LW fibres at an elevated temperature or with chemical treatment to soften the fibres and minimize the fibre cutting. That would certainly help improve the inter-fibre bonding potential;

2. Efforts on genetic engineering studies such as cloning and hybridization or other biological engineering approaches should be emphasized to produce Jack pine with thinner fibre wall and lower extractives content or different extractives composition that causes no adverse effect on pulping and bleaching. "Genetic engineering" most often refers to a technique used to transfer small fragments of genetic material (genes which are composed of DNA) from one organism to another. The transferred gene carries a new characteristic or trait that is expressed in the engineered organism [194]. In recent years, the genetic

engineering techniques were widely applied to modify pulpwood growth, structure, and to maximize pulp yields with minimal energy consumption and environment impacts [195, 196]. For example, Tsai *et. al.* [197] had genetically augmented syringyl lignin biosynthesis in low-lignin European aspen (*Populus tremula*) and enhanced lignin reactivity during chemical pulping. Moreover, Sundberg *et.al.* [198] had also genetically modified the structure of auxin (a kind of growth hormone) of a hybrid aspen to control the tree growth and wood properties. Based on these theories and present techniques, it is possible to change the EW and LW growth rates such as the reduction and (or) retardation of the formation of thick-walled LW, which would be beneficial for the fibre development in thermomechanical pulping of Jack pine.

3. The "pitch problem" may be minimized by biological, mechanical, physical or chemical treatments, which would be also useful to improve the pulp quality and extend the exploitation of Jack pine. For example, using seasoning at elevated temperature (e.g. oxidation) and biological treatment of chips would reduce wood resin and other extractives [199]. Deresination and extractives removal by mechanical extraction with or without chemical aid or by inter-stage washing could also eliminate the resins [200]. Furthermore, extraction of wood chips or pulp using a solvent might offer another solution to the pitch problem relating to wood extractives [201].

Bibliography

1. Reme, P.A., and Johnson, P.O., 1999. *"Changes Included in Early- and Latewood Fibres by Mechanical Pulp Refining"*, Nordic Pulp & Paper Res. J., 14(3), p. 256-262.

2. Law, K.N., 2001. *"Mechanical Behaviour of Early- and Latewood under Compression Load"*, Proc.Intl. Mech. Pulp. Conf., Helsinki, Finland, p. 159-166.

3. Wimmer, R. 1995. *"Intra-annual Cellular Characteristics and Their Implications for Modeling Softwood Density"*, Wood Fiber Sci. 27(4), p. 413-420.

4. Jain, K.K., and Seth, M.K., 1979. *"Intra-increment Variation in Specific Gravity of Wood in Blue Pine"*, Wood Sci. Technol. 13(4), p. 239-248.

5. Gamov, V.V., 1975. *"Tensile Strength of Larch Spring- and Summerwood in the Direction Perpendicular to the Fibers"*, Izv. VUZ. Lesnoi. Zh. 18(4), p. 161-163.

6. Hartler, N., and Nyren, J., 1969. *"Influence of Pulp Type and Post-treatments on the Compressive Force Required for Collapse"*, TAPPI STAP No. 8, p. 265-270; discn.,p. 271-277.

7. Jones, T.G., and Richardson, J.D., 1999. *"Relationships between Wood and Chemimechanical Pulping Properties of New Zealand Grown Eucalyptus Nitens Trees"*, Appita J. 52(1), p. 51-56.

8. Law, K.N., Valade, J.L., and Daneault, C., 1989. *"Chemimechanical Pulping of Tamarack, (2). Effects of pH and Sodium Sulfite"*, Cellul. Chem. Technol. 23(6), p. 733-741.

9. Reme, P.A., and Helle, T., 2001. *"Quantitative Assessment of Mechanical Fibre Dimensions During Defibration and Fibre Development"*, J. Pulp Paper Sci. 27(1), p. 1-7.

10. Murton, K.D., Richardson, J.D., Corson, S.R., and Duffy, G.G., 2001. *"TMP Refining of Radiata Pine Earlywood and Laterwood Fibres"*, Proc. Intl. Mech. Pulp. Conf., Helsinki, Finland, p. 361-371.

11. Agut, P., Gouttenoire, P., Michalowicz, G., Robert, A., Choudens, C.D., and Lombardo, G., 1987. *"Destructuration of Wood Chips in a Roller Press – Morphological Aspects and Application to Kraft and Bisulfite Cooks"*, Poster Presentations, Vol. 2, Proc. 4th Intl. Symp. Wood & Pulping Chem., Paris, France. p.149-155.

12. Luner, P. 1986. *"Wet fiber Flexibility as An Index of Pulp and Paper Properties, New Technologies in Refining"*, Vol. 1, Session 1, Paper 3, Proc. PIRA Intl. Conf., Birmingham, England. p. 26.

13. Hattula, T., and Niemi, H., 1988. *"Sulfate Pulp Fiber Flexibility and Its Effect on Sheet Strength"*, Paperi ja Puu 70(4), p. 356-361.

14. Uhmeier, A., and Salmén, L., 1996. *"Repeated Large Radial Compression of Heated Spruce"*, Nordic Pulp Paper Res. J. 11(3), p.171-176.

15. Mohlin, U.-B., 1975. *"Cellulose Fiber Bonding. (5) Conformability of Pulp Fibers"*, Svensk Papperstid. 78(11), p. 412-416.

16. Smith, W.E., and Byrd, V.L., 1972. *"Fiber Bonding and Tensile Stress-strain Properties of Earlywood and Latewood Handsheets"*, U.S. Forest Serv., Res. Paper FPL 193,11p.

17. Hartler, N., and Nyren, J., 1970. *"Transverse Compressibility of Pulp Fibers II, Influence of Cooking Method, Yield, Beating, and Drying"*, Tappi J. 53(5), p. 820-823.

18. Laurent, J.M., Rudie, A.W., and Shakhet, A.R., 1993. *"Mechanical Pulping by Fraction after Low Energy Refining"*, 1993 TAPPI Pulping Conference, Atlanta, USA, p. 95-99.

19. Salmén, L., Dumail, J.F., and Uhmeier, A., 1997. *"Compression Behavior of Wood in Relation to Mechanical Pulping"*, Proc. Intl. Mech. Pulp. Conf., Stockholm, Sweden, p. 207-211.

20. Rehmat, T., and Branion, R., 1995. *"Fibre Fractionation in Hydrocyclones"*, 81st Annual Meeting, Technical Section, CPPA, Montreal, Canada, p.B105-B125.

21. Salmén, L.; and De Ruvo, A, 1985. *"Model for Prediction of Fiber Elasticity"*, Wood Fiber Sci. 17(3), p. 336-350.

22. Law, K.N., and Yang, K.C., 2003. *"Characteristics of Fibre Separation in Wood Loaded by Normal-to-grain Compression"*, Appita. J., 56(1), p.42-45.

23. Saiki, H. 1970. *"Proportion of Component Layers in Tracheid Walls of Earlywood and Latewood of some Conifers"*, J. Japan Wood Res. Soc. (Kokuzai Gakkaishi) 16(5), p. 244-249.

24. Butterfield, B.G., 1998. *"Microfibril Angle in Wood"*, IAWA/INFRO International Workshop on the Significance of Microfibril Angle to Wood Quality, University of Canterbury, Christchurch, New Zealand.

25. Ifju, G., and Labosky, P. Jr. 1972. *"Study of Loblolly Pine Growth Increments (1), Wood and Tracheid Characteristics (2), Pulp Yield and Related Properties"*, Tappi. J. 55(4), p. 524-534.

26. Stamm, A.J. 1973. *"Maximum Effective Lumen and Pit Pore Sizes of the Earlywood and the Latewood of Never Dried Loblolly Pine Sapwood"*, Wood Sci. Technol. 7(3), p. 212-217.

27. Thomas, R.J., and Scheld, J.L., 1967. *"Distribution and Size of the Inter-tracheid Pits in an Eastern Hemlock"*, Forest Sci. 13(1), p. 85-89.

28. Petit-Conil, M., Robert, A., and Pierrard, J., 1997. *"Fundamental Principles of Mechanical Pulping from Softwoods and Hardwoods; Theoretical Aspects"*, Cellul. Chem. Technol. 31(1/2), p. 93-104.

29. McIntosh, D.C., and Uhrig, L.O., 1968. *"Effect of Refining on Load-elongation Characteristics of Loblolly Pine Holocellulose and Unbleached Kraft Fibers"*, Tappi. J. 51(6), p. 268-273.

30. Johansson, K., Thuvander, F., and Germgård, U., 2001. *"Single Fibre Fragmentation: a New Measure of Fibre Strength Loss during Brown Stock Washing and Oxygen Delignification"*, Appita. J. 54(3), p. 276-280.

31. Zink, A.G., Pellicane, P.J., and Shuler, C.E., 1994. *"Ultrastructural Analysis of Softwood Fracture Surfaces"*, Wood Sci. Technol. 28(5), p. 329-338.

32. Saka, S., and Tsuji, M., 1987. *"Relationship between the Microfibril Orientation in the Tracheid S2 Layer and the Lignin Content of Coniferous Woods"*, Cellul. Chem. Technol. 21(3), p. 225-231.

33. Khattak, T.M., and Mahmood, A., 1986. *"Estimation of Lignin, Holocellulose and Alpha-cellulose Content of Earlywood and Latewood among Innerwood and Outerwood of Blue Pine (Pinus wallichiana, A.B. Jacks.)"*, Pakistan J. Botany 18(2), p. 235-241.

34. Leask, R.A., and Kocurek, M.J., 1987. *"Pulp and Paper Manufacture"*, Volume 2, *"Mechanical Pulping"*, TAPPI/CPPA, Atlanta, USA, p.1-17, 98-112.

35. Sundholm, J., 1999. *"Papermaking Science and Technology, Mechanical Pulping"*, Finnish Paper Engineering's Association & TAPPI, Helsinki, Finland, p. 35-51, 223-249, 195-218, 313-343.

36. Harkonen, E., and Tienvieri, T., 2001. *"Energy Saving in TMP Pulping"*, Proc. Intl. Mech. Conf., Helsinki, Finland, p. 547-556.

37. Law, K.-N., and Lanouette, R., 2000. *"Effect of Mechanical Conditioning of Chips on the Quality of Softwood TMP"*, Pulp Pap Can. 101(7), p. 31-35.

38. De Montmorency, W.H., 1965. *"The Relationship of Wood Characteristics to Mechanical Pulping"*, Pulp Paper Mag. Can. 66(6), p. T325-T348.

39. Karnis, A., 1994. *"The Mechanism of Fibre Development in Mechanical Pulping"*, J. Pulp Paper Sci. 20(10), p. J280-J288.

40. Sundholm, J., 1993. *"Can We Reduce Energy Consumption in Mechanical Pulping"*, Proc. Intl. Mech. Pulp. Conf., Oslo, Norway, p. 133-142.

41. Kano, T., Iwamida, T., and Sumi, Y., 1982. *"Energy Consumption in Mechanical Pulping"*, Pulp Paper Can. 83(6), p. T151-T161.

42. Reme, P.A., and Helle, T., 2000. *"Fibre Characteristics of Shives Initiating Web Rupture"*, Nordic Pulp Paper Res. J. 15(4), p. 287-291.

43. Hattula, T., and Niemi, H., 1988. *"Sulfate Pulp Fiber Flexibility and Its Effect on Sheet Strength"*, Paperi ja Puu 70(4), p. 356-361.

44. Aspler, J.S., and Beland, M.C., 1994. *"Review of Fibre Rising and Surface Roughening Effects in Paper"*, J. Pulp Paper Sci. 20(1), p. 27-32.

45. Fengel, D., and Wegner, G., 1984. *"Wood Chemistry, Ultrastructure, Reactions"*, W. de Gruyter, New York, USA, p. 7-25.

46. Core, H.A., Côté, W.A., and Day, A.C., 1981. *"Wood Structure and Identification"*, 2nd Edition, Syracuse University Press, New York, USA, p. 14-18, 25-27, 49, 92, 106.

47. Panshin, A.J., and Zeeuw, C.D., 1970. *"Textbook of Wood Technology"*, 3rd Edition, Vol.1, McGraw-Hill, New York, USA, p. 48-51, 91, 151-177.

48. Larson, A., 1960. *"Physiological Consideration of Springwood Summerwood Transition in Red Pine"*, Forest Sci., 6, p. 110-122.

49. Little, C.H.A, and Eidt D.C., 1968. *"Effects of Abscisic Acid on Bud-break and Transpiration in Woody Species"*, Nature 220, p. 498-499.

50. Funada, R.; Kubo, T.; Tabuchi, M., Sugiyama, T.; and Fushitani, M., 2001. *"Seasonal Variations in Endogenous Indole-3-acetic Acid and Abscisic Acid in the Cambial Region of Pinus Densiflora Sieb. Et Zucc., Stems in Relation to EW-latewood Transition and Cessation of Tracheid Production"*, Holzforschung 55(2), p. 128-134.

51. Kocurek, M.J., and Stevens, C.F.B., 1983. *"Pulp and Paper Manufacture", Volume 1, "Properties of Fibrous Raw Materials and Their Preparation for Pulping"*, TAPPI/CPPA, Atlanta, USA, p. 11-21, 23.

52. Mark, R. E., Habeger, C.C., Jr., Borch, J, and Lyne, M.B., 2002. *"Handbook of Physical Testing of Paper"*, Volume 1, Second Edition, Marcel Dekker, New York, USA, p. 700-720.

53. Stenius, P., 1999. *"Papermaking Science and Technology", Book 3, "Forest Products Chemistry"*, Finnish Paper Engineering's Association/TAPPI, Helsinki, Finland, p. 50-65.

54. Bisset, I.J.W., 1950. Dadswell, H.E., Australian Forestry, 14(1).

55. Hatvani, T.G., Evans, R., Kibblewhite, R.P., and Parker, I.H., 1999. *"Relationships Between Tracheid and Kraft Pulp Fibre Transverse Dimensions"*, Proc. 53rd Appita Ann.Gen.Conf., Rotura, New Zealand, p. 87-94.

56. Strelis, I., and Kennedy, R.W., 1967. *"Identification of North American Commercial Pulp Wood and Pulp Fibres"*, University of Toronto Press, Toronto, Canada, p. 5-9, 29.

57. Ingruber, O.V., Kocurek, M.J., and Wong, A., 1985. *"Pulp and Paper Manufacture - Volume 2: Sulfite Science & Technology"*, TAPPI/CPPA, Atlanta, USA, p. 159-162.

58. Yang, K.C., and Benson, C., 1997. *"Ultrastructure of Pits in Pinus banksiana Lamb"*, Wood. Sci. & Technol., 31(1), p. 153-169.

59. Cochaux, A., D'Aveni, A., and Robert, A., 1995. *"Étude Fondamentale de la Résistance au Déchirement des Pâtes et Papiers I: Considérations Générales"*, Cellul. Chem. Technolo. 29(1), p. 45-54.

60. Cochaux, A., Moutet, B., D'Aveni, A., and Robert, A., 1995. *"Étude Fondamentale de la Résistance au Déchirement des Pâtes et Papiers. II. La Résistance au Déchirement en Fonction des Paramètres Morphologiques"*, Cellul. Chem. Technol. 29(2), p. 215-22.

61. Howard, E.T., and Manwiller, F.G., 1969. *"Anatomical Characteristics of Southern Pine Stemwood"*, Wood Sci., 2(2), p. 77.

62. Ward, R.J., Côté. W.A.,Jr., and Day, A.C., 1964. *"The Wood Structure-coating Interface"*, Official Digest, 36(477), p. 1091.

63. Sjöström, E., 1981. *"Wood Chemistry, Fundamentals and Applications"*, Academic Press, New York, USA, p. 1-20.

64. Ruel, K., and Barnound, F., 1978. *"Quantitative Determination of Tension Wood in Beech: Statistical Significance of the Galactose Content"*, Holzforschung, 32(5), p. 149-156.

65. Fengel, D., and Stoll, M., 1973. *"Variation in Cross-sectional Area of the Cell, and Thickness of the Cell Wall and Wall Layers in Sprucewood Tracheids within an Annual Ring"*, Holzforschung, 27 (1), p. 1-7.

66. Sjöström, E, 1992. *"Wood Chemistry: Fundamental and Applications"*, 2nd ed., Academic Press, New York, USA, p. 13-20, 56, 79.

67. Mork, O, 1928. *"Die Qualität des Fichtenholzes unter Besonderer Rücksichtnahme auf Scleif- und Papierholz"*, Der Papierfabrikant (48), p. 741-747.

68. Fjerdingen, H., and Foreseth, T.F., 1997. *"Some Mechanical Pulp Fibre Characteristics, Their Process Relationship and Papermaking Significance"*, 11th Fund.Res.Symp. *"The Fundamentals of Papermaking Materials"*, Mechanical Engineering Publications, London, England, p. 547-605.

69. Mohlin, U.-B., 1995. *"Fibre Development during Mechanical Refining"*, Proc. Intl. Mech. Pulp. Conf., Ottawa, Canada, p. 71-77.

70. Ladell, J.L., 1971. *"Variation Outwards and some Internal Correlations in the Wood of Black Spruce, Picea mariana (Mill), B.S.P."*, Unpublished Report, Ontario Research Foundation, Mississauga, Ontario, Canada.

71. Szanyi, L., and Sugden E.A.N., 1967. *"The Relationship between Specific Gravity and the Scanned Solid Volume of Wood"*, Unpublished Report (RSR No.539), Columbia Cellulose Company Ltd., New Westminster, Canada.

72. Sugden, E.A.N., 1962. *"Douglas Fir Sulphate Pulp Strength as Affected by Percentage Summerwood"*. Unpublished B.S.F. Thesis, Univ. of B.C., Vancouver, Canada.

73. Smith, D.M., and Miller, R.B., 1964. *"Methods of Measuring and Estimating Tracheid Wall Thickness of Redwood"*, Tappi J. 7(10), p. 599-604.

74. Johansson, D., 1940. *"Über Früh- und Spätholz in Schwedischer Fichte und Kiefer und über ihren Einfluss auf die Eigenschaften von Sulfit- und Sulfatzellstoff"*, Holz Roh- u, Werkstoof 3, p. 73-78.

75. Kollmann, F.F.P., and Côté, W.A., Jr., 1968. *"Principles of Wood Science and Technology, I: Solid Wood"*, N.Y., Springer-Verlag, New York, p. 9-12.

76. Goldstein, I.S., and Gould, R.f., 1977. *"Wood Technology: Chemical Aspects"*, ACS Symposium Series 43, American Chemical Society, Washington, USA, p.1-23.

77. Fengel, D., 1969. *"Ultrastructure of Cellulose from Wood (1): Wood as the Basic Material for the Isolation of Cellulose"*, Wood Sci. Technol. 3(3), p. 203-217.

78. Gladstone, W.T., and Ifju, G., 1974. *"Some Influences of Wood Morphology on Kraft Pulping of Loblolly Pine"*, Proc. TAPPI Forest Biology Conf., Seattle, USA, p. 13-19.

79. Larson, P.R., 1966. *"Changes in Chemical Composition of Wood Cell Walls Associated with Age in Pinus resinosa Ait"*, Forest Products J. 16(4), p. 37-45.

80. Ritter, G.J., and Fleck, L.C. 1926. *"Chemistry of Wood IX. Springwood and Summerwood"*, Industrial Engineering Chemistry, 18, p. 608-609.

81. Von Byrd, L., Ellwood, E.L., Hitchings, R.G., and Barefoot, A.C., 1965. *"Wood Characteristics and Kraft Paper Properties of Four Selected Loblolly Pines"*, Forest Products J. 15(8), p. 313-320.

82. Wu, B.J., Stonecypher, R., and Browne, C., 1967. *"Lignification within Coniferous Growth Zones"*, Pulp Paper Can. 68, p.159-164.

83. Bisset, I.J.W., and Dadswell, H.E., 1949. *"The Variation of Fiber Length within One Tree of Eucalyptus Regnans F.V.M."*, Australian Forestry, 13(2):, p.86-96.

84. Ranby, B.G., 1961. *"The Fine Structure of Cellulose Fibrils in F. Bolam(ed), Fundamentals of Papermaking Fibers"*, First Tech. Section Paper and Board Makers Association, London, England.

85. Roelofsen, P.A., 1959. *"The Plant Cell Wall, vol. III, part 4, Encyclopaedia of Plant Anatomy"*, Gebrüder Borntraeger, Berlin-Nikolassee, Germany.

86. Back, E. L., and Salmén, N. L, 1982. *"Glass Transitions of Wood Components Hold Implications for Modeling and Pulping Processes"*, Tappi 65 (7) p. 107-110.

87. Cousins, W. J., 1978. *"Young's Modulus of Hemicellulose as Related to Moisture Content"*, Wood Sci. Technol. 12 (3), p. 161-167.

88. Haun, J.L., and Britt, K.W., 1970. *"Handbook of Pulp and Paper Technology"*, Van Nostrand Reinhold, New York, USA, p. 25-31.

89. Glasser, W.G., and Sarkanen, K.V., 1989. *"Lignin Properties and Materials"*, ACS Symp. Ser.397. American Chemical Society, Washington, DC, USA.

90. Karnis, A., 1993. *"The Mechanism of Fibre Development in Mechanical Pulping"*, Proc. Intl. Mech. Conf.,, Oslo, Norway, p. 268-293.

91. Smook, G.A., 1992. *"Handbook for Pulp & Paper Technologists"*, 2nd edition, Angus Wilde Publications Inc., Vancouver, Canada, p. 45-64.

92. Alami, R., Boileau, I., Harris, G., Lachaume, J., Karnis, A., Miles, K.B., and Roche, A., 1995. *"Evaluation of the Impact of Refining Intensity on Energy Reduction in Commercial Size Refiners: The Effect of Primary Stage Consistency"*, Proc. Intl. Mech. Conf. Ottawa, Canada, p. 203-212.

93. Strand, B., 1997. *"Quality Control of High Consistency Refiners"*, Proc. Intl. Mech. Conf., Stockholm, Sweden, p. 127.

94. Härkönen, E., and Tienvieri, T., 2001. *"Energy Saving in TMP Pulping"*, Proc. Intl. Mech. Pulp. Conf., p. 547-556.

95. Pere, J., Liukkonen, S., Siika-Aho, M., Gullichsen, J., and Viikari, L., 1996. *"Use of Purified Enzymes in Mechanical Pulping"*, Proc. TAPPI Pulp. Conf., Atlanta, USA, p. 693-696.

96. Petit-Conil M., de Choudens C., and Espilit T., 1998. *"Ozone in the Production of Softwood and Hardwood High-yield Pulps to Save Energy and Improved Quality"*, Nordic Pulp Paper Res. J. 13(1), p. 16-22.

97. Mao, C.B., Law, K.N., and Kokta, B.V., 2003. *"Effect of Sulfonation on the Compression Behaviour of Early- and Latewood"*, 89th Annual Meeting, Pulp and Paper Technical Association of Canada, Montreal, Canada, p. 17-22.

98. Htun, M., and Salmén, L., 1996. *"Importance of Physical and Chemical Properties of Wood for Efficient Energy Input in Mechanical Pulping"*, Wochenblatt für Papierfabrikation 124(6), p. 232-235.

99. Easterling, K.E., Harrysson, R., Gibson, L., and Ashby, M.F. 1982. *"On the Mechanics of Balsa and other Woods"*, Proc. Royal Society London A383, p. 31-41.

100. Dumail, J.F., and Salmén, L., 1997. *"Compression Behaviour of Wood at Large Plastic Deformations: Comparison between Water and Ethylene-glycol Saturated Wood"*, Holzforschung, 51(4), p. 296-302.

101. Tabarsa, T., and Chui, Y.H., 2000. *"Stress-strain Response of Wood under Radial Compressions. Part I. Test Method and Influences of Cellular Properties"*, Wood and Fiber Sci. 32(2), p. 144-152.

102. Hartler, N., and Nyren, J., 1969. *"Influences of Pulp Type and Post-treatment on the Compressive Force Required for Collapse"*. TAPPI STAP No.8, p. 265-270, discn. p. 271-277.

103. Alexander, S.D., Marton, R., and McGovern, S.D., 1968. *"Effect of Beating and Wet Pressing on Fibre and Sheet Properties (1): Individual Fibre Properties"*, Tappi. J., 51(6), p. 277-283.

104. McIntosh, D.C., and Uhig, L.O., 1968. *"Effect of Refining on Load-elongation Characteristics of Lobbly Pine Holocellulose and Unbleached Kraft fibres"*, Tappi. J. 51(6), p. 268-273.

105. Reme, R.A., Johnsen, P.O., and Helle, T., 1998. *"Fibre Characteristics of Some Mechanical Pulp Grades"*, Nordic Pulp Paper Res. J. 13(4), p. 263-268.

106. Kure, K.A., 1999. *"On the Relationship between Process Input Variables and Fibre Characteristics in Thermomechanical Pulping"*, Doctoral Thesis 45/99, Dept. Chem. Eng., Norwegian Univ. Sci.Tech., Norway.

107. Cowan, W.F., 1968. *"The Screening of Groundwood - A Reappraisal"*, Proc. Intl. Mech. Pulp.Conf., Atlanta, USA, p. T011.

108. Reme, P.A., Tufa, L.D., Helle, T., and Johnson, P.O., 1998. *"The Fibre Characteristics of Shive Initiating Web Rupture"*, Paper Physics Seminar, Vancouver, Canada, A4.

109. Rudie, A.W., Morra, J., St. Laurent, J.M., and Hickey, K.L., 1994. *"The Influences of Wood and Fibre Properties on Mechanical Pulping"*, Tappi. J. 77 (6), p. 86-90.

110. Ahlgren, P.A., and Olausson, J.A., 1975. *"Kraft Pulping of Pine Earlywood and Latewood"*, Paperi ja Puu 57(2), p. 57-59.

111. Gladstone, W.T., and Ifju, G., 1974. *"Some Influences of Wood Morphology on Kraft Pulping of Loblolly Pine"*, Proc.Tappi. Forest Biology Conf., Seattle, USA, p. 13-19.

112. Gladstone, W.T., 1969. *"Response of Early- and Latewood from Loblolly Pine to Kraft Pulping"*, Ph.D. Thesis. North Carolina State Univ. ,USA, p. 138.

113. Hasuika, M., 1973. *"Physical Properties and Macrostructure of Earlywood and Latewood Pulp Sheets"*, J. Japan Wood Res. Soc. (Mokuzai Gakkaishi) 19(11), p. 547-553.

114. Franklin, C.L., 1945. *"Preparing Thin Sections of Synthetic Resin and Wood Composites and a New Maceration Method for Wood"*, Nature, 155, p. 51-54.

115. Law, K.N., Kokta, B.V., and Mao, C.B., 2001. *"Fibre Morphology and Soda-sulphite Pulping of Switchgrass"*, Bioresource Technology, 77, p. 1-7.

116 Koran, Z., 1967. *"Electron Microscopy of Radial Tracheid Surface of Black Spruce Separated by Tensile Failure at Various Temperatures"*, Tappi J., 50(2), p. 60-67.

117. Koran, Z., 1968. *"Electron Microscopy of Tangential Tracheid Surface of Black Spruce Produced by Tensile Failure at Various Temperatures"*, Svensk Papperstidning, 71(17), p. 567-576.

118 Koran, Z., 1970. *"Surface Structure of Thermomechanical Pulp Fibres Studied by Electron Microscopy"*, Wood and fibre, 2(3), p.247-258.

119. Atack, D., 1972. *"Characterization of Pressurized Mechanical Pulps"*, Svensk Papperstidning, 75(3), p. 89-94.

120. Sabourin, M.J., Cort, J.B., Waller, A., Xu, E. and Boileau, I., 1996. *"Optimizing Residence Time, Temperature and Speed to Improve TMP Pulp Properties and Reduce Energy"*, Preprints Tech. Sect., PAPTAC, Montreal, Canada, Book B, p. B69-B79.

121. Fuglem, G., Kure, K.-A., Løbben, O.P. and Helle, T., 2000. *"Full Scale Comparison of TMP Fibre Properties – RTS and Conventional Conditions"*, Preprints Ann. Meet., PAPTAC, Montreal, Canada, Book C, p.C187-C191.

122. Sabourin, M., Vaughn, J., Frith, M., Lauritzen, J., Wisenman, N. and Fraser, T., 2002. *"Characterization of Paper Properties from Spruce and Pine Thermomechanical Pulps – Effect of Refining Intensity"*, Preprints Ann. Meet., PAPTAC Book C, p. C125-C134.

123. Sabourin, M., 2004. *"Operating Experience with New RTS Lines at Holmen Hallstavik"*, Pulp Paper Can., 105(9), p. 27-29.

124. Münster, H., and Dahlqvist, G., 1995. *"Operational Experience with the First Commercial High Speed Refiner at Perlen Papier AG Switzerland"*, Intl. Mech. Pulp. Conf., Ottawa, Canada, p.197-203.

125. Law, K.N., 2000, *"New Insight into Chip Refining"*, Appita J., 53(3), p. 393-397.

126. Meyers, J., and Nanko, H., 2005. *"Effects of Fines on the Fiber Length and Coarseness Values Measured by the Fiber Quality Analyzer (FQA)"*, TAPPI 2005 Spring Technical Conference, Atlanta, USA, p.1-8.

127. Robertson, G., Olson, J., Allen, P., Chan, B., and Seth, R., 2004. *"Measurement of Fibre Length, Coarseness, and Shape with the Fibre Quality"*, http://api.mech.ubc.ca/PDF/fqa_tappi.pdf (consulted on 6, December, 2005)

128. Bently, R.G., Scudamore, P., and Jack, J.S., 1994. *"A Comparison between Fibre Length Measurement Methods"*, Pulp & Paper Canada, 95(4), p.41-45.

129. Page, D.H., Seth, R.S., Jordan, B.D. and Barbe, M.C., 1985. *"Curl, Crimps, Kinks and Microcompressions in Pulp Fibers - Their Origin, Measurement and Significance"* Trans. 8th Fund. Res. Symp., Oxford, Sept. 1985,V. Punton, Ed., Mech. Eng. Publications Ltd., London, p. 183–227.

130. Law, K.N., Yang, K.C., and Valade, J.L., 1997. *"Fibre Development in Thermomechanical Pulping: Comparison between Black Spruce and Jack Pine"*, Preprints of CPPA Technical Section Annual Meeting, Montreal, Canada, 1997, p. B113-B127.

131. Seth, R.S., 2002. *"The Measurement and Significant of Fines"*, 88th PAPTAC Annual Meeting, Montreal, Canada, p. c97-c101.

132. Sirvio, J., and Nurminen, I., 2004. *"Systematic Changes in Paper Properties Caused by Fines"*, Pulp Pap Can 105(8), p. 39-42.

133. Marton, R., and Robie, J.D., 1969. *"Characterization of Mechanical Pulps by A Settling Technique"*, Tappi. J., 52(12), p. 2400-2406.

134. Luukko, K., 1999. *"Fine Quantity and Quality in Controlling Pulp and Paper Quality"*, Intl. Mech. Pulp.Conf., Houston, USA, p.67-75.

135. Allender, B. M., and Waterhouse, J. F., 1986. *"Morphological Factors in the Refining of Eucalypt and Pinus Radiata Fibers"*, PIRA Int. Conf. New Technologies in Refining, Birmingham, England, Vol. 2, Session 3, Paper 8:, p. 22.

136. "Morfi Cell Wall Thickness Measurement", Website of Techpap, France, http://www.techpap.com/ (consulted on 4, Nov., 2005)

137. CMT, Appareil de mesure d'Épaisseur des parois, Manuel d'utilisation, Techpap 2005, France.

138. Reme, P.A., and Helle, T., 1999. *"Methods to Assess Detailed Cross-sectional Dimensions of Fibre Populations in Wood and Pulp Using SEM and Image Analysis, Microscopy as a Tool in Pulp and Paper Research and Development"*, A State of the Art Symposium, Concerted Action FAIR-98-3681, Stockholm, Sweden.

139. Website of Scalex Corporation, http://www.scalex.com/products.php#mapwheel (consulted on 6, December, 2006).

140. Cisneros, H.A., Williams, G.J., and Hatton, J.V., 1992. *"Fibre Surface Characteristics of Hardwood Refiner Pulps"*, TAPPI Pulp. Conf., Atlanta, USA, p. 1151-1160.

141. K.N., Law, J.J., Balatinecz, J.J., and Garceau, J.J., 1974. *"A Technique for Making Oriented Fibre Sheet"*, TAPPI, Vol. (57), No.12, p. 153-154.

142. M.J., Dykstra, 1993. *"A Manual of Applied Technical for Biological Electron Microscopy"*, Plenum Press., New York, USA, p. 38-42.

143. Reme, P.A., Johnsen, P.O., and Helle, T., 2002. *"Assessment of Fibres Transverse Dimensions Using SEM and Image Analysis"*, J. Pulp & Paper Sci., 28(4), p. 122-128.

144. Website of National Institute of Neurological Disorders and Stroke, USA , http://rsb.info.nih.gov/ij/ (consulted on Nov, 8, 2005)

145. Fjerdingen, H., and Houen, P.J., 1997. *"On the Effect of Recycling on Gross-sectional Shapes and Dimensions of Sulphate Pulp Fibres"*, Recycling Symposium, TAPPI, Atlanta, USA, p. 347-360.

146. Jang, H.F., and Seth, R.S., 1998. *"Characterization of the Collapse Behaviour of Papermaking Fibres Using Confocal Microscopy"*, 84th Annual Meeting, Technical Section CPPA, Atlanta, USA, p. 205-212.

147. Johnsen, P.O., Skinnarland, I., Helle, T., and Housen, P.J., 1995. *"Distribution of Lignin and Other Materials on Particles Surfaces in Mechanical Pulps"*, Proc. Intl. Mech. Pulp. Conf., Ottawa, Canada, p. 93-107.

148. Maxwell, M.H., 1978. *"Two Rapid and Simple Methods Used for the Removal of Resins from 1.0 μm T-epoxy Sections"*, J. Microscopy, 112(2), p. 253-255.

149. Williams, G.J., Drummond, J.G., and Cisneros, H.A., 1994. *"A Microscopic Approach for Examining Fibre and Paper Structure"*, J. Pulp & Paper, 20(4), p. J110-J114.

150. Carlsson, G., 1996. *"Surface Composition of Wood Pulp Fibres: Relevance to Wettability, Sorption and Adhesion"*, Department of Pulp and Paper Chemistry and Technology, Royal Institute of Technology and Institute for Surface Chemistry, Stockholm.

151. Österberg, M., 2000. *"On the Interactions in Cellulose Systems: Surface Forces and Adsorption"*, Department of Chemistry, Surface Chemistry, Royal Institute of Technology, Stockholm, Department of Forest Products Technology, Helsinki University of Technology, Helsinki, and Institute for Surface Chemistry.

152. Dorris, G.M., and Gray D.G.,1978. *"The Surface Analysis of Paper and Wood Fibers by ESCA (Electron Spectroscopy for Chemical Analysis)/I. Application to Cellulose and Lignin"*, Cellulose Chem. Technol, Vol.12, p. 9.

153. Dorris, G.M., and Gray D.G., 1978. *"The Surface Analysis of Paper and Wood Fibers by ESCA (Electron Spectroscopy for Chemical Analysis)/II. Surface Composition of Mechanical Pulps"*, Cellulose Chem. Technol, Vol.12, p.721.

154. Gray, D.G., 1978. *"The Surface Analysis of Paper and Wood Fibers by ESCA. III. Interpretation of Carbon (1s) Peak Shape"*, Cellulose Chem. Technol. Vol.12, p.735.

155. Hultén, A.H., and Paulsson, M., 2003. *"Surface Characterization of Unbleached and Oxygen Delignified Kraft Pulp Fibres"*, J. Wood Chem. Technol., 23(1), p. 31-46.

156. He, J.H., W.J., Batchelor, and R.E., Johnston, 2003. *"The Behaviour of Fibres in Wet Pressing"*, Tappi, J., 2(12), p.27-31.

157. Mohlin, U.B., Dahlbom, J., and Hornatowska, J., 1996. *"Fibre Deformation and Sheet Strength"*, Tappi J., 79(6), p. 105-111.

158. Stationwala, M.I., Mathieu, J., and Karnis, A., 1996. *"On the Interaction of Wood and Mechanical Pulping Equipment. Part I: Fibre Development and Generation of Fines"*, J. Pulp Paper Sci. 22(5), p. J155-J160.

159. Law, K.N., and Valade, J.L., 1994. *"The Status of the Utilisation of Jack Pine (Pinus banksiana) in the Pulp and Paper Industry"*, Canadian J. For. Res. 24, p. 2078-2084.

160. Jossart, D., Barbe, M.C., Lapointe, M., and Law, K.N., 1988. *"Properties of Mechanical and Chemi-Mechanical Jack Pine Pulps: Part I - Thermomechanical Pulps"*, Pulp Paper Can. 89(4), p. T115-T122.

161. Laliberté, D., Shallhorn, P.M., and Karnis, A., 1987. *"Comparison of TMP and CTMP Properties from Spruce and Pine Sawmill Chips"*, Pulp Paper Can., 88(3), p. 94-100.

162. Tyrväinen, J., Law, K.N., and Valade, J.L., 1997. *"Alkaline-Peroxide Inter-Stage Treated Mechanical Pulp from Jack Pine (Pinus banksiana), Part I. Introduction and Pulp Physical Properties"*, Pulp Paper Can., 98(6): T191-196.

163. Tyrväinen, J., Law, K.N., and Valade, J.L., 1997. *"Alkaline-Peroxide Inter-Stage Treated Mechanical Pulp from Jack Pine (Pinus banksiana), Part II. Pulp Optical Properties, Colour Reversion, Extractives Content and Process Implication"*, Pulp Paper Can., 98(7): T223-227.

164. Isenberg, I.H., 1980. *"Pulpwood of the United States and Canada, Vol.1-Conifers"*, The Institute of Paper Chemistry, Appleton Wis. USA.

165. Tay, C.H., and Manchester, D. F., 1982. *"Jack Pine as Pulpwood for Newsprint Manufacture. Part I. The Significance of fibre morphology and Extractives"*, Proceedings of the Canadian Wood Chemistry Symposium, Niagara Falls, p.119-114.

166. Beath, L.R., and Mihelich, W.G., 1977. *"Refiner Mechanical Pulping of chemically Pretreated Wood"*, Tappi.J., 60(12), p. 77-81.

167. Berzins, V. 1966. *"Chemical Composition of Woods"*, Pulp and Paper Research Institute of Canada, Pointe-Claire, Quebec, Canada, p.61.

168. Yuen, T.Y.C., 1999. *"Tree Gummies"*, Website of Econoteck, http://www.econotech.com/newsletters/007_fall_99.pdf (consulted on 20, January, 2006)

169. Allen, L.H., 1988. *"Pitch Control during the Production of Aspen Kraft Pulp"*, Pulp. Paper Can. 89(10), p. T342-T346.

170. Yemchuk, E.M., 1970. *"Oxidation of Black Liquor at Great Lakes Paper Co. Ltd"*, Pulp Paper Mag. Can., 71(14), p.T45-T50.

171. Rapson, W.H., Wayman, M., and Anderson, C.B., 1965. *"Hydrosulphite and Peroxide Bleaching of Nine Pure Species Groundwoods"*, Pulp Paper Mag. Can., 66(5), p.255-271.

172. Beath, L.R., Neill, M.T., and Masse, F.A., 1966. *"Latency in Mechanical Wood Pulps"*, Pulp & Paper Magazine of Canada, 67(10), p. T423-T430.

173. Karnis, A., 1993. *"Latency in Mechanical Pulp Fibres"*, Paperi Ja Puu, 75(7), p.505-511.

174. Harris, G., and Karris, A., 1986. *"Storage of Latent Mechanical Pulps"*, Journal of Pulp and Paper Science, 12(4), p. J100-J107.

175. Lunan, W.E., Sferrazza, M.J., Franzen, R.G., and May, W.D., 1986. *"Curl-Setting during Storage of Thermomechanical Pulp at High Consistency"*, Journal of Pulp and Paper Science, 12(4), p. J108-J115.

176. Page, D.H., Barbe, M.C., Seth, R.S., and Jordan, B.D., 1984. *"Mechanism of Curl Creation, Removal and Retention in Pulp Fibers"*, Journal of Pulp and Paper Science, 10(3), p. J74-J79.

177. Label, R.G., Nobleza, G.C., and Paquet, R. 1979. *"Water Retention Value Indicates Machine Runnability of Pulp"*, Pulp & Paper Canada, 80(5), p.T135-T140.

178. Law, K., Kokta, B.V., and Mao, C.B., 2006. *"Effect of Temperature on Compression Properties of Wood and Fibre Failures – A Problem Implication of Screw-pressing of Chips in TMP"*, PAPTAC 92nd Annual Meeting, Montreal, Canada, p. A429-A434.

179. Reme, P.A., Kure, K.-A., Gregersen, Ø., and Helle, T., 1999. *"Optimal Mechanical Pulp Fibres for Publication Paper: Targets and Treatments"*, Proc. Intl. Mechanical Pulping Conf., TAPPI PRESS, p.171–182.

180. Fjerdingen, H., Forseth, T., Gregersen, Ø., Helle, T., Johnsen, P.O., Kure, K.-A., and Reme, P.A., 1997. *"Some Mechanical Pulp Fibre Characteristics, Their Process Relationshipsand Papermaking Significance"*, Trans. 11th Fundamental Res. Symp., Fundamentals of Papermaking Materials, Cambridge,UK., p. 547–605.

181. Shallhorn, P.M., Karnis, A., 1979. *"Tear and Tensile Strength of Mechanical Pulps"*, Trans. Techn. Sect. (CPPA), 5(4), TR92-TR100.

182. Sabourin, M., Cort, J.B., Waller, A., Xu, E., and Boileua, I., 1996. *"Optimizing Residence Time, Temperature, and Speed to Improve TMP Pulp Properties and Reduce Energy"*, PAPTAC annual meeting, Montreal, Canada, p. B69-79.

183. Sabourin, M., Vaughn, J., Wiseman, N., and Welungoda, B., 2003. *"Industrial Scale Evaluation of Low Retention, High Temperature, High Speed (RTS) TMP Pulping of Southern Pine"*, APPITA Annual Gen. Conf. Proceedings, Melbourne, Australia, p. 119-126.

184. Lindholm C.A., 1980. *"Comparison of Some Papermaking Properties of Groundwood, Pressure Groundwood and Thermomechanical Pulp by Means of Pulp Fractions. (2) Fines Fractions"*, Paperi ja Puu, 62(12), p803-808.

185. Luukko, K., and Paulapuro, H., 1997. *"Mechanical Pulp Fines; Effect of Particle Size and Shape"*, 1997 Engineering & Papermakers: Forming Bonds for Better Papermaking Conference, TAPPI Press, p. 131-136.

186. Alince, B., Porrubská, J., and Vande Ven, T.G.M., 2001. *"Effect of Model and Fractionated TMP Fines on Sheet Properties"*, Proc. 12th Fundamental Research Symposium, Oxford, p. 1343-1355.

187. Norrström, H, 1969. *"Light-absorption Properties of Pulp and Pulp Components. (1). Method. (2). Sulfite Pulp"*, Svensk Papperstidning, 72(2), p.25-38.

188. L. T., Heikkurinen, 1997. *"Changes in Fibre Wall Structure during Defibration"*, 11th Fundamental Research Symposium in: the Fundamentals of Papermaking Materials, Cambridge, United Kingdom, p. 641-662.

189. Mohlin, U-B., 1980. *"Properties of TMP Fractions and Their Importance for Quality of Printing Papers. (1). Large Variations in Properties within Fractions"*, Svensk Papperstidning, 83(16), p.461-466.

190. Mohlin, U-B., 1980. *"Properties of TMP Fractions and Their Importance for Quality of Printing Papers. (2).Influence of Particle Properties and Particle Size Distribution on Pulp Properties"*, Svensk Papperstidning, 83(18), p.513-519.

191. Zha, Q. Q., and Lanouette, R., 2006, *"Co-refining Interfractions between White Birch, Hybrid Larch and Black Spruce"*, PAPTAC 92nd Annual Meeting, Montreal, Canada, p. A421-A426.

192. MacDonald, R.G., 1969. *"The Pulping of Wood"*, McGraw Hill Book Co., New York, USA, p.629.

193. Aravamuthan, R. G., Shriver, E. H., and Mishra, A. K. 1993, *"Effect of Refining on the Optical Properties of Softwood Thermo- and Chemithermomechanical Pulps"*, Cellul. Chem. Technol. 27, no. 5, p. 525-535.

194. Barrett, K., and Flora, G., 2000. *"Genetic Engineering and the Precautionary Principle"*, website of Northern Plains Sustainable Agriculture Society (NPSAS), www.npsas.org/GEPrecautionary.html (consulted on March 15, 2006)

195. Sykes M., Yang, V., Blankenburg, J., AbuBakr, S.,1999. *"Biotechnology: Working with Nature to Improve Forest Recourses and Products"*, Tappi International Environment Conference, Atlanta, GA., USA. p. 631-637.

196. Moffa, A., *"Genetic Engineering Turns to Trees"*, Science Vol.271, No. 5250, p. 761.

197. Tsai, C.J., Zhang, D. Y., Davis, M. F., Chiang, V.L., 2004. *"Genetic Augmentation of Syringyl Lignin in Low-lignin Aspen Trees"*, TAPPI Paper Summit - Spring Technical and International Environmental Conference. Atlanta, USA, p.363-366.

198. Sundberg, B., Tuominen, H., Nilsson, O., Moritz, T., Little, C., Sandberg, G., and Olsson, O., 1997. *"Growth and Development Alteration in Transgenic Populus [Poplar]: Status and Potential Applications - Micropropagation, Genetic Engineering, and Molecular Biology of Populus Poplar"*, Rocky Mountain Forest and Range Experiment Station, USA., p.74-83.

199. Nugent, H.M., Allen, L.H., and Bolker, H.I., 1977. *"Effect of Seasoning on the Acetone Extractives Composition of Wood from Black Spruce, Jack pine and Trembling Aspen"*, CPPA Trans. Tech. Sect. 3, no.4, Montreal Canada, p.103-109.

200. Tay, C. H., Fairchild, R. S., Manchester, D.F., 1983. *"Jack Pine as Pulpwood for Newsprint Manufacture. (2). Removal of Extractives During Ultra-High-Yield Pulping"*, Int. Symp. Wood & Pulping Chem. Vol. 2, Japan, p. 28-35.

201. Tay, C. H., Fairchild, R. S., Manchester, D.F., 1982. *"Jack Pine as Pulpwood for Newsprint Manufacture. (1). Significance of Fiber Morphology and Extractives"*, Can. Wood Chem. Symp. (Niagara Falls), Ontario, Canada, p. 109-114.

Appendix

- **"Effect of Primary Stage Refining Temperature on Fibre and Pulp Properties of Jack Pine TMP"**

This paper was presented at 92nd PAPTAC Annual Meeting, Montreal (Feb., 2006).

- **"Behaviour of Jack Pine Early- and Latewood in Refining"**

This poster was presented at 91st PAPTAC Annual Meeting, Montreal (Feb., 2005). It was also presented at the scientific poster competition of *Uiversité du Québec à Trois-Rivières* (March, 2005) and won the first prize in the papermaking division.

- **Micrograph: "Cross-section of early- and latewood TMP fibre"**

This micrograph was published as a cover image in *Journal of Pulp and Paper Science* (Vol.31, No. 1, 2006).

EFFECT OF PRIMARY STAGE REFINING TEMPERATURE ON FIBRE AND PAPER PROPERTIES OF JACK PINE TMP

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RÉSUMÉ
Pour vérifier la pertinence de raffiner à haute température les fibres de pin gris à parois épaisses, nous avons effectué des essais de raffinage à 120 et 160 °C en nous concentrant sur la morphologie des fibres et les propriétés des papiers produits. Pour un même indice d'égouttage, le raffinage à 160 °C nécessite 12 à 17% moins d'énergie spécifique. Malgré une épaisseur de paroi et une masse linéique des fibres longues plus élevées, les pâtes à haute température montrent des fines ayant une surface spécifique plus grande pour une même quantité. De plus, il n'y a pas de différence significative au niveau de l'indice de rupture ni de la rugosité des pâtes produites à ces deux températures.

ABSTRACT
This work deals with the question if it is beneficial to refine the thick-wall fibres of jack pine at elevated temperature in thermomechanical pulping. We conducted the refining at 120 and 160°C and studied the morphological characteristics of fibre and handsheet properties from the resulting pulps. Comparatively, for a given freeness, refining at 160°C required 12 to 17% less energy consumption. Despite its thicker cell wall and higher coarseness of long fibres the high-temperature pulp shows greater hydrodynamic specific volume for a given fines content. Additionally, there is no significant difference in tensile index and surface roughness between the pulps produced at these two temperatures.

KEYWORDS
Thermomechanical pulping, *Pinus banksiana*, fibre coarseness, cell wall thickness, physical properties, optical properties

INTRODUCTION

The common wisdom taught us that refining wood chips at elevated temperature well above the softening temperature of lignin results in fibre separation in the lignin-rich middle lamella [1-5]. Hence, mechanical pulping processes that use temperature above the lignin softening point produces fibres with smooth surfaces coated with lignin. Further mechanical treatment of these fibres requires higher energy consumption when compared to that of fibres produced at temperatures lower than the softening point of lignin [6].

It was shown that fibre properties of refiner pulps are determined by the conditions of the primary refining stage [7-9] and the secondary refining stage improves the pulp quality proportionally to the energy input. It was advisable that temperature lower than the lignin softening point be used in the first stage refining to prevent the production of smooth fibres. However, it had also been reported that the use of elevated temperature above the softening point of lignin in secondary refining or reject refining could reduce the refining energy requirement and improve the quality of the thick-walled long fibres [10-12]. This option is based on the principle that the higher temperature increases fibre flexibility and allow the fibre to be developed with improved papermaking characteristics more readily. The benefits of using high temperature in reject refining have also been reported recently [13, 14], in terms of fibre collapse and sheet surface smoothness.

On the other hand, higher temperature has been exploited in combination with high refining intensity during the first stage operation, which also leads to improved pulp quality and reduced energy, as reported by [15-18]. In this case, the real impact of high temperature on fibre separation mode is clouded by the high rotation speed used which should overwhelmingly determine the final fibre properties. However, a recent study [19] on high temperature primary refining of Canadian softwoods did not show any definite advantages of raising the refining temperature well above the softening point of lignin.

Amidst these conflicting views on the exploitation of elevated temperature in TMP, we initiated refining trials using a relatively low TMP temperature of 120 °C and an elevated temperature of 160 °C. Jack pine was used in these trials for its particularly high proportion of thick-walled latewood fibres. The main purpose was to see if high temperature refining could increase the collapsibility of the thick-wall fibres and improve the physical properties of handsheets.

EXPERIMENTAL

Material

Logs of freshly fell jack pine (*Pinus banksiana* Lam.) were used. The sample trees were about 25-30 years old. The logs were first debarked manually and cut into disks of about 2.5 cm thick in longitudinal direction. Chips were prepared manually using a chisel to separate the earlywood and latewood. The separation was based on the difference in color; the latewood in jack pine was relatively broad and much darker than the earlywood counterpart. The separation of earlywood from latewood permitted us to study the refining responses of the two types of wood within our global research framework. However, in this part of our research the earlywood and latewood chips were recombined according to the initial proportion and refined together. Due to the lengthy chip preparation it was necessary to air dry the chips for storage. However, they were rewetted prior to refining.

Refining

Pressurized refining at two temperatures, 120 and 160 °C, was performed using our pilot unit (Sunds Defibrator CD300 – Metso Inc.). The first stage pulp freeness was about 500 mL which was further reduced to several levels through atmospheric refining.

Fibre and Pulp Characterization

Pulp freeness and handsheet properties were determined following the appropriate PAPTAC standard methods while the fibre characteristics were measured using a Fiber Quality Analyzer (OpTest Equipment, Canada). Additionally, cell wall thickness was measured by means of a Cell Wall Thickness Analyzer (TECHPAP, France).

RESULTS AND DISCUSSION

Refining Energy

The increase in refining temperature from 120 to 160 °C had significantly reduced the energy requirement over a range of freeness from about 250 to 75 mL (Fig. 1). The corresponding energy ranged from about 9.5 to 12 MJ/kg; the savings in energy were approximately 12 to 17% when the first stage refining was raised from 120 to 160 °C. The drop in refining energy as a result of secondary refining at elevated temperature had been reported [9-11]. Albeit the use of high temperature in primary refining has been commercially exploited [12-15], the real contribution of high temperature to the reduction in refining energy is unclear because it is employed in combination with high intensity refining. However, the latter has in fact the most far-reaching impact on pulp quality compared with the former. In reality, the purpose of using high temperature in such a case is to flexibilize the fibres, minimizing fibre cutting in the high speed operation.

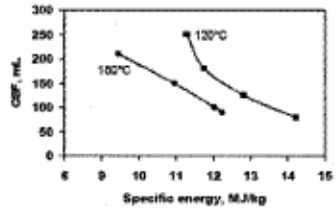

Figure 1. CSF vs specific refining energy.

Fibre Characteristics

The high temperature refining had different impact on fibre properties as compared to the low temperature operation. The high temperature (160 °C) produced coarser fibres (Fig. 2) because it is well above the lignin softening point, favouring fibre separation in the middle lamella. On the other hand, the lower temperature of 120 °C is expected to promote fibre separation in the S1 layer or at the S1/S2 interface [20]. With the subsequent second-stage atmospheric refining the fibre length and cell wall thickness decreased nearly in a linear manner, as revealed in Fig. 2. The trends were similar for both R28 and R48 Bauer McNett fractions. Similar relations between cell wall thickness and coarseness were also observed, as illustrated in Fig. 3. Most interestingly, Fig. 3 shows that the fibre development process was different depending on the initial defibration temperature; there was distinct trend for each fraction. This implies that the final pulp properties are dictated by the first stage refining conditions. Besides, the figure also reveals that there exists a close correlation between the two instruments measuring cell wall thickness and coarseness.

The primary fines produced with low energy consumption are comparable in terms of hydrodynamic specific volume (HSV) and quantity for both high and low refining temperatures (Fig. 4). However, the development of HSV during the subsequent second stages differed noticeably between the two temperatures. The high temperature refining would be more favourable in terms of fines quality development when compared to the low temperature process.

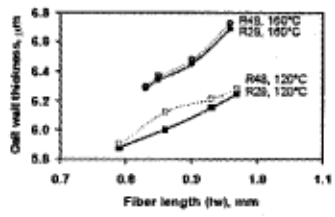

Figure 2. Cell wall thickness vs fiber length.

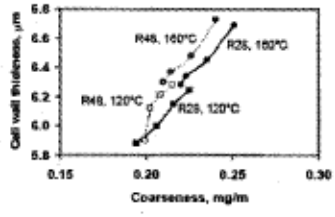

Figure 3. Cell wall thickness vs coarseness.

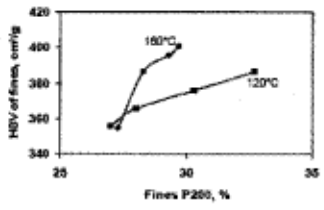

Figure 4. HSV of fines vs fines content.

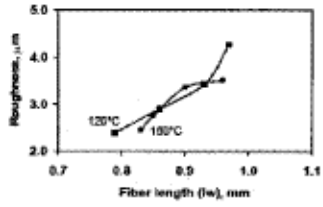

Figure 6. Roughness vs fiber length.

Handsheet Properties

Despite the seemingly unfavourable fibre characteristics mentioned earlier (example greater coarseness and cell wall thickness) the high temperature refining maintained the same tensile-sheet density relation when compared with the low temperature process (Fig. 5). This might imply that both processes produced fibres with similar collapsibility and conformability, which also leads to similar sheet surface smoothness (Fig. 6). It has been reported [12-14] that high temperature refining (secondary stage or rejects refining) could cause irreversible fibre collapse, which may explain why the coarser fibres produced at 160 °C yielded similar sheet density and roughness in comparison with those prepared at 120 °C.

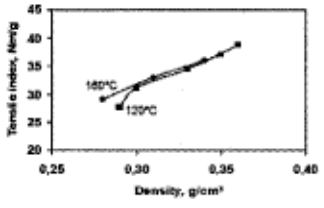

Figure 5. Tensile index vs density.

For a given tensile index, the low temperature (120 °C) pulp had higher light scattering coefficient when compared with high temperature (160 °C) one because the former had greater fines content (Fig. 7). The difference in light scattering coefficient between the pulps appeared to widen gradually with increasing refining energy or tensile strength. The difference might be also attributed to the fact noted earlier that the low-temperature fines had lower HSV as compared to the high-temperature fines.

The relation of tear index with the tensile index was quite different between the high and low temperature pulps, as shown in Fig. 8. The high temperature pulp maintained the same tear index of about 8.5 $mN*m^2/g$ until the tensile index reached approximately 36 N*m/g, after which its tear fell abruptly, probably due to the increased reduction in refiner plate clearance during the high temperature operation. As for the low temperature pulp, the development of tear index was more usual; it augmented sharply to a maximum of about 9 $mN*m^2/g$ and then dropped steeply at a tensile index of about 34 m*N/g. We suppose that, at low temperature, it is required to put more energy to reach the maximal tear development. On the other hand, at high temperature, the maximum tear index was reached at the early stage of refining. However, its magnitude was lower than that for the low temperature process.

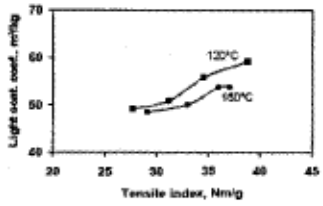

Figure 7. Light scattering coefficient vs tensile index.

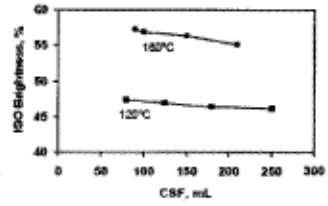

Figure 9. ISO Brightness of pulps.

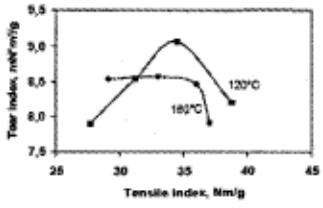

Figure 8. Tear vs tensile indexes.

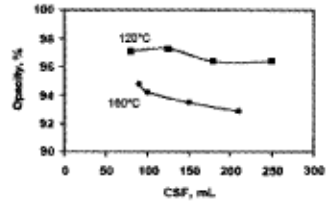

Figure 10. Opacity of pulps.

The greatest drawback of high temperature refining was a substantial drop in pulp brightness, nearly 10% point drop for the case of jack pine used in this investigation (Fig. 9). The particularly high extractives content of the species under study might have contributed to such a sharp fall in brightness. The decrease in initial pulp brightness would mean higher bleaching cost. As a result of the decreased brightness, the sheet opacity increased considerably, from 93 to 97% (Fig. 10).

CONCLUSION

The results of this study suggest that:

Raising the refining temperature from 120 to 160 °C in the primary refining is beneficial for jack pine TMP in terms of significant reduction in refining energy consumption, and comparable tensile index , surface characteristic and acceptable tear index.

High temperature primary refining of jack pine can incur substantial decrease in pulp brightness, increasing the cost of bleaching.

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REFERENCES

1. ASPLUND, A., "The origin and development of the Defibrator process", *Svensk Papperstidning*, 56(14):550, 1953.
2. KORAN, Z., "Electron microscopy of radial tracheid surface of black spruce separated by tensile failure at various temperatures", *Tappi J.*, 50(2): 60-67, 1967.
3. KORAN, Z., "Electron microscopy of tangential tracheid surface of black spruce produced by tensile failure at various temperatures", *Svensk Papperstidning*, 71(17): 567-576, 1968.
4. KORAN, Z., "Surface structure of thermomechanical pulp fibres studied by electron microscopy", *Wood and fibre*, 2(3):247- ? , 1970.
5. ATACK, D., "Characterization of pressurized mechanical pulps", *Svensk Papperstidning*, 75(3): 89-94, 1972.
6. ALLISON, R.W., "Effect of ozone on high-temperature thermomechanical pulp", *Appita J.*, 32(4):279-284, 1979.

7. STATIONWALA, M. and A. KARNIS, "Pulp grinding – a new method for producing mechanical pulp", *Tappi J.*, 73(12):187-195, 1990.
8. STATIONWALA, M., MILES, K.B. and A. KARNIS, "The effect of first stage refining conditions on pulp properties and energy consumption" *J. Pulp Paper Sci.*, 19(1):J12-J18, 1993.
9. HEIKKURINEN, N., VAARASALO, J. and A. KARNIS, "Effect of initial defiberization on the properties of refiner mechanical pulp", *J. Pulp Paper Sci.*, 19(3):J119-J124, 1993.
10. HÖGLUND, R., BÄCK, B., FALK, B. and M. JACKSON, "Thermopulp – a new energy-efficient mechanical pulping process" *Pulp Paper Can.*, 98(6):82-89, 1997.
11. JOHANSSON, O., FRITH, M., FALK, B. and R. GAREAU, "Thermopulp® - recent process developments and experiences", *Preprints 84th Ann. Meet. Tech.* Sect., PAPTAC, Book B:B241-B251, 1998.
12. NORGREN, S., HÖGLUND, R. and B. BÄCK, "Irreversible long fibre collapse at high temperature TMP reject refining – initial studies", *Pulp Paper Can.*, 105(7):47-51, 2004.
13. MUENSTER, H., FERRITSIUS, O., LECOURT, M. and M. PETIT-CONIL, "Energy savings in TMP by high temperature LC/MC refining", *Proc. Intl. Mech. Pulp. Conf.*, Oslo, 7-9 June: 213-223, 2005.
14. NORGREN, S. and H. HÖGLUND, "Irreversible long fibre collapse at high temperature TMP reject refining", *Intl. Mech. Pulp. Conf.*, Oslo, 7-9 June: 163-168, 2005.
15. SABOURIN, M.J., CORT, J.B., WALLER, A., XU, E. and I. BOILEAU, "Optimizing residence time, temperature and speed to improve TMP pulp properties and reduce energy", *Preprints Tech. Sect., PAPTAC*, Book B:B69-B79, 1996.
16. FUGLEM, G., KURE, K.-A., LØBBEN, O.P. and T. HELLE, "Full scale comparison of TMP fibre properties – RTS and conventional conditions", *Preprints Ann. Meet.*, PAPTAC Book C:C187-C191, 2000.
17. SABOURIN, M., VAUGHN, J., FRITH, M., LAURITZEN, J., WISEMAN, N. and T. FRASER, "Characterization of paper properties from spruce and pine thermomechanical pulps – effect of refining intensity", *Preprints Ann. Meet.*, PAPTAC Book C:C125-C134, 2002.
18. SABOURIN, M., "Operating experience with new RTS lines at Holmen Hallstavik", *Pulp Paper Can.*, 105(9):27-29, 2004.
19. OMHOLT, I., "Preheating and refining of mechanical pulp at high temperature", *Intl. Mech. Pulp. Conf.*, Oslo, 7-9 June: 59-62, 2005.
20. Law, K.N., and K.C. Yang, "Characteristics of fibre separation in wood loaded by normal-to-grain compression", *Appita J.*, 56(1):42-46, 2003.
21. SHALLHORN and P.M., KARNIS, "A. Tear and tensile strength of mechanical pulps", *Trans. Techn. Sect.*, PAPTAC, 5(4):TR92-00,1979.

Behavior of Jack pine early- and latewood in refining

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ABSTRACT

Thick-walled latewood fibers are known to have inferior papermaking properties relative to the thin-walled earlywood counterpart (Fig. 1). In this study we studied the refining behavior of early- and latewood of Jack pine under the same refining conditions by examining the breakdown of these two types of wood in pressurized refining.

INTRODUCTION

In comparison to spruce, Jack pine has higher proportions of latewood and its latewood fibers have thicker cell wall. As results, mechanical pulp such as thermomechanical pulp (TMP) of Jack pine has poor inter-fiber bonding, which means lower tensile and burst strengths. Due to their morphological differences early- and latewood fibers could behave differently in refining, in terms of cell wall rupture modes (inter-wall, trans-wall and intra-wall failures), cell wall thickness reduction, fiber breakage, cell wall splitting, surface fibrillation, etc. These differences in refining responses would have a significant impact on paper properties. Such information will be of great value for understanding the refining mechanism.

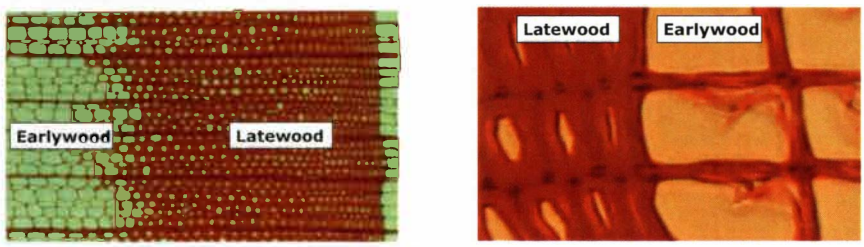

Fig.1 Cross-section view of early- and latewood

EXPERIMENTAL

Early- and latewood chips were refined separately using a Metso (Sunds Defibrator) CD300 pilot refiner. The chips were first pre-steamed for 10 min at atmospheric pressure and then pressed in a plug-screw with a 2:1 compression ratio. First-stage pulps of about 500 ml CSF were produced at 120° and 160 °C. The first-stage pulps were then refined atmospherically to different levels of freeness (50 – 250 mL). The pulps were fractionated in a Bauer McNett classifier to obtain R14, R28, R48, R100, R200 and P200 fractions which were used for microscopic analysis. Fibre length distribution, and physical and optical properties of standard handsheets were also determined. The experimental procedures are shown in Fig.2.

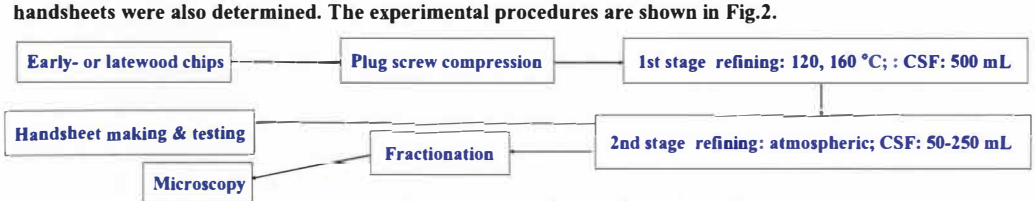

Fig. 2. Flow diagram showing the experimental procedure

RESULTS

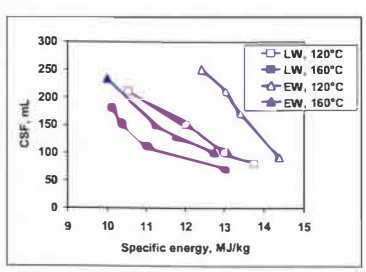

Fig. 3. Refining energy of early- and latewood

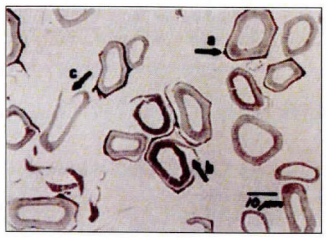

Fig.4: Cross-section view of TMP fibers

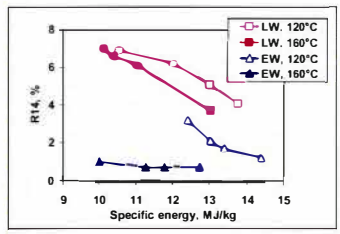

Fig. 5. R14 fraction of early- and latewood.

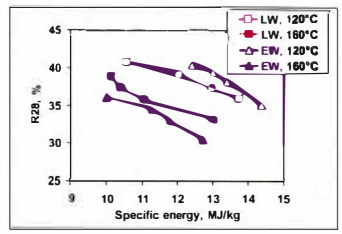

Fig. 6. R28 fraction of early- and latewood

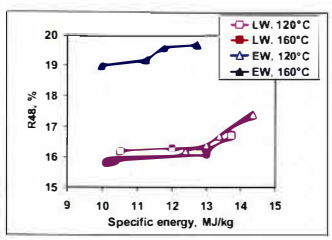

Fig. 7. R48 fraction of early- and latewood

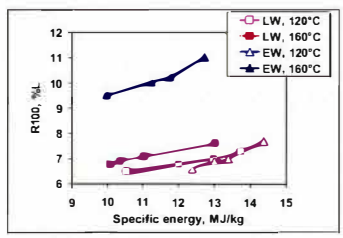

Fig. 8. R100 fraction of early- and latewood

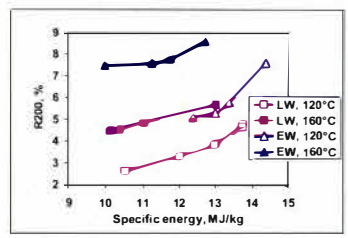

Fig. 9. R200 fraction of early- and latewood

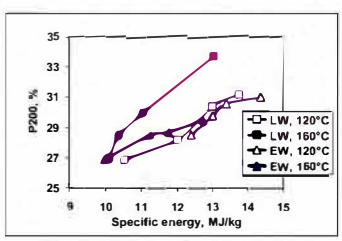

Fig. 10. P200 fraction of early- and latewood

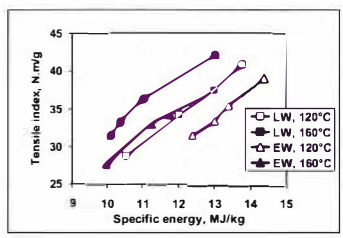

Fig. 11. Tensile index of early- and latewood

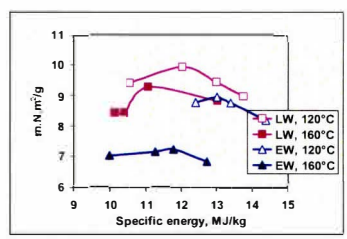

Fig. 12. Tear index of early- and latewood

CONCLUSION

1. For a given freeness earlywood required more specific refining energy than latewood did, despite the fact that there was no clear dissimilarity in fines generation between earlywood and latewood.

2. Regarding the refining behavior in terms of fiber fractions, there was no definite trend differentiating earlywood from latewood when they were refined separately. However, they might response differently when they are refined together. High temperature refining at 160 °C did yield significant higher proportion of earlywood fiber in the short fractions such as R48, R100 and R200.

3. Surprisingly, latewood pulp showed higher tensile strength compared to earlywood counterpart, for a given refining energy and temperature. Tear index was also higher for latewood than for earlywood, under similar refining conditions.

4. Cell wall delamination was more significant for latewood fibers than for earlywood ones.

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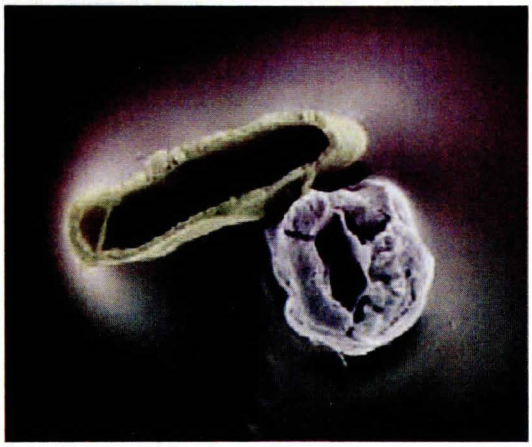

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