Faculty of Technology and Science
Chemical Engineering

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Strength Properties of Paper produced from Softwood Kraft Pulp

Pulp Mixture, Reinforcement and Sheet Stratification

DISSERTATION
Karlstad University Studies
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Bli pappersexpert på 5 minuter

Vad papper är

Kort och gott fibrer (förr ofta lump/tyg, nu trä) som finfördelats så grundligt att varje enskild fiber är skild från de andra. Sedan blandas de med vatten och trycks ihop, oftast i en vals. Voilà: ett papper är fött!

Vad papper inte är

Papyrus har inget med finfördelade fibrer att göra. Det tillverkas av papyrusgräs som skivas på längden och sedan limmas ihop. Rispapper karvas ur kung-shu-trädets märg i smala skivor. Pergament består av skrapade djurhudar.

Varde papper!

I Kina skrev man förr på bambu, som var tunt, och siden, som var dyrt. Ett alternativ behövdes. Enligt bevarade dokument uppfann Tsai Lun är 105 en metod att finfördela ”trädbark, hampa, trasigt tyg och fisknät för att göra chi”, det vill säga skrivunderlag. Dundersuccé på stört. På 800-talet berättade en kinesisk fånge för araberna om tillverkningshemligheten och papperets segertåg över världen kunde börja.

Runstenar kontra papper

Vikingarna meddelade sig med oss via runstenar, ofta intakta efter 1000 år eller mer. Runstenarna hade nackdelar: de var tunga och svårarbetade, inget praktiskt anteckningsblock direkt. Men de har en fördel: de står pall mot tidens tand. Ett ständigt problem med papper är dess flyktighet: det brinner lätt, gulnar, ruttnar, faller sönder. Papper är toppen på många sätt – men sätt ut det på en åker och kom igen om femhundra år…

Bildningsakuten, Göran Everdahl. ICA-KURIREN 42/03

Abstract

For paper producers, an understanding of the development of strength properties in the paper is of uttermost importance. Strong papers are important operators both in the traditional paper industry as well as in new fields of application, such as fibre-based packaging, furniture and light-weight building material. In the work reported in this thesis, three approaches to increasing paper strength were addressed: mixing different pulps, multilayering and reinforcement with man-made fibres. In specific:

The effects of mixing Swedish softwood kraft pulp with southern pine or with abaca (*Musa Textilis*) were investigated. Handsheets of a softwood kraft pulp with the addition of abaca fibres were made in a conventional sheet former. It was seen that the addition of abaca fibres increased the tearing resistance, fracture toughness, folding endurance and air permeance. Tensile strength, tensile stiffness and tensile energy absorption, however, decreased somewhat. Still it was possible to add up to about 60% abaca without any great loss in tensile strength. As an example, with the addition of 30% abaca, the tear index was increased by 36%, while the tensile index was decreased by 8%.

To study the effect of stratification, a handsheet former for the production of stratified sheets, the *LB Multilayer Handsheet Former* was evaluated. The advantage of this sheet former is that it forms a stratified sheet at low consistency giving a good ply bond. It was shown to produce sheets with good formation and the uniformity, evaluated as the variation of paper properties, was retained at a fairly constant level when the number of layers in the stratified sheets was increased. The uniformity of the sheets produced in the LB Multilayer Handsheet Former was generally at the same level as of those produced in conventional sheet formers.

The effects of placing southern pine and abaca in separate layers, rather than mixing them homogeneously with softwood pulp were studied. Homogeneous and stratified sheets composed of softwood and southern pine or softwood and abaca were produced in the LB Multilayer Handsheet Former. It was found that by stratifying a sheet, so that a pulp with a high tear index and a pulp with a high tensile index are placed in separate layers, it was possible to increase the tear index by approximately 25%, while the tensile index was decreased by 10-20%. Further, by mixing a pulp with less conformable fibres and no fines with a pulp with more flexible fibres and

fines, a synergy in tensile strength (greater strength than that predicted by linear mass fraction additivity) was obtained.

The effects of stratifying sheets composed of softwood and abaca were compared to the effects of refining the softwood pulp. Homogeneous and stratified sheets composed of softwood with three different dewatering resistances and abaca were also produced in the LB Multilayer Handsheet Former. It was found that by stratifying the sheets the tear index was retained while the tensile index was increased by the refining.

The effects of reinforcing softwood pulp with man-made fibres were also investigated. Man-made fibres (i.e. regenerated cellulose, polyester and glass fibres) were added in the amounts 1, 3, or 5% to softwood pulp of three different dewatering resistances. It was found that with refining of a softwood pulp and subsequent addition of long fibres with low bonding ability, the tensile-tear relationship can be shifted towards higher strength values. The bonding ability of the man-made fibres was evaluated by pull-out tests and the results indicated that, in relation to the fibre strength, regenerated cellulose (lyocell) was most firmly attached to the softwood network while the glass fibres were most loosely attached.

List of papers

The thesis is based on the following papers, which are referred to by their Roman numerals in the thesis:

I Karlsson, H., Beghello, L., Nilsson, L. and Stolpe, L. (2007):
Abaca as a reinforcement fibre for softwood pulp.
TAPPI JOURNAL, vol. 6, no 10, pp 25-32.

II Karlsson, H., Nilsson, L., Beghello, L., Stolpe, L. (2009):
Handsheet former for the production of stratified sheets.
Appita, vol. 62, no 4, pp 272-278

III Karlsson, H., Stolpe, L., Beghello, L. and Nilsson, L. (2010):
Paper strength evaluation both in stratified and in homogeneous sheets with selected fibres – Part I: Effect of fibre properties.
Submitted to TAPPI JOURNAL

IV Karlsson, H., Stolpe, L., Beghello, L. and Nilsson, L. (2010):
Paper strength evaluation both in stratified and in homogeneous sheets with selected fibres – Part II: Effect of refining.
Submitted to TAPPI JOURNAL

V Karlsson, H., Stolpe, L., Beghello, L. and Nilsson, L. (2010):
The effect on paper strength of man-made fibres added to a softwood kraft pulp.
Manuscript in preparation

Reprint of paper I and II has been made with permission from the publisher.

List of symbols and abbreviations

Symbols, Latin

A	Cross sectional area, in Page's equation
A	Crack area, for the calculation of energy release rate
b	Shear strength of fibre-fibre bond, in Page's equation
g	Acceleration due to gravity, in Page′s equation
G	Energy release rate
J	Calculated value of the J-integral
l_f	Fibre length, in the modified Page's equation
L	Fibre length, in Page's equation
P_{CD}	Value of any property measured in the cross machine direction of a sheet
P_{Geo}	Geometrical mean value of any property of an anisotropic sheet
P_i	Value of any property of pulp component i, for the calculation of properties for pulp mixtures
P_{MD}	Value of any property measured in the machine direction of a sheet
P_{Mix}	Value of any property for a pulp mixture
s	Arc length of the integration path round a crack, for the calculation of the J-integral
T	Tensile strength, in Page's equation
T_i	Traction vector, for the calculation of the J-integral
U	Potential energy, for the calculation of energy release rate
u_i	Displacement vector, for the calculation of the J-integral
w_f	Fibre width, in the modified Page's equation
w_i	Mass fraction of pulp component i, for the calculation of properties for pulp mixtures
Z	Zero-span tensile strength, in Page's equation

Symbols, Greek

Γ	Integration path round a crack, for the calculation of the J-integral
τ_f	Breaking stress of fibre-fibre bonds, in the modified Page's equation
ϖ	Strain energy density, for the calculation of the J-integral

Abbreviations

A	Abaca
A30	Sheet composed of abaca with a target grammage of 30 g/m²
A60	Sheet composed of abaca with a target grammage of 60 g/m²
Ad A	Prediction for a sheet composed of abaca and softwood according to the linear mass fraction additivity
Ad SP	Prediction for a sheet composed of southern pine and softwood according to the linear mass fraction additivity
CD	Cross machine direction of a sheet
CMC	Carboxymethyl cellulose, can be used as dry strength agent in paper
Comm 1-4	Commercial papers used in Paper II
HW	Hardwood pulp used in Paper II
IR	Infra-red radiation, used for drying on a paper machine
ISO	Isotropic sheets
LB	Luciano Beghello, originator of the LB Multilayer Handsheet Former
LEFM	Linear elastic fracture mechanics
MD	Machine direction of a sheet
NLFM	Non-linear fracture mechanics
OCC	Old corrugated container
PAM	Polyacrylamide, can be used as dry strength agent in paper
PET	Polyethylene terephtalate
PFI	Papir- och fiberinstituttet AS Paper and Fibre Research Institute, Trondheim, Norway
PQM	Pulp Quality Monitor
RBA	Relative bonded area
S	Softwood
S16.5	Softwood with a dewatering resistance of 16.5 SR
S24.0	Softwood with a dewatering resistance of 24.0 SR
S33.0	Softwood with a dewatering resistance of 33.0 SR

S30	Sheet composed of softwood with a target grammage of 30 g/m²
S60	Sheet composed of softwood with a target grammage of 60 g/m²
S/A	Stratified sheets of softwood and abaca
S+A	Homogeneous sheets of softwood and abaca
S/SP	Stratified sheets of softwood and southern pine
S+SP	Homogeneous sheets of softwood and southern pine
SP	Southern pine
SP30	Sheet composed of southern pine with a target grammage of 30 g/m²
SP60	Sheet composed of southern pine with a target grammage of 60 g/m²
SR	Schopper Riegler, unit for dewatering resistance of a pulp
STFI	Skogsindustrins Tekniska Forskningsinstitut Swedish Pulp and Paper Research Institute, Stockholm, Sweden. Current name: Innventia
SW1	Softwood pulp used in Paper II
SW2	Softwood pulp used in Paper II
TAD	Through-air drying
TEA	Tensile energy absorption
TMP	Thermomechanical pulp

Table of contents

1 Introduction

1.1 Background

The increasing competition on the world paper and board market inspires paper makers to continually develop and improve their products. One way to meet this competition is to increase paper strength properties. Strong papers are important not only in the current paper industry, they are also of interest for new fields of application of fibre-based products, such as packaging, furniture and light-weight building materials.

Long-fibre chemical pulp has long been used as a reinforcement in mechanical pulps, and softwood pulp is used as an additive to improve the strength properties of hardwood pulp, but the possibility of improving the strength properties of softwood chemical pulps with other types of pulps has not been investigated so thoroughly. There is much to be done to investigate other fibres with special properties with regard to length, strength, coarseness, stiffness and bonding potential from this point of view.

Multilayer forming techniques are commonly used in the manufacture of paper board where a low-cost, bulky mechanical pulp is placed in the middle layer and a chemical pulp with higher strength and better printing properties is placed in the outer layers. Multilayer forming is also widely used in tissue production where the middle layer is composed of a pulp with high bulk for good absorbency while the top layer is composed of a pulp with shorter fibres to give a smoother surface. Multilayer forming of paper products with a lower grammage than board, such as wood-free fine paper, is gaining some attention, but there is a need for further studies in this area to improve the strength properties and/or lower the cost of the raw material.

1.2 Objectives of this work

The objective of the work reported in this thesis was to examine the possibilities of increasing the overall strength of paper products. The work was divided into subprojects where the effects on paper strength of 1) mixing different pulps, 2) adding reinforcement fibres to a softwood pulp and 3) stratification, were examined.

In Paper I, the reinforcement potential of abaca (*Musa Textilis*) fibres for a softwood sulphate pulp was investigated.

In Paper II, the operating function of a multilayer handsheet former for the production of stratified sheets, i.e. the LB Multilayer Handsheet Former, was studied.

In Paper III, the question at issue was whether a more positive combined effect on paper strength properties can be achieved by placing fibres with special properties, i.e. high fibre length or coarseness, in separate layers rather than by making homogeneous sheets of a pulp mixture.

Paper IV is a continuation of Paper III where the effects on paper strength of stratification are compared with those of refining.

In Paper V, the objective was to examine whether the tearing resistance of a homogeneous sheet could be increased by addition of long fibres with low bonding ability while the tensile strength was increased by refining of the base pulp.

2 Papermaking

2.1 Paper production

The process of producing a paper consists of several steps;

- Production of pulp from a selected raw material
- Refining and other treatments of the pulp
- The addition of chemicals
- Forming the paper sheet on a wire
- Dewatering and consolidation to form a web
- Pressing
- Drying
- If necessary, surface sizing, coating and calendering

The most common raw materials for papermaking are vegetable fibres, mainly from wood, grass or cotton. Synthetic fibres are used in special papers. In Sweden, most of the total paper production is based on virgin fibres, although recycled fibres are used, mainly in the production of newsprint, tissue and corrugated board materials. There are several different methods for the production of pulp from the raw material and the pulps produced can be classified as mechanical, chemimechanical, semi-chemical and chemical pulps. Within these categories, there are several processes for pulp production. In Sweden, most of the mechanical pulp produced is thermomechanical pulp (TMP). The dominating cooking method for chemical pulp is the sulphate process, but the sulphite process and other cooking methods are also used to some extent.

In the refining of the pulp, the single fibres are subjected to a mechanical treatment, with the main purpose of improving the bonding ability. Chemicals and other additives can be added to the pulp to improve the runnability on the paper machine, to reduce the production cost or to give or enhance special properties of the end product.

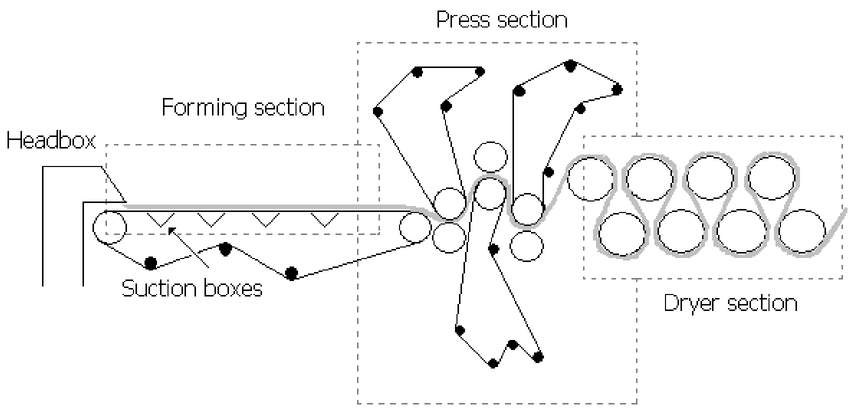


Figure 1. Schematic sketch of a fourdrinier paper machine. After the dryer section, the paper web may be subjected to surface sizing, coating and calendering before being transferred to a converting operation.

To start the forming process the pulp is distributed evenly onto the wire through a headbox, as shown in Figure 1. There are several designs of headboxes with different functions. In the forming section, the pulp is dewatered on a wire and a continuous web of fibres is formed. Three main types of paper machines exist where the forming section has different designs, the fourdrinier, the gap and the hybrid former. In the fourdrinier machine, the pulp is distributed onto a single wire and the water is removed by gravity, foils and vacuum applied in suction boxes. The gap and the hybrid formers are twin-wire machines where the dewatering is two-sided.

In the press section, the paper web is pressed against one or between two press felts through several roll nips and the dry solids content is increased. The paper web is then transferred to the drying section. The most common way of drying the paper web is by passing the web over a number of dryer cylinders heated by steam. Other drying methods that are used are convection drying, where hot air is blown onto the paper web, through-air drying (TAD), where hot air is pulled through the sheet by vacuum, and infra-red (IR) drying.

Sizing is used to make the fibres or the paper surface more hydrophobic. A coating may be applied to the paper to provide a smooth surface, to improve optical properties and to give a good printing surface. During calendering, the paper web is led through several nips to smoothen the surface and to give a higher gloss.

2.2 Multilayer forming on the paper machine

The technique of multilayer forming in paper production started in 1830 with the addition of a second forming cylinder to John Dickinson's cylinder machine (Attwood 1998). Figure 2 shows the principle of operation of the two-cylinder machine. Multiply paperboard has been produced continuously for almost 150 years (Attwood 1980). At first, the main target was to increase the speed at which heavyweight paperboard products were produced. Nowadays, multilayer forming is also used to produce paper products with improved or maintained stiffness properties at a lower grammage, to improve the fibre economy by placing a low-cost pulp in the middle layer, to improve surface properties by placing a high-quality pulp in the top layer, and to produce products with special properties by placing selected pulps in the different layers.

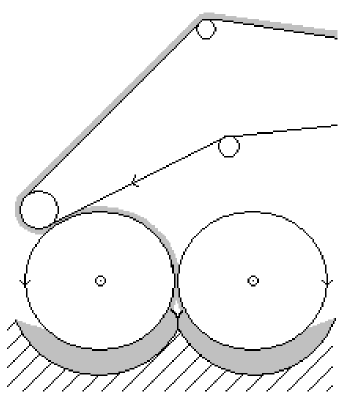


Figure 2. Operating principle of an early two-cylinder paper machine. (Redrawn from Attwood 1998).

Multilayer papers are produced by forming separate layers and combining them into one web, by forming a new layer on a pre-formed layer or by simultaneously forming a web in a multilayer headbox. The bond between the layers, i.e. the ply bond, is important for the properties of the end product. Critical parameters for the ply bond strength when combining different layers together are the dry solids content and the amount of fines at the interface. Several design concepts are used for the successive forming of layers. For example, the first layer can be formed on a fourdrinier wire, with the subsequent layers being formed in minifourdriniers with separate headboxes and wire sections, as shown in Figure 3. In a multilayer headbox, the different pulps are distributed through channels separated by vanes, as shown in Figure 4. Paper produced in a multilayer headbox is referred to as stratified paper, in contrast to multiply or multilayer paper.

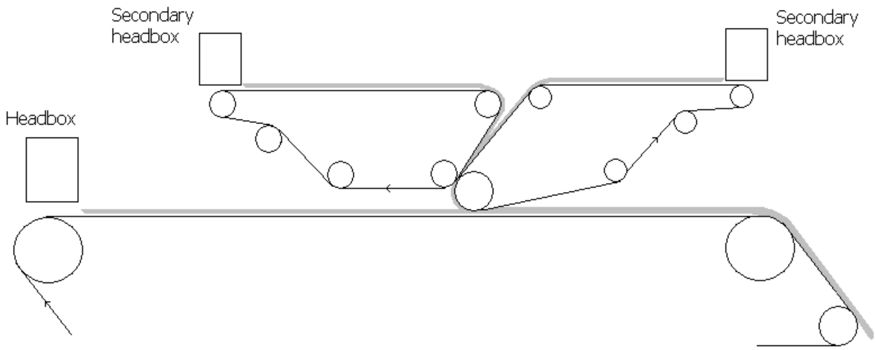


Figure 3. Schematic sketch of a machine concept for multilayer forming: A fourdrinier with two top minifourdriniers. (Redrawn from Foulger 1998).

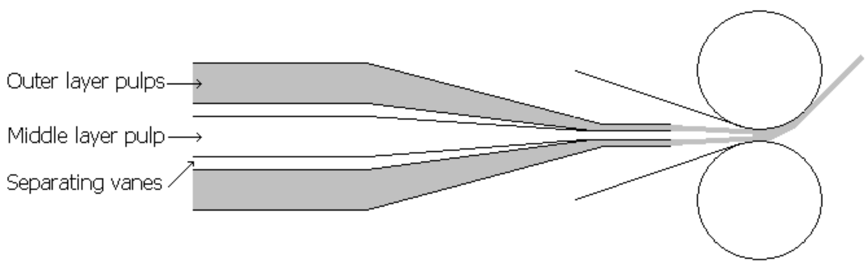


Figure 4. Schematic sketch of a multilayer headbox for stratified forming in a gap former. (Redrawn from Page and Hergert 1989).

2.3 Production of multilayer handsheets

Over the years, different approaches to the production of multilayer handsheets have resulted in a number of formers with their own advantages and disadvantages.

Stöckmann (1974a) modified a conventional isotropic handsheet former with a gate that divides the pulp container into two compartments. Figure 5 shows a sketch explaining the function of the sheet former. The lower compartment is filled with the bottom-layer pulp. The gate is then closed and the second-layer pulp is added. When the gate is opened permitting the sheet former to drain, the bottom layer is formed on the wire. The dewatering is interrupted when the water level reaches the gate. The gate is then again closed and a third pulp may be added. When the gate is opened to continue the dewatering, the second layer is formed on top of the previous layer and drained through it. Additional layers can be added by repeating the procedure. Gentle stirring during the dewatering prevents flocculation and provides some mixing between the layers enhancing the ply bond strength. Stöckmann's sheet former concept was used in several studies (Stöckmann 1974a; Stöckmann 1974b, Bergström and Peel 1979; Erickson 1977).

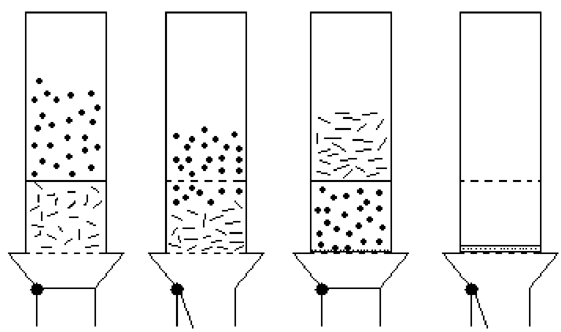


Figure 5. Schematic sketch of the function of a modified handsheet former. Dots and lines represent different pulps. (Redrawn from Stöckmann 1974a).

Bergström and Peel (1979) introduced a slightly different function in the sheet former. To separate the pulps, two plates with drilled holes were used, and one of the plates could be rotated relative to the other. When the holes are lined up, the gate may be moved through the pulp container and when the plate is rotated the gate is closed to flow. Once a layer is dewatered, the plates are lowered and closed and the next pulp is added. The gate is then opened and slowly elevated, and the next layer is dewatered. To hold the formed layers on the wire during the forming of subsequent layers, a valve is opened to permit a small flow through the fibre mat.

Another modification of the conventional isotropic handsheet former, the Pira Plug Flow Former, has been described by Moore (1998). The pulp is added at the top of the former in the ordinary manner. Jets with water and air create a turbulent area in the top of the sheet former giving an uniform dispersion of the pulp. Below the turbulent area, at the base of the former, the pulp is allowed to settle evenly, creating a sheet with good formation. Pulp for additional layers may then be added and the degree of interlayer mixing is controlled by the time interval between the pulp additions.

Puurtinen et al. (2003) chose a different approach when developing a multilayer handsheet former, as illustrated in Figure 6. The main difference from the previously described methods is the two-sided dewatering. The pulp container can be divided by plates into two or three compartments. Movable fabric frames are mounted vertically at the two ends of the chamber. The frames are pushed towards each other and the two outer layers are dewatered. When the frames approach the dividing plates, the plates are removed. The frames are then pushed further towards each other and the middle layer is dewatered through the two outer layers.

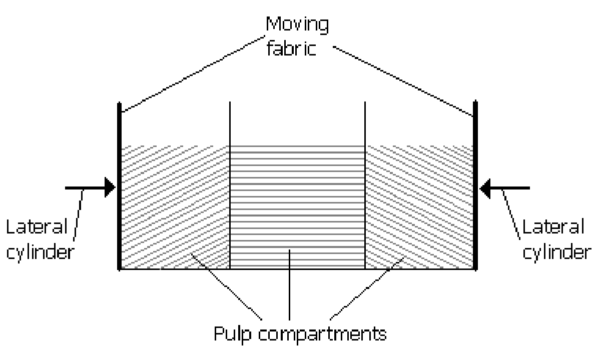


Figure 6. Function of multilayer handsheet former. (Redrawn from Puurtinen et al. 2003).

The dynamic sheet former Formette Dynamique (Sauret 1971), which produces anisotropic handsheets, can be used to produce multilayer sheets. The pulps are added consecutively in the pulp container, or two separate pulp containers can be used alternately. The added layers are dewatered through the previously formed layers. Multilayering in the Formette Dynamique has been reported in studies on both chemical and mechanical pulp (Bristow and Pauler 1983; Tubek-Lindblom and Salmén 2003; Nesbakk and Helle 2003).

2.3.1 LB Multilayer Handsheet Former, Paper II

In the present work, a multilayer handsheet former for the production of stratified sheets, originally developed at Åbo Akademi (Beghello et al. 1996), was used. In Paper II, this LB Multilayer Handsheet Former, Figure 7, is presented and evaluated. This former produces isotropic sheets and is easy to handle. The LB Multilayer Handsheet former can be used to produce single-layer sheets and stratified sheets with two, three or four layers. Figure 8 shows the basic design. The pulp container can be divided into two, three or four compartments using sliding plates, which are moved manually. Each compartment is equipped with a propeller to stir the pulp. The bottom compartment can be filled with water from below, while the top three compartments are top-filled. Each compartment has a volume of 15 dm^3 and the area of the sheets produced is 230 by 310 mm. The former is drained downwards and the wire aperture is chosen according to the International Standard for the preparation of laboratory sheets in a conventional sheet former, ISO 5269-1:05. The sheet production followed this standard as closely as possible. The bottom-layer pulp is added at the top of the former and diluted with water to a level just above the first divider plate, which is pushed into the former to seal the bottom compartment. The divider plate should be inserted when some of the fibre-water mixture from the bottom compartment is in the second compartment, to avoid air bubbles in the bottom compartment. The second-layer pulp is added from the top of the sheet former and diluted with water to a level just above the next divider plate. For further layers, the procedure is repeated. When the desired number of compartments have been filled with pulp and the stirring has been switched off, the divider plates are firmly pulled out in succession from the top to the bottom and the valve to the drainage pipe is opened. The sheets produced can be wet-pressed and dried under standardised conditions.

Figure 7. LB Multilayer Handsheet Former.

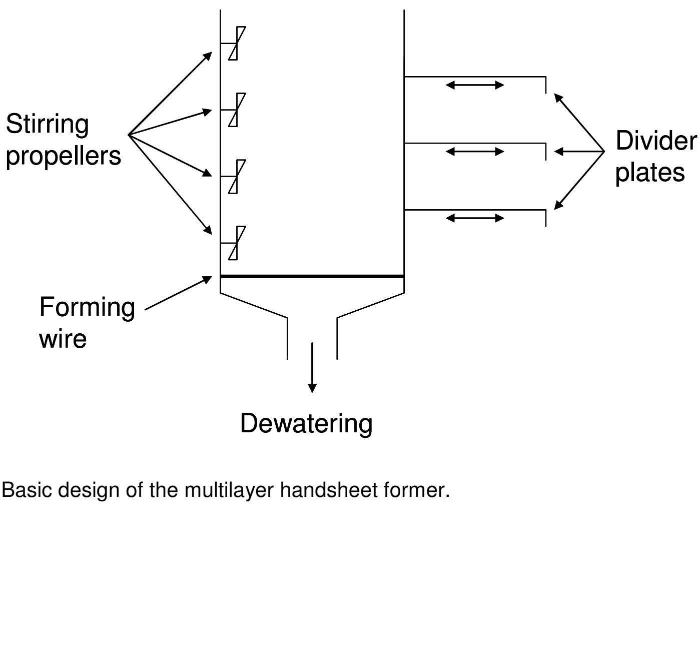


Figure 8. Basic design of the multilayer handsheet former.

3 Fibre and fibre network properties

3.1 Fibre strength

Several chemical and physical properties of the fibre affect the strength of the paper. Strength of single fibres is one of those properties and it can be measured directly on single fibres or indirectly on sheets. The two different approaches are described briefly in this section.

The fibre strength can be evaluated from tensile tests of single fibres. This method usually gives results with a large variation between fibres due to variations in the morphological properties of the fibres, and it requires a large number of fibres to give a meaningful average. Also, care has to be taken to ensure a representative selection of fibres. Due to the difficulties of measurement on single fibres, the zero-span tensile strength of paper is widely used as an indirect measurement of fibre strength. However, the fibres need to be straightened before sheet preparation since deformations in the fibres affect the zero-span tensile strength (Mohlin and Alfredsson 1990, Mohlin et al. 1996, Seth and Chan 1999, Seth 2001). Fibres can be straightened by refining in a PFI mill (Seth and Chan 1999) or by careful laboratory cooking (Seth 2001). The zero-span tensile strength is also affected by grammage: a higher grammage reduces the zero-span strength because of stress gradients in the thickness direction (Batchelor et al. 2006). Batchelor et al. thus suggest that, when using zero-span as a comparsion of fibre strength between different fibre types, the sheets evaluated should be of the same, low grammage.

In the present work fibre strength of the man-made fibres used in Paper V was evaluated from tensile tests curves of single fibres. For the pulp fibres used in Paper I and III-V the fibre strength was evaluated from zero-span tensile strength. The zero-span strength was evaluated on handsheets produced from the pulps without preceding straightening of the fibres.

3.2 Fibre network build-up

During the pressing of the paper web in the press section of the paper machine, the dry solids content is increased from about 20% to 40-55%. Fibres and fines are drawn towards each other by the capillary forces created by the surface tension in the liquid meniscus formed at the contact area between two fibres. Mechanical entanglement of fibrils also pulls fibres closer together and fines give an increase in the capillary forces and lead to a stronger network. Flexible fibres more easily conform to each other and give a larger fibre-fibre contact area.

Hydrogen bonds are generally believed to be the dominating force that builds up a paper. Dispersion forces between the fibres and van den Waals′ forces are other factors that slightly contribute to the strength. The hydrogen bonds between cellulose fibres are mainly formed between hydroxylic groups where a hydrogen atom oscillates between two oxygen atoms. Hydrogen bonds exist between glucose units in the cellulose molecule, between fibrils in the fibre wall, and between fibres in the fibre network (Retulainen et al. 1998).

3.3 Bond strength

Bond strength is a property of the fibre network which it is not easy to determine. The specific bond strength (SBS) is the ratio of the bond strength to the bond area. The methods for determining bond strength or specific bond strength are based on measurements on either single fibre bonds or on paper. The measurement of the strength of a single fibre bond is tedious, both because it requires careful precision to form and measure a bond, and because the great variation among fibre and bond properties demands that a large number of bonds be tested. Measurements on paper have the advantage that a large number of bonds are represented in each test, the bond structure is that resulting from the forming process and is not constructed artificially, and the measurement methods are rational. However, the sheet structure affects the loading behaviour to a great extent and the bond strength cannot be separated from the network strength. Thus the result of a measurement of specific bond strength is valid only for the specific structure and loading mode used (Retulainen 1997).

Mayhood et al. (1962) were among of the first to measure the shear bond strength of single fibre crosses. They produced fibre crosses by standard handsheet preparation using an extremely low concentration of the fibre suspension. The bonded area was determined by measuring the area of optical contact with a polarizing microscope. The fibre crosses were mounted on a specimen holder using strain-gauge cement, the lower fibre being attached to a stationary mount and the upper to a moving mount. The specimen holder was placed in a chainomatic balance and the load at shear failure was recorded. Mayhood et al. measured fibre crosses from different pulps but, due to the large deviation, no significant difference could be seen in the shear stress of the fibre crosses from different pulps.

At the same time, Schniewind et al. (1964), independently developed another method for measuring the shear strength of fibre-fibre bonds. They produced fibre crosses by placing two wet fibres at right angles on top of each other on a Teflon disk. Another Teflon disk was placed on top and the fibre crosses were dried under applied pressure. The bonded area was evaluated by microscopic determination of the size of the fibre overlap area. The fibre crosses were mounted on a paper strip with a pre-cut slot. One of the fibres was placed across the slot and its ends attached to the paper strip by double-sided tape. The other fibre was aligned with the slot having one free end protruding beyond the strip. The paper strip with the fibre cross was mounted in a tensile tester so that the paper strip was attached to one of the clamps and the free fibre end of the top fibre in the fibre cross was attached to the other clamp. Schneiwind et al. also reported large deviations in the results, but they concluded that the bond strength between latewood fibres was higher than that between earlywood fibres and that the strength of a bond between one latewood fibre and one earlywood fibre was intermediate.

Stratton and Colson (1990) evaluated the bond strength of single fibre bonds by measurement of the breaking load of fibre crosses. Their method resembled that of Schniewind et al. The fibre crosses were produced by placing two fibres perpendicular to each other on a silicone rubber plate. The fibre pair was dried in an oven under compression and then attached to a Mylar mount as shown in Figure 9. The bonded area was determined with light scattering and the breaking load of the bond was recorded in a tensile testing device. The bond strength was then calculated by dividing the load by the bonded area. Since the fibres were free to rotate, both shear and peel were present in the bond rupture process. A large standard deviation was observed for the breaking load and it was suggested that this was due to variations in the properties of the fibres themselves as well as in the bonds. Like Schneiwind et al., Stratton and Colson also reported a higher breaking load for latewood than for earlywood. They also investigated the effect of refining but saw no effect within the interval studied.

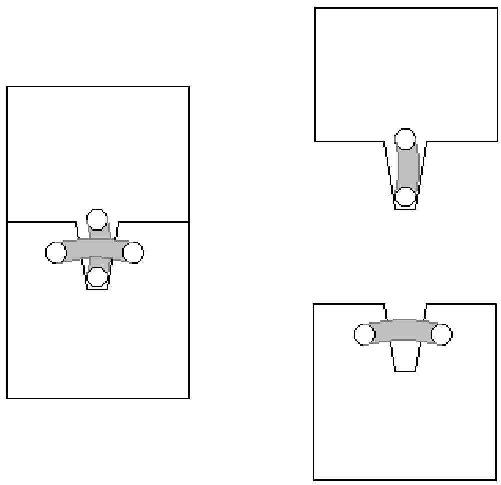


Figure 9. Fibre cross before and after straining the bond to failure. (Redrawn from Stratton and Colson 1990).

Torgnysdotter and Wågberg (2003) used the technique of Stratton and Colson to produce fibre crosses of chemically modified viscose fibres with different cross sectional areas. The regenerated fibres were used in order to reduce the variations in fibre surface morphology in order to reduce the standard deviation of the measurements. For 25 measured fibre crosses the variation coefficient was around 20%. Torgnysdotter and Wågberg found that the bond strength increased with increasing surface charge of the fibres.

Davison (1972) used a different approach for determining the bond strength. Using a tensile tester, fibres protruding from the torn edge of standard handsheets were pulled out from the fibre network. Davison compared sheets having two different levels of tensile strength and observed that, in the case of the sheets of higher tensile strength, a larger fraction of the tested fibres were broken rather than being pulled out intact compared to the situation when testing fibres in sheets of lower tensile strength. Davison discussed three types of typical stress-strain curves, see Figure 10 Curve *a* shows the curve for a fibre being pulled out intact, curve *b* represents a fibre resisting the pull-out at first but then being pulled out intact, and curve *c* is that of a fibre that broke during the pull-out.

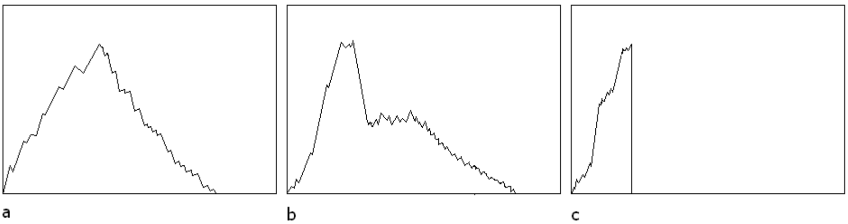


Figure 10. Typical stress-strain curves for fibres pulled out of handsheets. (Freely from Davison 1972).

Yan and Kortshot (1997) also used single fibre pull-out to compare the energy required for fibre pull-out and for fibre fracture. They made two slits in a paper strip and uncovered single fibres in the fibre network. The paper strip was mounted in a tensile tester and the load-elongation curves obtained. In the case of fibre pull-out, their observations were similar to those of Davison; first an initial force increase up to a critical value at which was a significant drop in the force, indicating debonding followed by a tail where the force gradually decreased to zero due to frictional forces. Yan and Kortshot reported that the energy required to initiate fibre debonding was lower than that required to break the fibre.

Page (1969) proposed a model for tensile strength that can be used to calculate the fibre-fibre bond strength. The method is based on the measurement of relative bonded area, RBA, and fibre properties. Page's equation for tensile strength is:

$$\frac{1}{T} = \frac{9}{8Z} + \frac{12A\rho g}{bPLRBA} \qquad (1)$$

where T is the tensile strength expressed as the now obsolete term breaking length, Z is the zero-span tensile strength of paper expressed as breaking length, A is the mean fibre cross-sectional area, ρ is the fibre density, g is the acceleration due to gravity, b is the shear strength per unit area of the fibre-fibre bonds, P is the perimeter of the average fibre cross section, L is the mean fibre length and RBA is the relative bonded area. A modified version of

Page's equation, where the tensile strength is given as force per cross-sectional area (Niskanen and Kärenlampi 1998), can be written:

$$\frac{1}{T} = \frac{9}{8Z} + \frac{3w_f}{\tau_f l_f RBA} \qquad (2)$$

where T is the tensile strength, Z is the zero-span tensile strength of paper, w_f is the fibre width, τ_f is the breaking stress of bonds, l_f is the fibre length and *RBA* is the relative bonded area. Paavilainen (1994) studied the impact of morphological properties of softwood sulphate pulp fibres on the bonding potential, evaluating the bond strength by Page's equation. Paavilainen argues that the bonding potential is determined by the bonded area, which in turn is controlled by the cell wall thickness of the fibres. She saw an increase in bond strength with increased refining of the pulp and suggested that the formation of fines together with increased fibre conformability during the refining, and thus an increase in RBA, explained the increase in the calculated bond strength. The observation of increasing bond strength with increasing refining is in agreement with the findings of Retulainen and Ebeling (1993), who also obtained higher bond strengths for latewood than for earlywood when using the Page equation.

Retulainen and Ebeling compared different ways of determining the bond strength from measurements on paper. Eight methods were studied; Nordman's bonding strength, Skowronski's SBS, Clark's cohesiveness, SBS from the Page tensile strength equation, SBS calculated from pull-out length, tensile strength at constant density level, Scott Bond divided by RBA and drying stress divided by RBA. The RBA was determined by the light-scattering method of Ingmanson and Thode (1959). They concluded that three factors lead to discrepancies between the different methods: a) the method of measurement of bonded area, b) the method of measurement of strength; force or energy, and c) the loading mode during the strength measurement; in-plane or z-directional (thickness). Koubaa and Koran (1995) compared the z-directional tensile strength test, the delamination test and the Scott-Bond test and suggested that the z-directional tensile strength test was the most suitable method among those for measuring bond strength. Section 4.7 further discusses the measurement of the internal bond strength in paper.

3.3.1 Pull-out tests used in Paper V

In the present work, the bonding potential of the man-made fibres used in Paper V were evaluated with pull-out tests. Long man-made fibres were pulled out from handsheets produced from softwood pulp. A 19 mm wide tape was placed across the center of the wire screen in the sheet former. Long fibres were then placed across the tape, as shown in Figure 11a. Pulp of softwood was added to the sheet former and a sheet was formed. The tape prevented fibres from forming a network in the center of the wire screen and the resulting sheet was in two parts, held together by the long fibres. Strips for tensile testing were cut from the sheet, as shown in Figure 11b. The man-made fibre was cut 10 mm into the upper part of the strip. The strip was mounted in a tensile tester so that the lower set of clamps held both the sheet and the fibre, while the upper set of clamps held only the sheet. The distance between the clamps was 50 mm. The force-displacement curve was then recorded at a constant elongation rate of 3 mm/min. Typical force-displacement curves for glass fibres, polyester and lyocell fibres being pulled out from handsheets produced from softwood pulp are shown in Figure 12. It can be seen that the fibres resist the force at first. The polyester and the glass fibres are then pulled out intact. The lyocell fibre however breaks. The area under the curves, i.e. the energy required to pullout the fibre, was used as an indication of the bonding potential of the fibres. It was thus concluded that, in relation to the individual fibre strength, the glass fibres have lowest bonding potential while the lyocell fibres have highest bonding potential.

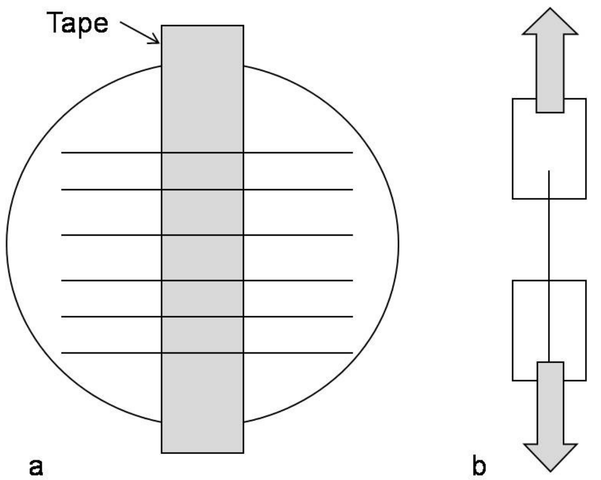


Figure 11. a: Handsheet former with a tape across the wire screen and long man-made fibres placed across the tape, b: Tensile test strip from the handsheets including one man-made fibre.

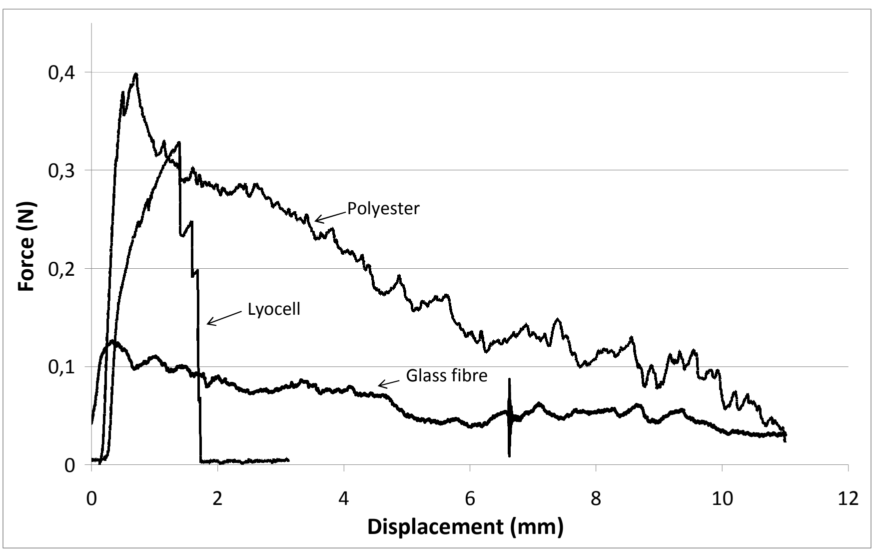


Figure 12. Typical force-displacement curves for glass fibre, polyester and lyocell fibres being pulled out from handsheets produced from softwood pulp.

3.4 Man-made fibres

Man-made fibres are not widely used in papers. They are more often used in the closely related non-woven industry. However, some studies of the application of man-made fibres in paper can be found in the literature. In the production of man-made fibres, a spin finish is often included to control the friction and to disperse static electricity. The spin finish affects the bonding ability of the fibres, and a common problem with composites incorporating man-made fibres is the low bonding ability between the fibres and the matrix. To overcome this, coupling agents can be added or the fibre surface can be modified.

In the present work, the fibres were used as delivered and no additives were used. This section gives a short description of the man-made fibres used in this work and their application in the paper industry.

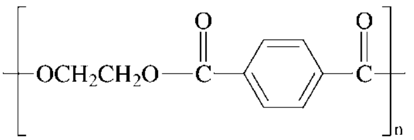
Lyocell

Lyocell is regenerated cellulose from a solution of dissolving pulp developed primarily for the preparation of chemical derivatives of cellulose. It is by one manufacturer marketed under the trademark Tencel®. The production process includes dissolution in amine oxide, filtration, spinning, washing and drying. The lyocell fibre should be biodegradable and the solvent is to a large extent recovered and reused. The fibres can thus be considered relatively eco-friendly. However, considerable amount of energy are consumed in the production process. The lyocell fibre can be fibrillated during wet processing to give a web of high wet strength. The largest application for lyocell fibres by far is in the textile and non-wovens industry, but lyocell fibres are also used in filter and specialty papers. (Kadolph 2007)

In the present study, lyocell was chosen since it was believed that it was able to form hydrogen bonds with wood fibres, based on its origin from cellulose fibres. Lyocell was chosen instead of viscose because of its lower extensibility.

Polyester

Polyesters are polymers containing esters of a substituted aromatic carboxylic acid. Polyesters can be saturated, most often thermoplastic, and unsaturated, thermosetting, depending on whether or not they contain reactive double bonds. Polyesters can be synthesized by sequential polymerisation from a diol and a carbonic acid but more common is synthetizing by rearrangement reactions through hydrolysis (Terselius 2010). The most common polyester is polyethylene terephthalate, commonly known as PET, and produced from aromatic terephthalic acid and ethylene glycol. Polyester fibres are particularly used in the textile industry where they are commonly blended with cotton or wool, but polyester fibres are also used as rubber reinforcement. (Reese 2001)



Poly(ethylene terephthalate) (PET)

The polyester fibres were used in this study since their ability to create chemical and mechanical bonds with wood fibres was judged to be intermediate between that of lyocell and glass fibres.

Glass fibre

The term “glass fibre” is a generic term for inorganic materials produced from a melt shaped as fibres of a diameter of about 0.001 to 0.1 mm. Glass fibres include, for example, glass wool, mineral wool and textile glass fibres (Linzander and Lundberg 2010). Glass fibres are produced from aluminium boron silicate and extruded into fibres. To obtain the desired surface properties, depending on the end use, a spin finish can be used. Glass fibres are strong and ductile but not elastic. Glass fibre mats are used for example in protective textiles and as cord in tires. Short cut fibres are used as reinforcement in plastics and concrete.

The fibres used in the present project were textile glass fibres, henceforth referred to as glass fibres. Glass fibres were chosen in this study as a model for fibres with a very low ability to form bonds with wood fibres.

4 Paper properties

Some of the paper properties discussed in this thesis and in the appended papers are described briefly below. Standardized methods are used as far as possible. It is common practice to report most paper strength properties in the form of an index, i.e. normalized with respect to the grammage, in order to eliminate the effect of grammage variations.

4.1 Formation

Formation is an expression for the small-scale variations in grammage. Large variation in the grammage means a poor formation and can cause variations of both optical and strength properties in the sheet. Poor formation also affects the runnability of the paper in the machine and is liable to lead to web breaks. The measure is called formation number. This is dimensionless and defined as the coefficient of variation of the local grammage. The specific formation index is the formation number divided by the square root of the grammage. There are different methods for the evaluation of formation, the most common being based on optical methods or β-radiation absorbency. The β-radiation has a low scattering, and this reduces the problems connected with light transmission when evaluating sheets containing different components with variation in the optical properties. In this project an AMBERTEC Beta Formation Tester was used to evaluate the formation. It measures the grammage at a number of points over a given area of the sheet and yields profiles of the grammage variation and charts of the fibre floc distribution.

4.2 Thickness

The thickness of a paper sheet determines primarily its bending stiffness, and an even thickness profile is desirable in for example converting processes. The dominating method for the determination of paper thickness is to measure the distance between two parallel plates under a given load (ISO 534:2005). The method can be used both on single sheets and on a pile of several sheets. When several sheets are measured, the effect of unevenness in the surfaces is reduced. However, this method still overestimates the thickness (Fellers and Nordman 1998, Schultz-Eklund et al. 1991).

In the present work, the thickness was determined by evaluation of the thickness profile (Fellers et al. 1986, Schultz-Eklund et al. 1991) (i.e. SCAN-P 88:01), where a test piece is fed at constant speed through a nip between two spherical probes. The probes thus follow the thickness profile of the sheet and the distance between the probes is continuously recorded. An average thickness for the sheet can be calculated from the profile and this is referred to as the structural thickness and is reported in µm.

4.3 Tensile properties

The tensile properties of paper are measured by clamping a strip between two grips and applying a tensile load until the strip breaks (ISO 1924-3:05. The applied load and the elongation are constantly measured throughout the test. A curve, as shown in Figure 13, can be obtained, where the load is plotted versus the elongation. Stress is the force required to elongate unit cross-sectional area of the material, but in the paper literature, the term stress is often used for force divided by width, since paper is a porous material and the thickness includes also the voids that do not contribute to strength. The elongation can be reported as a fraction, the elongation divided by the original length, or as a percentage, the fractional elongation multiplied by 100. Factors affecting the test results are grip pressure, specimen size, strain rate, moisture content and temperature. The test should be performed with sufficiently long strips to give a state of pure tensile stress in the middle of the strip, a standardized sample width, a constant strain rate and in a standardized climate. The tensile test is performed both in the machine direction, MD, and the cross machine direction, CD, and, when comparing papers with different anisotropies, a geometric mean can be calculated according to:

$$P_{Geo} = \sqrt{P_{MD} * P_{CD}} \qquad (3)$$

where P_{Geo} is the geometric mean value of the property, P_{MD} the value of the property in the machine direction and P_{CD} the value of the property in the cross machine direction.

Tensile strength is defined as the breaking force divided by the width of the strip and has the units kN/m. Tensile index is the tensile strength divided by the grammage and the unit is kNm/kg.

Strain-at-break, is defined as the ratio of the increase in length until the onset of rupture to the original length. Stretch-at-break is the ratio of the length at rupture to the original length. Both has units of m/m or as a percentage.

Tensile energy absorption, TEA, or tensile strain energy, is the amount of energy per unit area of the paper absorbed during straining until the onset of rupture in a tensile test. It can be illustrated as the area under the load-elongation curve and has the units J/m^2.

Tensile stiffness is determined from the slope of the initial linear elastic part of the load-elongation curve. Tensile stiffness is the force divided by the elongation and the width of the strip, and it has the unit kN/m. The elastic modulus can be obtained by dividing the tensile stiffness by the thickness of the strip. The unit for elastic modulus is MN/m^2, or MPa.

The nature of the fracture is dependent on the degree of bonding in the sheet (Page 1969, Davison 1972). A high degree of bonding leads to a large propagation of fibre breaks, whereas in a poorly bonded sheet the dominating process in the fracture zone is that fibres are pulled out of the network as the fibre-fibre bonds break. Refining improves the tensile properties in several ways. The fibres become more flexible, which facilitates the formation of fibre-fibre bonds. Also the fines content in the pulp is increased, leading to an increase in bond strength. Wet pressing increases the density and improves the tensile properties as a result of an increase in bonded area. The stretch-at-break is highly dependent on the strain during the drying of the paper; freely dried paper has a much higher stretch-at-break than paper dried under restraint. Seth (1990) showed that the tensile strength increased with increasing density and decreased with decreasing fibre strength. He also showed a similar correlation for stretch-at-break. Further, Seth showed that both tensile strength and stretch-at-break increased with increasing fibre length. Paavilainen (1993a) saw a decrease in tensile strength with increasing coarseness and suggested that the most important factors for high tensile strength are good bonding ability and high intrinsic fibre strength. In a different study, Paavilainen (1993b) continued the discussion, suggesting that the tensile strength is determined by the bonded area, thus collapsibility, external fibrillation, amount of fines and especially wet fibre flexibility. Mohlin at al. (1996) studied the impact of fibre deformation on sheet strength. They saw a decrease in tensile and tensile stiffness indices with increasing total number of fibre deformations, while the stretch-at-break was increased with decreasing shape factor.

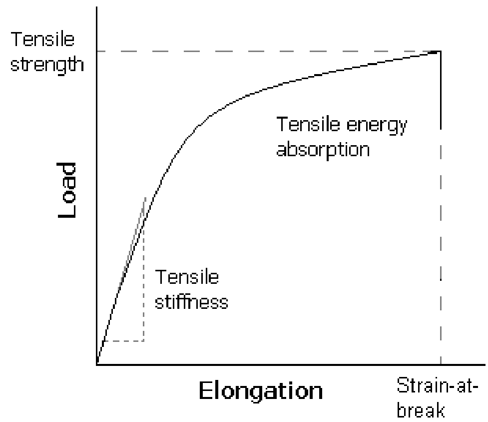


Figure 13. Schematic sketch of a typical load-elongation curve for paper, for the determination of tensile strength, tensile stiffness, tensile energy absorption and strain-at-break.

4.4 Fracture toughness

Fracture mechanic theory, where the loading situation, the geometry of the test piece and the fracture toughness are mathematically related, can be applied to paper. Mäkelä (2000) has presented a thorough literature review of the fracture of paper. Over the years, both linear elastic fracture mechanics, LEFM, and non-linear fracture mechanics, NLFM, have been applied to paper. However, Mäkelä's conclusion is that the application of LEFM to paper is limited since paper shows no pronounced linear elastic behaviour. Instead, NLFM should be used. In fracture mechanics, the energy release rate is the potential energy released per unit area of the propagating crack (Irwin 1956) according to:

$$G = -\frac{dU}{dA} \qquad (4)$$

where G is the energy release rate, U is the potential energy and A is the crack area.

For non-elastic materials, the energy release rate can be calculated by the J-integral (Rice 1968) according to:

$$J = -\frac{dU}{dA} = \oint_{\Gamma} \left[\varpi dy - T_i \frac{\partial u_i}{\partial x} ds \right] \quad (5)$$

where Γ is an integration path surrounding the crack tip, ϖ is the strain energy density, T_i and u_i are the traction vector and displacement vector, respectively, and s is the arc length of the integration path, as shown in Figure 14.

The fracture toughness is the critical value of the energy release rate when a crack starts to propagate. However, in the case of paper, it is of more practical interest to analyse failure than crack growth initiation, and the point of crack growth initiation is in any case ill-defined. It has therefore become common practice to use the load at failure as the fracture toughness instead of the load at the point of crack growth initiation. In practical terms, the fracture toughness of a paper is its ability to resist crack propagation. In this work, fracture toughness has been measured by means of a tensile test after an initial crack of a given length has been introduced into the strip. The fracture toughness is then calculated by the J-integral method using an evaluation procedure described by Wellmar et al. (1997) (i.e. SCAN-P 77:95). Fracture toughness has the unit J/m, and the fracture toughness index has the unit Jm/kg. Fracture toughness can be determined both in MD and CD and a geometric mean value can be calculated according to Equation (3).

Seth (1996) showed a linearly increasing relationship between fracture toughness of paper and fibre length. He also showed that more slender fibres gave a higher fracture toughness than fibres with a higher coarseness. Yu (2001) showed for different mixtures of softwood kraft, TMP, birch and straw pulps that the fracture energy increased approximately linearly with increasing fibre length, confirming Seth's findings.

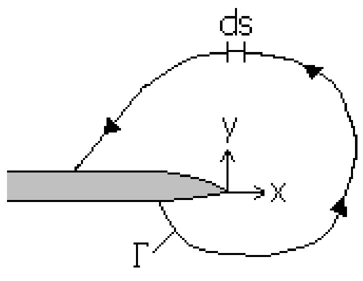


Figure 14. The J-integral contour. (Redrawn from Mäkelä 2000).

4.5 Tearing resistance

Tearing resistance is a measure of the force needed to tear the paper. There are different methods for measuring the tearing resistance of paper. In the work described in this thesis, the Elmendorf method (ISO 1974:90), which is an out-of-plane test, has been used. The test is performed, after introducing a crack in the test piece, by applying a load perpendicular to the face to pull the paper apart, as indicated in Figure 15. A swinging pendulum completes the tearing and the energy consumed during the tearing is measured. The total work is divided by the length of the test piece, and the tearing resistance is given in mN. When tearing resistance is normalized with respect to grammage, i.e. tear index, the unit is mNm^2/g. Tearing resistance can be determined both in MD and CD and a geometric mean value can be calculated according to Equation (3).

The tearing process is complex since the plane of fracture changes during the propagation of the tear. The fracture surface is oriented at 90° to the faces in the beginning of the tear and at almost 180° at the end. Further, the behaviour of the weak plane parallel to the faces is important in the process and the paper tends to split.

The mechanism of tearing resistance was first addressed by van den Akker (Instrumentation Studies XLVI 1944). He proposed that the energy consumed during a tear test is the sum of two processes: fibre fracture and fibre pull-out, the fibre pull-out providing the dominant contribution. The importance of the two factors has however been discussed over the years (Helle 1963, Shallhorn and Karnis 1979, Page 1994, Yan and Kortschot 1997). Helle considered that the larger amount of fibre fracture in a tear test than in a tensile test indicates that a higher force is needed in the tear test. One of the assumptions on which the Shallhorn and Karnis model is based is that the energy required to break a fibre makes a negligible contribution to the tearing resistance. Page however stresses that the energy required to break a fibre is not negligible. Further, Page also considers that Helle's results contradict van den Akker's theory, and suggests a new mechanism of tearing resistance divided into two parts: in loosely bonded sheets, below the maximum tearing resistance, both fibre failure and fibre-fibre bond failure contribute to the energy consumed, whereas in well-bonded sheets, the predominant mechanism is fibre failure. Yan and Kortschot added another contribution to the theory. They performed pull-out tests and proposed that when a fibre is pulled out from the network, energy is consumed both in breaking bonds and in overcoming frictional forces. Further, Yan and Kortschot concluded that if the fibres are very poorly bonded, are very strong, or are very short, all the fibres are pulled out intact and the fibre strength does not affect the tearing resistance. This is in agreement with the findings of Seth and Page (1988) who concluded that the tearing resistance of a sheet with a low degree of bonding is, to a great extent, determined by the fibre length, while the fibre strength is more important for the tearing resistance of well-bonded sheets. This was further supported by Page and McLeod (1992) who showed that at a given tensile strength the tearing resistance of a well-bonded sheet is proportional to the fibre strength raised to a power between 2,5 and 3,0. Further support was provided by Yan and Kortschot (1996) who modelled the out-of plane tear energy absorption. Their model showed that, for constant fibre length and strength, the tear energy passes through a maximum when the degree of bonding is increased. For a weaker fibre, the maximum occurs at a lower degree of bonding, whereas for a stronger fibre the maximum tear energy is reached in a well-bonded sheet. This is in agreement with the early finding of Parsons (1969), who showed that the tearing resistance is highly influenced by the degree of bonding and that a maximum occurs in a narrow range of bond density.

The effect on tearing resistance of fibre coarseness has been studied by some researchers. Seth and Page (1988) and Yu (2001) showed that coarser fibres gave sheets with a higher tear index than finer fibres. Lee et al. (1991), however, saw no correlation between fibre coarseness and tearing resistance.

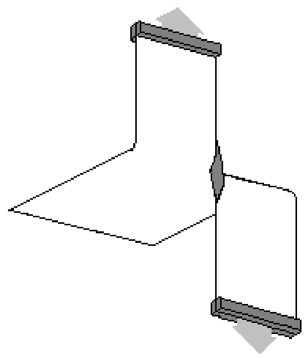


Figure 15. Schematic sketch of the direction of load during the Elmendorf tear test. (Redrawn from Fellers and Norman 1998).

4.6 Folding endurance

When folding endurance is measured, a strip is subjected to a constant tensile load and is folded backwards and forwards (ISO 5626:93). This is the only fatigue test used for paper. Folding number is the number of double folds that the strip resists before breakage. Since the fold number varies over a large range and the distribution is severely skew, it is more convenient to report folding endurance, which is the common logarithm of the fold number. Folding endurance can be determined both in MD and CD and a geometric mean value can be calculated according to Equation (3). There are various principles for measurement of the folding endurance and in this study the Köhler-Mohlin instrument, see Figure 16, was used. The test strip is clamped vertically between an upper and a lower clamp. The upper clamp can rotate 156° in each direction from the starting point. A weight is attached to the lower clamp so that the test strip is under a tensile load during the test. The folding is conducted by the upper clamp rotating backwards and forwards.

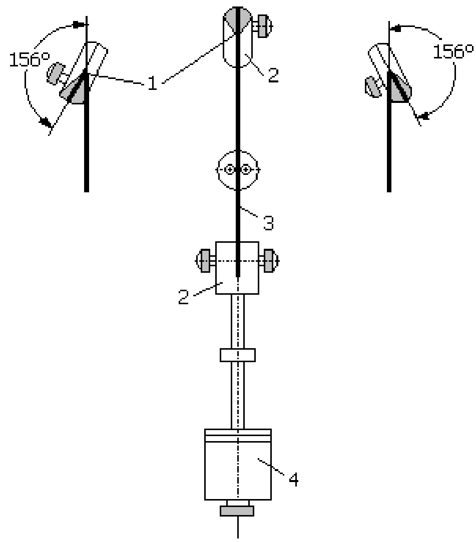


Figure 16. Schematic sketch of the Köhler-Mohlin instrument for folding endurance test. 1-Turning point, 2-Clamps, 3-Test strip, 4-Weight. (Redrawn from Fellers and Norman 1998).

Seth (1990) showed an increase in folding endurance with increasing fibre length and increasing sheet density. He also showed that folding endurance decreased rapidly with decreasing fibre strength.

4.7 Internal bond strength

Since paper has to a large extent a layered structure, especially in handsheets, measurement of the z-directional tensile strength provides an indication of the internal bond strength. It is important to bear in mind that commercial papers contain fibre entanglements and that the z-directional fibre distribution affects the z-directional strength. The internal bond strength of a paper is commonly measured by the Scott Bond test (TAPPI T 569) or the z-directional tensile strength test (TAPPI 541). In the Scott Bond method, an L-shaped block is attached with double-sided tape to the top face of the test sample and the bottom face is attached to a flat block, see Figure 17. The angled block is hit by a pendulum and the energy required to delaminate the test sample is measured. The energy absorbed is then divided by the cross-sectional area of the sample to give a measure of the internal bond strength. The z-directional tensile strength is defined as the force perpendicular to the test sample required to produce unit area fracture. In the z-directional tensile strength test, double-sided tape is used to attach two flat, smooth plates to the test sample, one on each face, as shown in Figure 18. The plates are then pulled apart in a tensile tester and the failure stress is measured. Stress-strain or stress-time curves can be obtained.

Neither of the two methods measures the true internal bond strength because of difficulties in determining the number and size of the bonds involved, and because breakage of fibre walls may occur and affect the test result. Further the adhesive on the double-sided tape may penetrate the test sample and provide unwanted reinforcement. The z-strength method is perhaps preferable, since in the Scott Bond method involves non-uniform stress and shear distributions during the fracture process. However, the z-directional strength method involves difficulties in applying a uniform tensile load over the specimen. The plates and the tensile forces have to be perfectly aligned. The gauge length of the test is the thickness of the paper and thus the result is sensitive to thickness variations within the sample. In the present study the z-directional tensile strength method was used to measure the internal bond strength, with the unit kPa.

Andersson (1981a; 1981b) measured the internal bond strength using the z-directional tensile strength test and found no correlation between either fibre length or intrinsic fibre strength and the internal bond strength. However, he saw an approximately linear relationship between z-strength and light scattering coefficient and a strong relationship between z-strength and density, supporting the idea that z-directional tensile strength is a measure of the internal bond strength. Further, Seth (1990) showed that the internal bond strength, measured by the Scott Bond test, is not affected by fibre length for sheets of a given density. Retulainen and Ebeling (1993) measured the Scott Bond strength of sheets with different RBA of latewood and earlywood respectively. They saw an increase in Scott Bond with increasing RBA and, for a given RBA, the sheets produced from latewood fibres showed a higher internal bond strength.

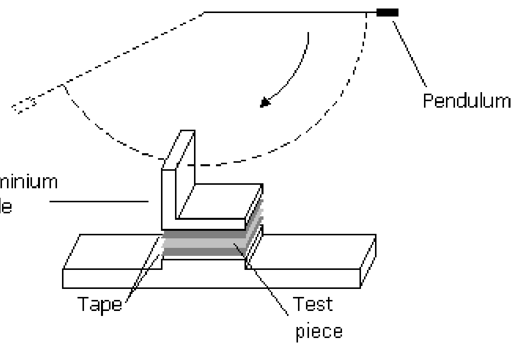


Figure 17. Schematic sketch of the Scott Bond testing method. (Redrawn from Fellers and Norman 1998).

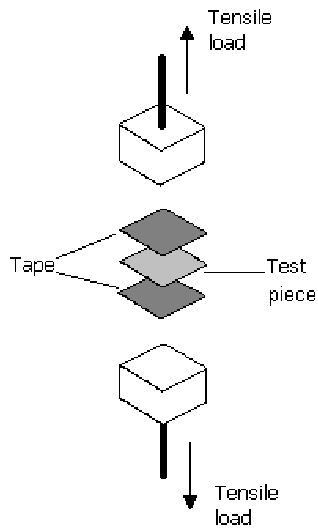


Figure 18. Schematic sketch of the z-strength testing method. (Redrawn from Fellers and Norman 1998).

5 Increasing paper strength

5.1 Mechanical and chemical treatment of fibres

The strength properties of paper may be enhanced in several ways by mechanical or chemical treatment of the fibres. Refining, the addition of wet and dry strength agents and chemical modification of the fibre surfaces are discussed briefly here.

The main target of refining is to improve the bonding ability of fibres so that they form a strong and smooth paper. The fibres become more flexible, the fibre walls are partly damaged exposing new sites available for hydrogen bonds and fines are created, and this promotes bonding and thus enhances the strength properties. Tensile strength, fracture toughness and folding endurance increase with increasing refining, but they eventually reach a maximum and decrease with further refining. Tearing resistance often passes through a maximum at a fairly low level of refining and then decreases with further refining (Fellers and Norman 1998; Lumiainen and Partanen 1997). The increase in tensile strength and fracture toughness is a result of the increased bonding level, while the decrease in tearing resistance is due to the shortening of fibres and also the increase in fibre bonding, so that fibres break instead of being pulled out of the fibre network, see further section 4.5.

When a paper comes into contact with water, hydrogen bonds between fibres are broken and replaced with bonds to water. The strength of a normal paper sheet in a wet condition is thus low. To protect the fibre-fibre bonds and enhance the wet strength, different types of wet strength agent resins may be added to the pulp. Depending on the type of resin, different mechanisms have been proposed for their action. They can form a protective network around the fibres and thus prevent bond failure or they can react with chemical groups at the fibre surface forming covalent bonds which enhance the strength of the fibre network. Further, the resin can penetrate and close the pores in the fibres, thus preventing fibre swelling and also stabilizing the network (Fellers and Norman 1998; Bates et al. 1999).

Dry strength additives are used to increase the strength of the dry paper. Most commercial dry strength agents are polymers such as starches, gums, carboxymethyl cellulose – CMC – and synthetic polymers (Davison 1980;

Ketula and Andersson 1999). Among the synthetic polymers, polyacrylamide, PAM, is the most commonly used. The dry strength agents may take part in the hydrogen bonding in the fibre network and enhance the degree of bonding. Further, they may form gels which promote the consolidation of the sheet by dissipating stress concentrations (Fellers and Norman 1998).

An interesting approach is to modify the fibre surfaces in order to improve paper strength properties. A number of articles by Wågberg and co-authors have been published on the subject (Wågberg et al. 2002; Gernandt et al. 2003; Gärdlund et al. 2003; Torgnysdotter and Wågberg 2003; Torgnysdotter and Wågberg 2004; Gärdlund et al. 2005). In the first of these articles, Wågberg et al. deposited layers of polyelectrolytes on the fibre surface. By building up 5-10 layers on unrefined fibres, they achieved sheets with the same tensile strength as sheets made of conventionally refined pulp. Further, they saw a doubling of the tensile strength in sheets produced from fibres with 5 layers of polyelectrolyte compared to that of sheets made of untreated fibres. The subsequent articles support the conclusion that it is possible to enhance paper strength properties by treatment of fibres with polyelectrolytes.

5.1.1 Refining of the pulps in Paper I

In the present study, only mechanical treatment (i.e. refining) was used to increase the fibre strength.

Figures 19 show the effect on tensile index and tearing resistance of refining of the two pulps used in Paper I, softwood and abaca. It can be seen that the tensile index of both the softwood and the abaca pulp increased sharply with refining at the beginning of the sequence. Both pulps then reached a plateau where the tensile index remained at a fairly constant level when the refining was further increased. At the end of the refining study the tensile index seemed to start to decrease, which would be a result of fibre weakening and shortening. The tear index showed a different behaviour. For the softwood pulp, there was first a sharp increase and then a sharp decrease at a low dewatering resistance level. When the dewatering resistance was over 20 SR, the tear index slowly decreased with increased refining. Abaca showed a sharp decrease in tear index with increased refining up to about 30 SR and then a moderate decrease up to about 40 SR. Thereafter the tear index seems unaffected by the refining up to a dewatering resistance of about 70 SR after which the tear index again apparently decreased.

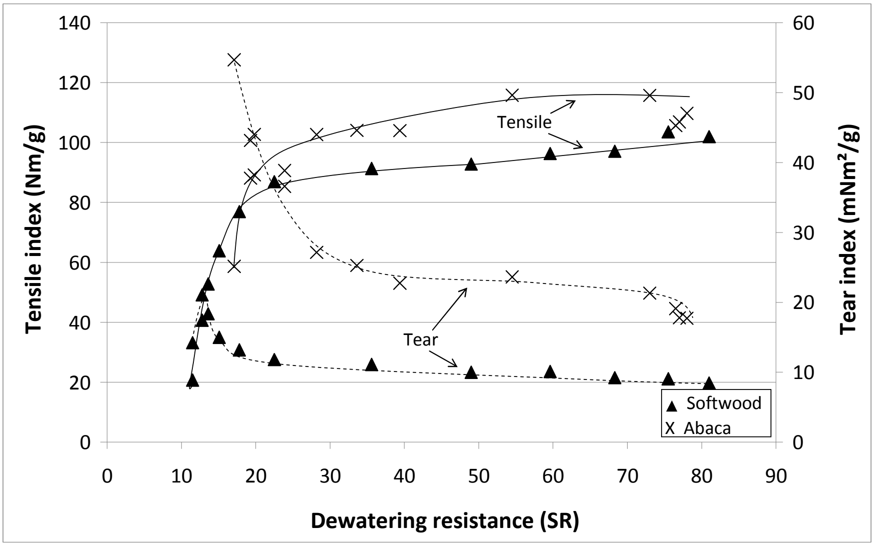


Figure 19. Tear and tensile indices plotted against dewatering resistance for the softwood and abaca pulps used in Paper I. (Results from Paper I).

5.2 Mixtures of different pulps

Mixtures of several pulp components are frequently used in the paper industry. The possibilities of enhancing paper strength by mixing different pulps have been studied extensively, but the performance of different pulp mixtures is not yet fully understood. In a mixture of two pulps, three different types of bonds exist; bonds within each pulp type and bonds between the different types of pulp. The formation of these bonds and their impact on the load-bearing capacity of the fibre network have not yet been clarified. It is well known that the linear rule of mixtures, or mass fraction additivity,

$$P_{Mix} = \sum_{i=1}^{n} w_i P_i \qquad (6)$$

where P_{Mix} is the value of any property for the mixed sheet, w_i is the mass fraction of pulp component i, P_i the value of the property for a sheet of pulp component i and n is the number of pulp components, is not always valid for pulp mixtures, and that synergetic or negatively deviating results can arise.

Görres et al. (1996) proposed that, in order to predict the properties of sheets made from mixtures of pulps, the fibre properties of the component pulps rather than the properties of sheets made from these pulps should be used. They developed a model for computing the density of mixed sheets using the average fibre properties of each pulp component weighted with the respect to their contribution to the total fibre length.

Bovin and Teder (1971) studied different kinds of pulp mixtures and found that it was possible to predict whether or not the tearing resistance of a mixture would deviate from linearity. When the tearing resistance is plotted against tensile strength for a pulp, a maximum usually appears on the curve. Bovin and Teder found that if the pulp components have the tensile strength on the same side of the maximum, the tearing resistance of the mixture is linearly additive.

Strength properties of mixtures of chemical and mechanical pulps have been shown to deviate from linear mass fraction additivity (Mohlin and Wennberg 1984; Retulainen 1992; Kazi and Kortschot 1996; Zhang et al. 2002; Hiltunen 2003; Honkasalo 2004). Results both higher and lower than those predicted have been reported. The reason for this behaviour is not clear, but most theories are based on the development of bonds in the mixture.

Mohlin and Wennberg (1984) observed a positive deviation from linear additivity for tearing resistance, whereas internal bond strength, measured as the Scott Bond strength and tensile properties showed a negative deviation. The results indicated that the bonding level of the kraft fibres cannot be fully utilized and Mohlin and Wennberg suggested that the interaction between the different pulps is weak and that the bonds between chemical and mechanical fibres are so weak that the pulps behave as if they form two separate networks.

Retulainen (1992) added earlywood and latewood kraft fibres to a TMP and saw a negative, non-linear deviation from linear mass fraction addivitity for the apparent density of the sheets. Further, the tearing resistance increased linearly with the addition of latewood fibres but showed a positive deviation when the more conformable earlywood fibres were added. The tensile strength and stiffness were affected only at an earlywood content greater than 40%, where there was a slight increase in strength. Retulainen suggested that the stiffer fibres prevent the flexible fibres from utilizing their full bonding potential and that the kraft fibres are not fully activated in the tensile test due to curls and kinks. Further, Retulainen investigated the bond strength of the different bonds in the sheet by measuring the z-directional tensile strength of two-layered sheets, i.e. the ply bond strength. He found that the z-strength of the TMP/kraft sheet was higher than that of the TMP/TMP sheet but lower than that of the kraft/kraft sheet, and he suggested that the bond strength between the different fibres should follow the same trend.

Kazi and Kortschot (1996) studied kraft-containing TMP sheets and showed a fracture toughness lower than the predicted value, although it increased with increasing kraft addition. However, with a 2.5% addition of kraft, a drop in the fracture toughness was observed. Light scattering measurements indicated that the kraft fibres, even in small amounts, were well bonded into the network. Kazi and Kortschot suggested that the kraft/TMP bonds are weak and do not contribute to the energy absorption during the fracture. This would explain the decrease in strength at low kraft contents and the increase when the kraft content increased allowing stronger bonds to form between kraft fibres.

Hiltunen (2003) saw a synergetic effect on fracture toughness when kraft pulp was added to TMP-based sheets. His results suggested that the linearity was dependent on the degree of refining of the kraft pulp, the addition of unrefined kraft pulp gave non-linear results.

Honkasalo (2004) found a synergism, especially in tearing resistance but also in fracture toughness, stretch-at-break and TEA, in sheets consisting of mixtures of groundwood, TMP and softwood kraft. The synergism seemed more likely to appear in well-bonded sheets. Further, the synergism in tearing resistance was most prominent when the compounds had bonding levels on opposite sides of their tearing resistance maxima, in agreement with the results of Bovin and Teder. Honkasalo suggested that an optimum combination of fibre length and bonding degree could give a synergism in paper strength properties.

Fernandez and Young (1994) analysed the results of Mohlin and Wennberg (1984) and Retulainen (1992) and suggested that collapse of the kraft pulp fibres is the main reason for the deviation from linearity in density and tearing resistance. The tension induced during drying causes the fibres to collapse but, when the bonding level is low, the fibres are able to shrink in the longitudinal direction and do not collapse to the same extent as they do in well-bonded sheets. Fernandez and Young argued that the mechanical pulp reduces the bonding between kraft fibres and thus decreases the tendency for the kraft fibres to collapse.

Zhang et al. (2002) saw both linear and non-linear behaviour of the strength properties in sheets consisting of kraft and TMP. They argued that the behaviour was dependent on the degree of bonding in the sheet. When the bonding potential of both the pulps was fully utilized, they saw a linear relationship in the strength properties, whereas a deviation, generally negative, was seen when the bonding potential was not fully utilized.

5.2.1 Mixtures of pulps in Paper I and Paper III

To summarize the literature, the behaviour of pulp mixtures seem to depend on the bond density in the fibre network. Some of the results in the present work are briefly discussed with respect to linear mass fraction additivity below. When using linear mass fraction additivity to predict properties of mixed sheets, the properties of sheets of the pulps were used rather than pulp or fibre properties. It is believed that hydrogen bonds are created between all fibre types in the sheets, so that a homogeneous network (as homogeneous as is possible in paper) is created. In Paper I, a mixture of two pulps, softwood pulp and abaca, was studied. In Paper III, three different pulps were used: Swedish softwood, abaca and southern pine. Sheets were produced with softwood and abaca or softwood and southern pine and the sheet properties were examined.

In Figure 20 the tensile and tear indices for sheets composed of softwood and abaca are plotted as a function of abaca content. The results predicted by linear mass fraction additivity are marked by solid lines. It can be seen that the tensile index follows the rule of mixing whereas the tear index deviates. This result is not in agreement with the hypothesis of Bovin and Teder, since these two pulps were on the same side of the tear-tensile maximum. The abaca fibres are believed to give a high tearing resistance because of their long fibre length and high strength. Based on the discussion on tearing resistance in section 4.5, the positive effect on tearing resistance of abaca fibres should be greatest in a sheet with low degree of bonding. The abaca fibres are more curled and less flexible than the softwood fibres. Thus, in a sheet with a high amount of abaca, the degree of bonding should be lower than that in a sheet where softwood is the dominating pulp. The negative deviation from linear mass fraction additivity can therefore be explained, at least partly, by the abaca fibres being relatively firmly bonded in the network with the flexible softwood fibres.

Figure 21 shows the tensile and tear indices of sheets composed of a homogeneous mixture of 50% Swedish softwood and 50% southern pine and sheets composed of 50% Swedish softwood and 50% abaca. The results predicted by linear mass fraction additivity are indicated in the figure. In the present work (Paper III), the southern pine and abaca pulps were fractionated before use to remove most of the fines. The sheets composed of Swedish softwood and southern pine followed the prediction for tear index whereas the tensile index was somewhat higher. The situation may be similar to that reported by Hiltunen, showing deviations from linearity when adding unrefined chemical pulp as reinforcement in a mechanical pulp rich in fines. The density of the sheets is somewhat higher than that predicted by linear mass fraction additivity, suggesting that the bond density was higher, resulting in a higher tensile strength. The sheets containing abaca deviated from the predicted result both in tensile and tear indices: approximately 15% higher tensile index and 20% lower tear. The lower tear can be explained by the same argumentation as in the work in Paper I, the abaca fibres being well bonded into the network with the flexible softwood fibres and fines and thus contributing less to the tearing resistance. The high bond density can also explain the high tensile index. When abaca fibres are activated in the network they contribute with their high fibre strength to the tensile strength of the sheet.

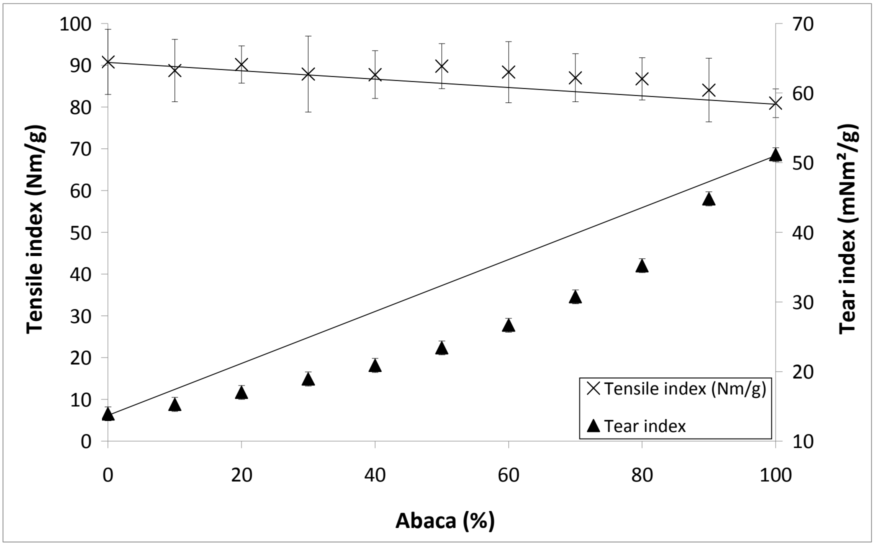


Figure 20. Tensile and tear indices plotted against abaca content for sheets consisting of abaca and softwood pulps. The lines represent linear mass fraction additivity. (Results from Paper I).

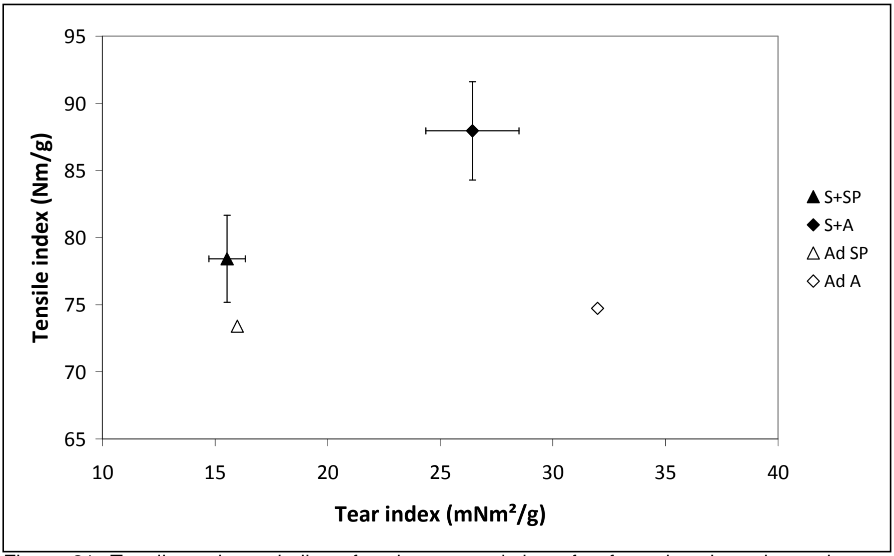


Figure 21. Tensile and tear indices for sheets consisting of softwood and southern pine or softwood and abaca. Ad SP and Ad A represent linear mass fraction additivity for sheets composed of softwood and southern pine and softwood and abaca respectively. (Results from Paper III).

5.3 Reinforcement

An important type of pulp mixture is that in which a reinforcement pulp is added. Reinforcement fibres are added to a pulp to improve the runnability of the paper web in the paper machine and the mechanical properties of the paper produced.

The dominant reinforcement situation in papermaking is the use of softwood pulp to improve the properties of weaker pulps such as mechanical or hardwood pulp. Several studies have been devoted to finding the optimal properties of a reinforcement pulp (Seth and Kingsland 1990, Retulainen 1992, Seth 1996, Alava and Niskanen 1997, Ebeling 2000, Hiltunen et al. 2002, Mansfield et al. 2004). Fibre coarseness, fibre length, mechanical properties and load activation of the fibres are factors that affect the reinforcement potential of a pulp. Most of the works referred to in the chapter 5.2 actually concern the interaction between mechanical pulps and reinforcement kraft pulps. Few studies have been devoted to the study of reinforcing kraft papers with other types of fibres.

In Paper V, man-made fibres were used to reinforce a softwood kraft pulp. The aim was to increase the tearing resistance by the addition of man-made fibres while the tensile strength could be increased by refining of the softwood pulp. The hypothesis was that fibres with low ability to form bonds with the softwood fibres should increase the tearing resistance regardless of the bond density in the sheet. Lyocell, polyester and glass fibres were chosen. No literature was found on the use of lyocell fibres in conventional papermaking, but a few studies in which viscose was used are summarized below, together with studies concerning the use of polyester and glass fibres in papermaking.

Heikal and El-Shinnawy (1980) added 5 mm long viscose fibres to a wood pulp in amounts from 1% up to 20%. The tearing resistance increased with increasing amount of viscose. The tensile strength, measured as breaking length, showed a remarkable increase with the addition of 1% viscose fibres. The tensile strength then decreased with increasing amount of viscose, but the sheets containing 5 and 10% viscose still showed a higher tensile strength than the reference sheet. The increase in tearing resistance was larger when the viscose fibres were added after refining of the wood pulp than when they were added before the refining. However, the tensile strength was higher when the viscose was added before the refining. Heikal and El-Shinnawy suggested that the result was a combined effect of an increase in mean fibre length and a decrease in sheet density.

Yu et al. (1999) studied the potential of viscose fibres, 3 or 6 mm long, to reinforce a softwood kraft pulp. They observed a decrease in density and in Scott Bond strength with the addition of viscose fibres, suggesting a decrease in RBA. As a result, the tensile strength also decreased with increasing amount of viscose fibres. However, the addition of up to 50% viscose increased the fracture toughness, with a maximum at 30% viscose.

Thomson et al. (2007) studied the interface between two viscose fibres with fluorescence resonance energy transfer and fluorescence microscopy and compared it to the interface between two wood fibres. Their results showed both greater and closer contact between the wood fibres, indicating a higher degree of bonding than between the viscose fibres. They suggested that the lack of fibrils and hemicelluloses on the viscose fibre surface was the cause of the lower contact in the fibre-fibre interface than in that between wood fibres.

El-Shinnawy et al. (1979) investigated the effect of adding polyester fibres to different pulps before or after refining of the bulk pulp. When polyester fibres were added to wood pulp it was seen that, with amounts up to 15%, the tearing resistance increased. Adding the fibres after refining of the bulk pulp resulted in a higher tearing resistance than adding the fibres before refining. Further, it seemed that the tensile strength remained at a constant level regardless of the amount added.

Ambardekar and co-authors (Ambardekar et al. 1998, Bhuwania et al. 1999) reported results from both laboratory studies and machine trials where short cut polyester fibres were used as reinforcement in different pulps and in different paper grades. With polyester levels up to 10%, they reported an increase in tearing resistance, while the tensile strength remained constant or was slightly reduced.

Singh and Wood (1996) prepared wet-laid glass fibre sheets, and studied the tensile strength of the sheets. They showed, not surprisingly, that the tensile strength of pure glass fibre sheets was lower than that of sheets based on wood fibres. However, the tensile strength was higher if the sheets were dried freely, without restraint. This was explained as being due to a tendency for the sheets to expand during drying, rather than to shrink, as in the case of wood fibres. This was believed to straighten the fibres making them ready to bear a load. When the sheets were dried under restraint the fibres were not able to expand, and the fibres were not activated to the same extent.

Singh (2000) later reported results for the addition of up to 10% glass fibres to bleached sulphite pulp and to old corrugated container pulp, OCC. In the case of the sulphite pulp, the tear resistance showed an increasing trend, while the tensile strength decreased. It was reported that an addition of 3 or 7% glass fibres to OCC pulp increased the tensile strength. The tearing resistance of the sheets containing 3% glass fibres was higher than that of the sheets of 100% OCC, while that of the sheets containing 7% glass fibres was somewhat lower.

Park et al. (2007) modelled the drying of wood fibres and compared it with glass fibres. They discussed the fact that, since the glass fibres are hydrophilic but not porous, there is no water inside the fibres, only between fibres in a network.

5.3.1 Reinforcement with man-made fibres in Paper V

The properties of the fibres used in Paper V are listed in Table 1. The coarseness of the softwood pulp, before refining, was 175 µm/g, or 1.75 dtex. Despite the length of the man-made fibres, the formation was only affected in the case of the sheets containing 3% glass fibres and the sheets containing 1% polyester. For these sheets, the specific formation index was twice that of the pure softwood sheet. The energy required to pull out a fibre from the softwood fibre network was evaluated as a measure of the ability to form bonds, see section 3.3.1. The results indicated that lyocell was most firmly attached to the fibre network and that the glass fibre was the least attached. Figure 22 shows the tear index of softwood sheets reinforced with different amounts of man-made fibres. The tear index was increased with the addition of man-made fibres and 5% glass fibres gave the highest tear index. Figure 23 shows that the tensile index decreased with the addition of man-made fibres but that, with the addition of glass fibres, the decrease was less than 10%. Thus it seems possible to increase the tearing resistance of softwood sheets by adding long man-made fibre with low bonding ability.

Table 1. Properties of man-made fibres. (Reprinted from Paper V).

Fibre	Glass fibre	Polyester	Lyocell
Fibre length, mm	12	12	12
Fibre diameter, µm	15	23	13
Coarseness, dtex	10	6,6	1,7
Breaking load, cN	31	44	5,1
Specific breaking load, cN/µm²	0.17	0.11	0.038
Strain at break, %	3	20	6

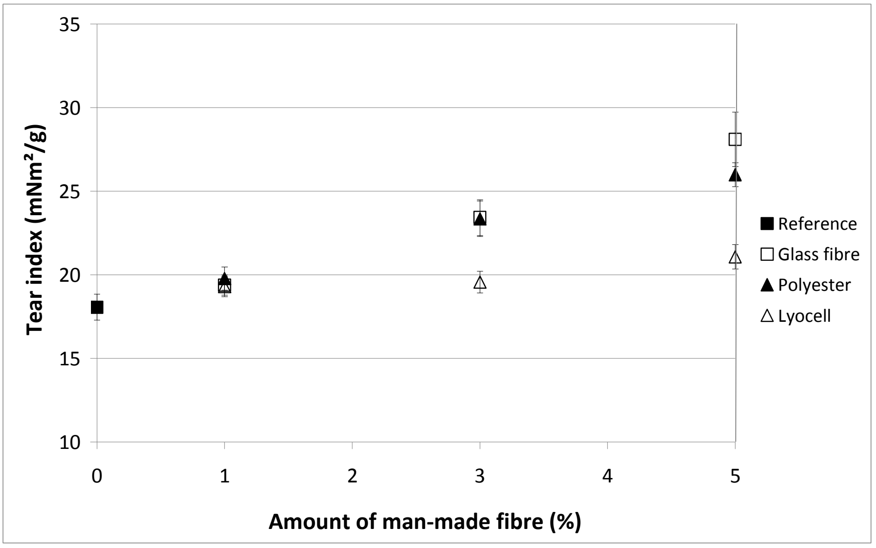


Figure 22. Tear index of softwood sheets reinforced with man-made fibres. (Reprinted from Paper V).

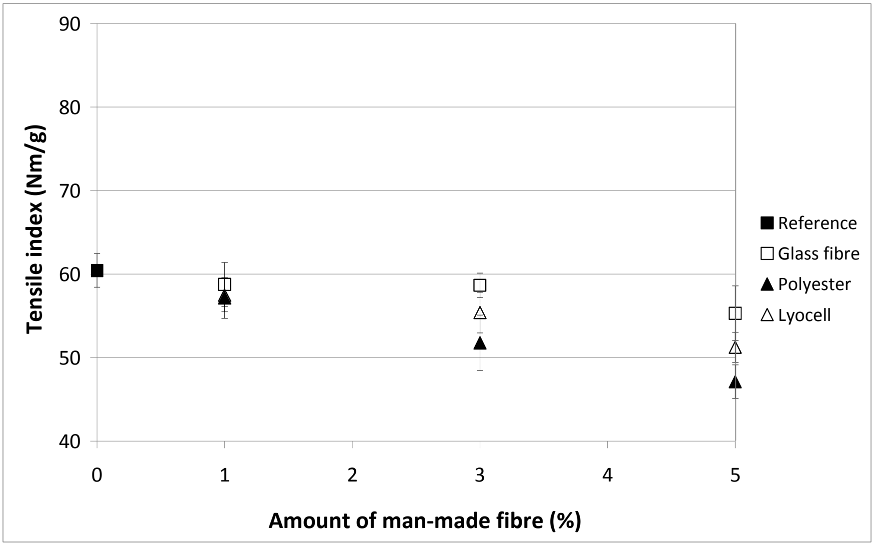


Figure 23. Tensile index of softwood sheets reinforced with man-made fibres. (Reprinted from Paper V).

To evaluate the effect of the man-made fibres in sheets with different bond densities, the softwood pulp was refined to three different dewatering resistances and handsheets were produced from each pulp. Figures 24-26 show the tear and tensile indices respectively for sheets produced from the softwood pulps reinforced with man-made fibres.

The glass fibers and polyester fibres increase the tear index to a similar extent, but the lyocell fibres gave a much lower contribution to the strength, probably due to a combination of the low breaking load of the fibres and the firm attachment in the softwood network. Reinforcement with glass fibres was expected to give a higher tear index than reinforcement with polyester because of the high specific breaking load and low bonding ability of the glass fibres. This was however only the case only in the sheets produced from the 19 SR softwood pulp. In the sheets made of a pulp with higher dewatering resistance, the man-made fibres are more firmly attached in the network. The fracture zone should thus be smaller, and the more brittle and less extensible glass fibres are more likely to break during the tear test than the polyester fibres. Further, the number of fibres for a given weight percentage is greater for polyester than for glass fibres and the polyester fibres have a larger fibre diameter, thus a greater force should be required to pull out the fibre from the network.

The effect on tensile index of adding the different man-made fibres was similar for all three fibre types, the tensile index decreasing with increasing addition. The higher the bond density in the sheets, the smaller was the negative effect on the tensile index. The glass fibres have the lowest number of fibres for a certain weight, a small fibre diameter and also a high specific breaking load, and this can explain why the tensile index seemed to be less affected when glass fibres were used.

The effects of adding polyester fibres are in agreement with those reported earlier (El-Shinnawy et al. 1979, Ambardekar et al. 1998, Bhuwania et al. 1999). The tearing resistance was increased and the tensile strength was slightly reduced. The effects of adding glass fibres are in agreement with those reported by Singh (2000) regarding the addition of glass fibres to a sulphite pulp. Singh saw however an increase in tensile strength with addition of glass fibres to OCC pulp, whereas the tensile index decreased in the present study. However, the tensile strength of the OCC pulp was probably considerably lower than that of the softwood pulp used in this work, and the polyester fibres might therefore give a positive contribution to the tensile strength.

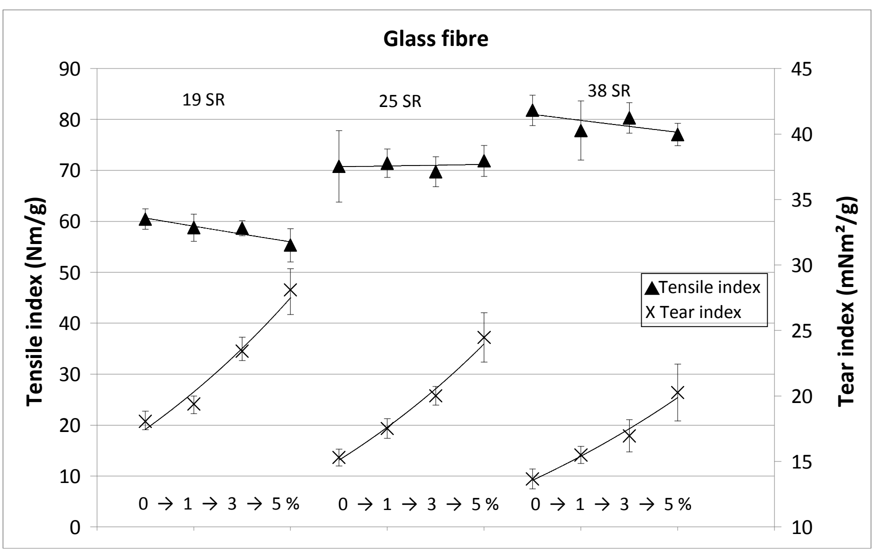


Figure 24. Tensile and tear indices of handsheets made from softwood kraft pulp reinforced with glass fibres. The amount of glass fibres was 0, 1, 3 and 5%. Three different dewatering resistances were used for the softwood pulp: 19, 25 and 38 SR. (Results from Paper V).

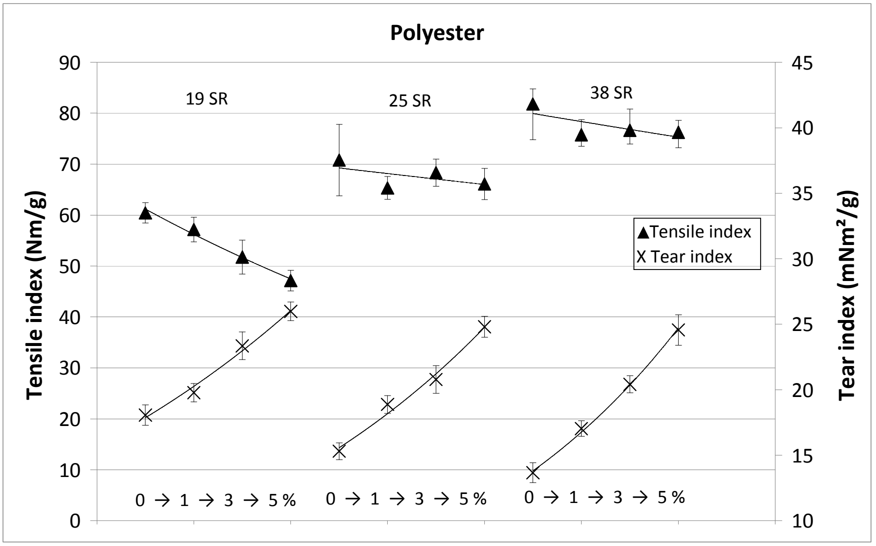


Figure 25. Tensile and tear indices of handsheets made from softwood kraft pulp reinforced with polyester fibres. The amount of polyester fibres was 0, 1, 3 and 5%. Three different dewatering resistances were used for the softwood pulp: 19, 25 and 38 SR. (Results from Paper V).

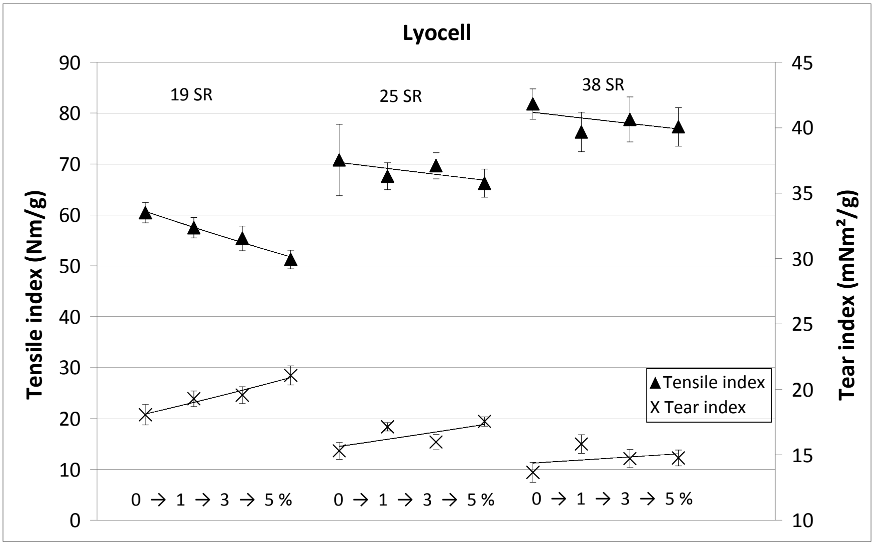


Figure 26. Tensile and tear indices of handsheets made from softwood kraft pulp reinforced with lyocell fibres. The amount of lyocell fibres was 0, 1, 3 and 5%. Three different dewatering resistances were used for the softwood pulp: 19, 25 and 38 SR. (Results from Paper V).

5.4 Multilayering or stratification

As mentioned in section 2.2, multilayering or stratification can be used to increase the stiffness of the paper, improve the surface properties and improve the fibre economics. This section presents a summary of studies on multilayer or stratified papers for improvement of paper strength properties and a discussion on relevant results from the work included in this thesis.

In 1974, Stöckmann (1974b) studied the effect of layering 50% kraft and 50% groundwood. With a three-layer sheet with kraft in the outer layers and groundwood in the middle, he obtained a tensile strength at the same level as that of a single-layer sheet of a homogeneous pulp mixture, despite a decrease in density, and the tearing resistance of the layered sheets was higher than that of the homogeneous sheets. When groundwood was placed in the outer layers, however, both the tensile strength and the tearing resistance were lower than those of the homogeneous sheet.

Erickson (1977) showed that three-layer sheets consisting of softwood kraft and hardwood sulphite had the highest tensile strength when the hardwood pulp was placed in the outer layers. With softwood in the outer layers, the tensile strength was lower than that of the homogeneous single-layer sheets. The tearing resistance was highest in the mixed sheets.

Bristow and Pauler (1983) studied the layering of a chemical pulp (50% birch kraft and 50% softwood kraft) and a groundwood pulp. Three-layer sheets with the pulp composition chemical / groundwood / chemical or groundwood / chemical / groundwood were compared to homogeneous single-layer sheets. It was seen that the tensile strength was slightly higher for the layered sheets, both for the sheets with chemical pulp in the outer layers and for the sheets with groundwood in the outer layers. The strength of the layered sheets was close to that predicted by linear mass fraction additivity while that of the mixed sheets was about 10% lower. With regard to tensile stiffness, the mixed sheets showed values close to the predicted additivity level, while the layered sheets showed a slightly higher value. Again, the relative order of the layers seemed unimportant. The layering had no clear effect on tearing resistance, but the results indicated that the sheets with chemical pulp in the outer layers had a slightly higher tearing resistance.

Terland and Fellers (1986) studied the possibility of increasing paper strength properties by layering a single pulp. Two separate layers of non-bleached softwood sulphate pulp were formed and combined into a two-layer sheet. At constant forming concentration, Terland and Fellers saw no effect of layering on tensile index, tensile energy absorption or tear index.

Fredlund et al. (1989) studied the layering of hardwood and softwood pulp. The top layer was produced in a Fourdrinier machine while the other layers were produced in a cylinder machine. Different sheet compositions, layered and homogeneously mixed, were compared and no effect of the layering was seen on either tensile stiffness index or tear index. The fracture toughness index of the layered sheets was slightly lower than that of the homogeneous sheets.

Koran and Kamdem (1990) saw a decrease in the geometric mean of the tearing resistance of two- and three-layer sheets consisting of 50% kraft and 50% thermomechanical pulp, TMP, compared to that of a homogeneous single-layer sheet. The geometric mean tensile strength was higher in the two-layer sheets and in the three-layer sheets with kraft in the outer layers, whereas the three-layer sheets with TMP in the outer layer showed a tensile strength lower than that of the mixed sheets.

Nesbakk and Helle (2003) studied the properties of three-layer sheets composed of different combinations of fractions from two TMP pulps, and found that the tensile strength in MD was somewhat decreased while that in CD was increased for the layered sheets compared to that of the homogeneous single-layer sheets.

5.4.1 Stratification in Paper III

The literature does not give a concordant picture on the effects of layering. Thus, in this study, only two layers were used, to minimize the factors affecting the result. All the sheets composed of two pulps consisted of 50% of each of the pulps. The work reported in Paper III is based on three pulps: Swedish softwood kraft pulp, southern pine and abaca. In Paper IV only softwood and abaca were used, but the softwood pulp was refined to three different dewatering resistances. Figure 27 show the tensile and tear indices for single-layer sheets of softwood, southern pine and abaca, for single-layer sheets containing a homogeneous mixture of softwood and southern pine or softwood and abaca, and for two-layer stratified sheets composed of one layer of softwood and one layer of southern pine or abaca. The results predicted by linear mass fraction additivity are also indicated. Both southern pine and abaca had a lower tensile index but a higher tear index than the softwood pulp. It can be seen that, by placing southern pine and abaca in separate layers, the tear index increased compared to that of the mixed sheets, but that the tensile index decreased. Both stratified sheets showed results close to the predicted values according to linear mass fraction additivity.

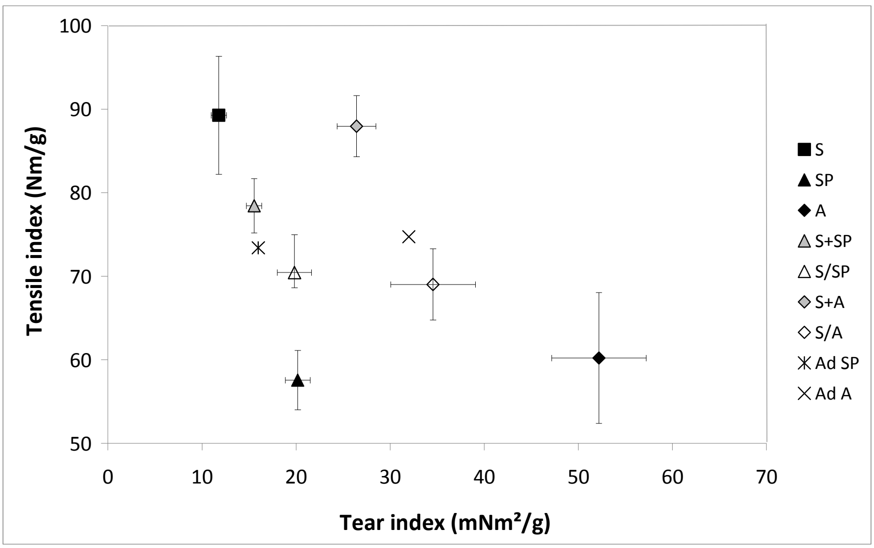


Figure 27. Tensile and tear indices for single-layer and two-layer sheets composed of softwood, S, southern pine, SP, and abaca, A. S: Single-layer sheet composed of 100% softwood. S+SP: Single-layer sheet composed of a homogeneous mixture of 50% softwood and 50% southern pine. S/SP: Two-layer sheet composed of one layer of softwood and one layer of southern pine. Ad SP: The result predicted by linear mass fraction additivity. The corresponding terms are used for the softwood/abaca sheets. (Results from Paper III).

The loading situation in and around the fracture zone differs between tensile and tear tests, and the loading situation also differs between single-layer and two-layer sheets. When a sheet composed of a homogeneous mixture of different fibres is exposed to an in-plane load, as in tensile testing, the stress and strain in the different fibres depend on the activation of the fibres in the fibre network, i.e. the extent to which the fibres carry a load. This in turn is governed by the number of bonds on each single fibre and the degree of straightening of curl and kinks in the free fibre segments between the bonds. In a two-layer sheet, the layers experience the same external strain under load. The strength of a two-layer sheet should be the sum of the two constituent layers.

The load-strain curves of single-layer sheets from softwood and abaca, for combinations of single-layer abaca sheets placed on top of single-layer softwood sheets, for stratified sheets with softwood and abaca and for homogeneous mixed sheets of softwood and abaca are shown in Figure 28. In the case of the two single-layer sheets placed on top of each other, the curve indicates that, when the strain at break of the abaca layer is reached this layer brakes. In some cases the softwood layer continued to carry load up to the point when the breaking strain for the softwood layer was reached, but for most cases the softwood layer also broke when the abaca layer broke. The load-strain curve for the stratified sheet lied approximately where it was expected to be if the A30 curve is added to the S30 curve. The stratified sheet did however carry load up to a higher strain than the two single-layer sheets placed on top of each other, thus the layers in the stratified sheets interact and gives a strong sheet with a higher strain than its components. The homogeneous sheet showed the highest breaking load and breaking strain, higher than expected. It is suggested that the load-bearing capacity of the less collapsed and less flexible abaca fibres was activated to a greater extent when they were mixed with the softwood fibres. In the network together with the more flexible and conformable softwood fibres, the number of fibre-fibre contacts was greater, giving a larger number of bonds in the sheet. Also, the fines in the softwood pulp contributed to the degree of bonding. With a higher degree of activation of the abaca fibres, their individual fibre strength, indicated by the zero-span tensile index, should contribute to a high tensile strength of the sheets.

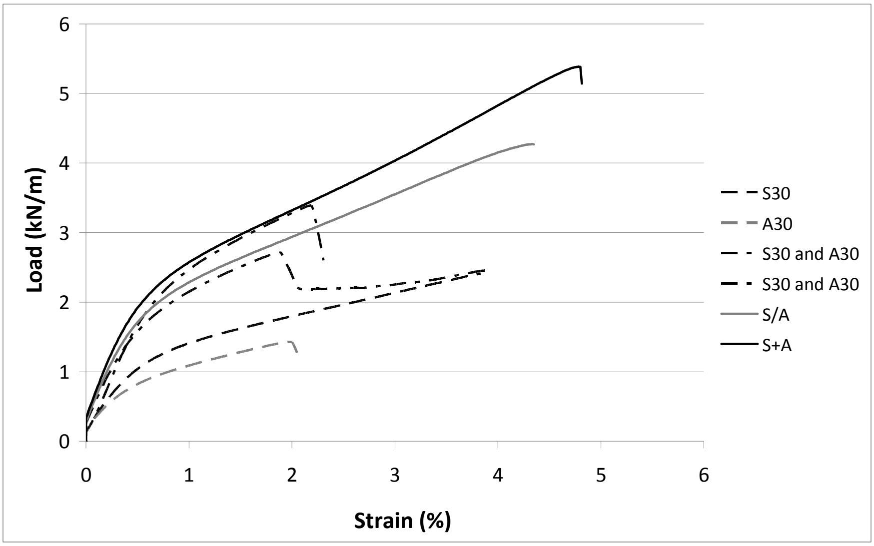


Figure 28. Load-strain curves for single-layer sheets of softwood, S30, abaca, A30, single-layer sheets of softwood and abaca placed on top of each other, S30 and A30, stratified sheets of softwood and abaca, S/A, and homogeneous sheets of softwood and abaca, S+A. The load has been normalized with respect to test strip width. (Results from Paper III).

As mentioned above, the load distribution in the sheet during a tear test differs from that during a tensile test. When a layered sheet is torn, the rupture line runs simultaneously through all the layers and thus all layers contribute to the tearing resistance throughout the entire fracture process. The crack propagation in the stronger layers cannot be accelerated by the presence of weaker layers, thus the tearing resistance of a layered sheet cannot be lower than that of its strongest layer. In the work reported in Paper III, the tear index of the stratified sheets was at the same level as that of the stronger component, i.e. 30 g/m² abaca sheets, and was higher than that of the mixed sheets. The lower tear index of the mixed sheets is probably a result of the abaca fibres being more firmly bonded in the network when mixed with the flexible softwood fibres and fines.

5.4.2 Stratification compared to refining in Paper IV

In Paper IV, the effect of stratifying sheets composed of softwood and abaca was compared to that of refining the softwood pulp. The tensile-tear relationship for mixed and stratified sheets composed of abaca and softwood pulp (three different dewatering degrees) is shown in Figure 29. The mixed and the stratified sheets show different responses to the increased refining of the softwood pulp. The mixed sheets behave as the pure softwood sheets, showing about the same gradient in the tensile-tear relationship with increased refining. In general, a decrease in tearing resistance with increasing refining is due to a higher degree of bonding and fibre shortening. In the mixed sheets containing abaca, these long fibres thus contribute less and less to the tearing resistance the higher the degree of bonding in the sheet.

The synergy in tensile index for the mixed sheets was also seen in Paper III, cf. Figure 27. The present results strengthen the suggestion that this phenomenon is related to the degree of bonding and activation of the fibres in the sheet. When the long, curled and moderately refined abaca fibres are mixed with the flexible, well-refined softwood fibres they experience a larger number of fibre-fibre contacts than in a network with only abaca fibres. This is because the softwood fibres contribute to a denser network since they are able to come into closer contact with the abaca fibres as a result of their greater flexibility and conformability and also because the softwood pulp includes fines. Thus the abaca fibres are more firmly bonded to the fibre network, the load-bearing capacity of the network is increased, and the strong abaca fibres therefore contribute to a high tensile strength. However, the high degree of bonding will reduce the contribution from the abaca fibres to the tearing resistance and result in a lower tearing resistance than that predicted by linear mass fraction additivity.

The stratified sheets, like the mixed sheets and the pure softwood sheets, also showed an increase in tensile index with increasing refining, but the tear index seems to be unaffected. The tearing resistance of the abaca fibres was considerably higher than that of the softwood fibres, so that the abaca layer provided the greatest resistance to crack propagation and dominated the result. The tearing resistance of the stratified sheet is therefore expected to remain at the same level even if the tearing resistance of the softwood layer was decreased. There was no significant variation, at a 5% statistical level in the tear index, of the stratified sheets with increasing refining, although the decrease in the tear index with increased refining in the case of the mixed sheets was significant. Thus, by placing the abaca pulp in a separate layer, it is possible to increase the tensile strength by refining the softwood pulp while retaining the high tearing resistance provided by the abaca pulp.

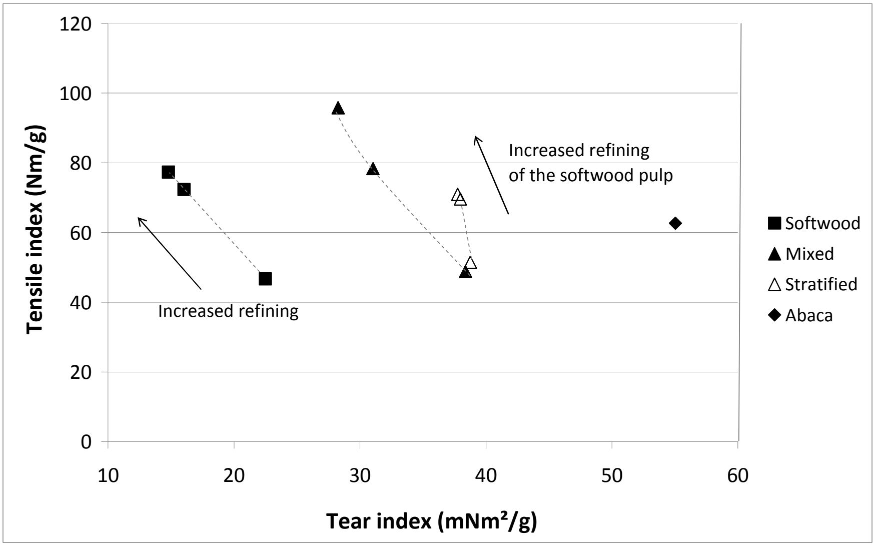


Figure 29. Tensile and tear indices of pure (100% of one pulp), homogeneously mixed and stratified sheets produced from softwood pulp with three different dewatering resistances and abaca pulp. The amount of abaca in the mixed and stratified sheets is 50%. (Results from Paper IV).

6 Materials and methods

6.1 Pulps and fibres

Paper I

Flash-dried, unbleached softwood sulphate pulp and bleached abaca neutral sulphite anthraquinone pulp sheets were used. The pulps were refined separately in a Voith LR1 conical refiner, with a cutting angle of 60° and a specific edge load of 3.0 Ws/m. The softwood pulp was refined at 240 kWh/ton and had a dewatering resistance of 27 SR. The abaca pulp was refined at 46 kWh/ton and had a dewatering resistance of 20 SR. The refined abaca pulp was dewatered and stored in a deep-freeze before use.

Paper II

Two softwood sulphate pulps, SW1 and SW2, and one hardwood sulphate pulp, HW, were used. They were collected from a local mill after refining. The hardwood pulp was refined at 88 kWh/ton and had a dewatering resistance of 21 SR, and SW1 was refined at 115 kWh/ton and had a dewatering resistance 32 SR. SW2 was refined at 128 kWh/ton. The pulps were collected at a concentration of about 3-4% and stored in a cold-storage room before use.

Paper III

Never-dried, bleached softwood sulphate pulp was collected from a local mill, after industrial refining at 110 kWh/ton. Bleached abaca neutral sulphite anthraquinone and bleached southern pine kraft pulp sheets were macerated and refined in a Voith LR1 conical refiner, with a cutting angle of 60° and a specific edge load of 3.0 Ws/m at 45 and 200 kWh/ton respectively. The abaca and southern pine pulps were fractionated on a bow screen, type ST-1, with a standard plate with 150 µm slot width to remove the fines. The dewatering resistances of the pulps were 31.5 SR for softwood and 16.0 SR for both abaca and southern pine.

Paper IV

Flash-dried, bleached softwood sulphate pulp was collected from a local mill and refined in three batches in a Voith LR1 conical refiner, with a cutting angle of 60° and a specific edge load of 3.0 Ws/m. The first batch was refined at 30 kWh/ton, the second at 135 kWh/ton and the third at 200 kWh/ton.

The dewatering resistances of the pulps were respectively 16.5, 24.0 and 33.0 SR. The abaca pulp from Paper III was used.

Paper V
Flash-dried, bleached softwood sulphate pulp (same as in Paper IV) was collected from a local mill and refined in three batches in a Voith LR1 conical refiner, with a cutting angle of 60° and a specific edge load of 3.0 Ws/m. The first batch was refined at 63 kWh/ton, the second at 123 kWh/ton and the third at 185 kWh/ton giving dewatering resistances of 19, 25 and 38 SR respectively. Commercial man-made fibres, lyocell, polyester and glass fibres, were used. The lyocell and polyester fibres were delivered with a length of 12 mm, while the glass fibres were obtained from a loosely woven fabric and cut to 12 mm.

6.2 Commercial papers

Four different types of commercial papers were collected from a local mill and used in Paper II. Comm 2, 3 and 4 were produced in a double Fourdrinier machine, where two separately formed layers are combined to a two-layer paper in the press section.

Comm 1
Single-layer, machine glazed paper of a homogeneous mixture of 50% softwood, 40% hardwood and approximately 8% broke and 2% clay. Grammage 60 g/m^2. The paper was acid-sized.

Comm 2
Two-layer sack kraft paper with softwood in both layers. Total grammage 70 g/m^2, each layer 35 g/m^2.

Comm 3
Two-layer paper with softwood in the bottom layer and a homogeneous mixture of 60% hardwood and 40% broke in the top layer. Total grammage 80 g/m^2, each layer 40 g/m^2.

Comm 4
Two-layer paper with a homogeneous mixture of 90% softwood and 10% broke in both layers. Total grammage 80 g/m^2, each layer 40 g/m^2.

6.3 Handsheet preparation

Standard isotropic sheets
Isotropic handsheets with a target grammage of 60 g/m^2 were produced according to ISO 5269-1:00 in a conventional sheet former. Standard isotropic handsheets were produced in Papers I, II and V.

PFI Sheet Former
In Paper II, isotropic handsheets were also produced in a PFI Sheet Former, producing square sheets of 22x22 cm. The sheets, with a target grammage of 60 g/m^2, were produced according to ISO 5269-1:00 except that the sheets were pressed at 224 kPa.

Dynamic sheet former
Anisotropic handsheets with a target grammage of 60 g/m^2 were produced in Paper I on a Formette Dynamique sheet former (Sauret 1971) at a rotational speed of 1200 rpm and a nozzle pressure of 2 bar. The sheets were pressed twice in a roll press, starting at 0.6 MPa followed by 1.6 MPa, and dried under restraint in an STFI plate dryer (Htun and Fellers 1982).

LB Multilayer Handsheet Former
Isotropic single-, two-, three- and four-layer sheets were produced in a multilayer handsheet former (Beghello et al. 1996). In Paper II, the forming concentration, stirring frequency and time, and the time for the decay of eddies were varied in order to study the operating function of the sheet former. In Papers III and IV, the forming concentration was 0.068 g/l, the stirring frequency 16 Hz, the stirring time 30 s for single-layer sheets and 90 s for multilayer sheets. In the rest of the sheet production, ISO 5269-1:00 was followed. See further section 2.3.1.

Handsheets for pull-out tests
In Paper V, handsheets with long man-made fibres (> 200 mm) were produced from softwood pulp in a standard handsheet former for isotropic sheets. The sheets were produced according to ISO 5269-1:00 with some modifications. A 19 mm wide tape was placed across the center of the wire screen in the sheet former. Long fibres were then placed across the tape. Stock of softwood pulp was added into the sheet former and a sheet was formed. The tape prevented fibres from forming a network in the center of the wire screen and the resulting sheet was in two parts, held together by the long fibres. See further section 3.3.1.

6.4 Test methods

The methods used for the determination of fibre, pulp and paper properties are listed in Table 2. The method used for evaluating the bond energy in Paper V is described in section 3.3.1.

The purity of each layer in the stratified and the combined sheets in Paper II was evaluated by producing two-layer sheets where one layer consisted of blue-dyed fibres. The total target grammage was 120 g/m^2, each layer having a target grammage of 60 g/m^2, and both layers were composed of HW. The sheets were cut in halves and the halves were cut clean to 150 by 228 mm in size. The sheet halves were gradually ground down, approximately 10 µm a time, one half from the top side, i.e. the white side, and the other half from the wire side, i.e. the blue side. A rotating grinding cylinder mounted on a precision holding fixture with a level adjustable to +/- 1 µm was used. The surfaces were scanned with a HP Scanjet 8200 after each grinding and image analysis in MATLAB R2007b was used to assess the proportion of dyed fibres after each grinding. A mean value for the coverage of the surface was calculated. These figures were then used to calculate the distribution of dyed fibres in the z-direction of the sheet.

Table 2. Methods used for the determination of fibre, pulp and paper properties.

Property	Method	Paper
Air permeance, Gurley	SCAN-P 19:78	I
β- formation	Ambertec Beta Formation Tester	II, V
Breaking load, single fibres	Zwick Tensile Tester (3 mm/min, 50 mm clamp distance)	V (man-made fibres)
Coarseness	STFI Fibermaster (Karlsson et al. 1999)	I, III - V
Coarseness	Weighing a known number of fibres of known length	V (man-made fibres)
Dewatering resistance	ISO 5267-1:00	I - V
Fibre diameter	Microscopic analyses	V (man-made fibres)
Fibre length	PQM1000 (Heikkurinen and Leskelä 1998)	I
Fibre length	STFI Fibermaster (Karlsson et al. 1999)	II - V
Fibre wall thickness	STFI Fibermaster (Karlsson et al. 1999)	III
Fibre width	STFI Fibermaster (Karlsson et al. 1999)	III, IV
Folding endurance	ISO 5626:93	I
Fracture toughness	SCAN-P 77:95	I
Kappa number	ISO 302:04	I
Shape factor	STFI Fibermaster (Karlsson et al. 1999)	I, III, IV
Stiffness, single fibres	Zwick Tensile Tester (3 mm/min, 50 mm clamp distance)	V (man-made fibres)
Stretch-at-break	SCAN-P 67:93	II
Strain-at-break, single fibres	Zwick Tensile Tester (3 mm/min, 50 mm clamp distance)	V (man-made fibres)
Tearing resistance	ISO 1974:90	I, III - V
Tensile strength	SCAN-P 67:93	I, II
Tensile strength	ISO 1924-3:05	III - V
Tensile stiffness	SCAN-P 67:93	I, II
Tensile energy absorption	SCAN-P 67:93	I
Thickness	SCAN-P 88:01	I, III, IV
Thickness	ISO 534:2005	II
Water retention value	SCAN-C 62:00	I, III
Wet fibre flexibility	Steadman-Mohlin (Steadman and Young 1978)	I, III
Viscosity	ISO 5351:04	I
Z-directional tensile strength	TAPPI 541 om-89	II, III, IV
Zero-span tensile strength	TAPPI 231 cm-85	I, III - V

7 Summaries of the papers

7.1 Paper I

- *Abaca as a reinforcement fibre for softwood pulp*

In this study, the effects of adding abaca (*Musa Textilis*) fibres to a softwood kraft pulp were investigated. Abaca fibres were chosen because of their great fibre length and fibre strength. Properties of the pulps used in the study are listed in Table 3.

Table 3. Properties of the pulps used in the study. (Reprinted from Paper I).

Pulp	Softwood	Abaca
Mean fibre length, mm	2.05	3.20
Coarseness, µg/m	173	155
Curl index, %	16.8	22.7
Wet fibre flexibility index	37.2	37.1
Water retention value, g/g	1.17	1.26
Zero-span tensile index, Nm/g	120	155
Kappa number	28.3	7.01
Viscosity, ml/g	1240	925
Initial drainage resistance, SR	12.4	17.1
Fibre diameter, µm	25-30 [1]	19 (6-53) [2]
Cell wall thickness, µm	2.2 [1]	4.4 (1.6-16) [2]
Lumen width, µm	10-30 [1]	12 (1-33) [2]

[1] Literature values from (Varhimo and Tourminen 1998; Fellers and Norman 1998)
[2] Literature values from (Estudillo and Torres 1997)

The beatability of the two pulps was examined. It was observed that to reach a given dewatering resistance, abaca required less refining energy than softwood, as shown in Figure 30. The tensile index of the refined pulps is shown in Figure 31 and it was observed that at a dewatering resistance in the range of 20-25 SR, the two pulps had approximately the same tensile index, while at a higher dewatering resistance abaca had a higher tensile index. Abaca showed a remarkably high initial tearing resistance and, although it rapidly decreased with refining up to about 35 SR, the tear index of abaca was well above that of softwood, as shown in Figure 32.

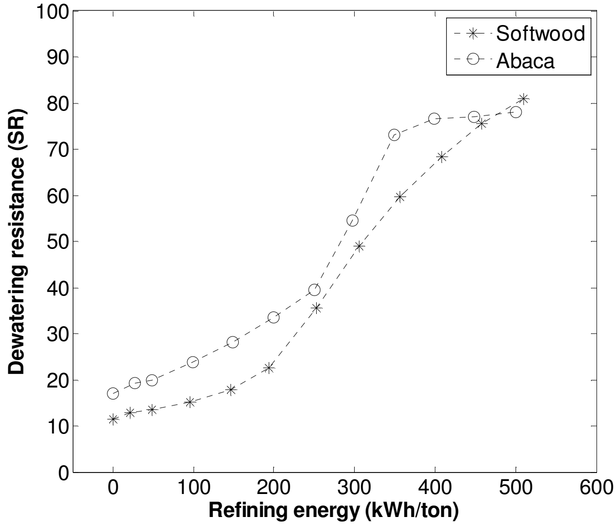


Figure 30. Dewatering resistance as a function of refining energy for softwood and abaca pulps. The pulps were refined in an Escher-Wyss refiner with a cutting angle of 60° and a specific edge load of 3.0 Ws/m. (Reprinted from Paper I).

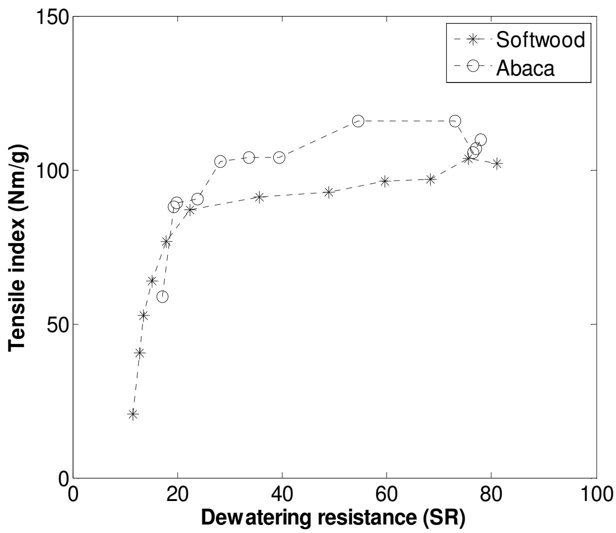


Figure 31. Tensile index plotted against dewatering resistance for the softwood and abaca pulps used in Paper I. (Reprinted from Paper I).

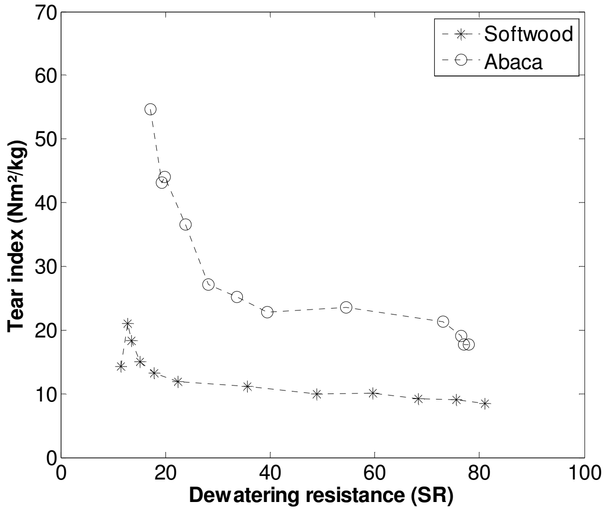


Figure 32. Tear index plotted against dewatering resistance for the softwood and abaca pulps used in Paper I. (Reprinted from Paper I).

In order to study the effects on paper strength of the addition of abaca, isotropic handsheets with various abaca contents were made in a conventional handsheet former. The density of the sheets decreased with the addition of abaca, as shown in Figure 33, probably because of the low conformability and low collapsibility of the abaca fibres. The decrease in density also indicates a decrease in bonded area and this is probably the reason for the slight decrease in tensile index with the addition of abaca, as shown in Figure 34. The tearing resistance of well-bonded sheets depends mostly on the fibre strength, but the tearing resistance of sheets with a lower degree of bonding depends on the fibre length. This might explain the deviation from the results predicted by linear mass fraction additivity. When the amount of abaca is low the fibre network is well-bonded and the abaca fibres do not contribute as much to the tearing resistance. When the amount of abaca fibres becomes greater, the network becomes more loosely bonded and the abaca fibres are able to give a larger contribution to the tearing resistance. This is also reflected in the increase in tear index with increasing abaca content, the increase in tear index was most significant with the higher additions of abaca where the density was lower.

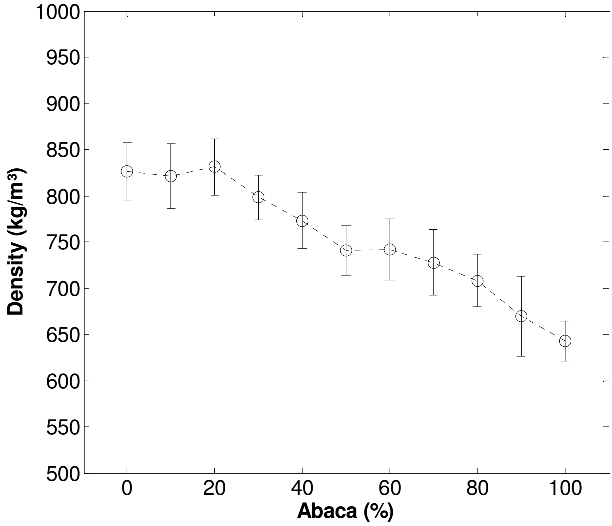


Figure 33. Density for mixed isotropic sheets of abaca and softwood as a function of abaca content. (Reprinted from Paper I).

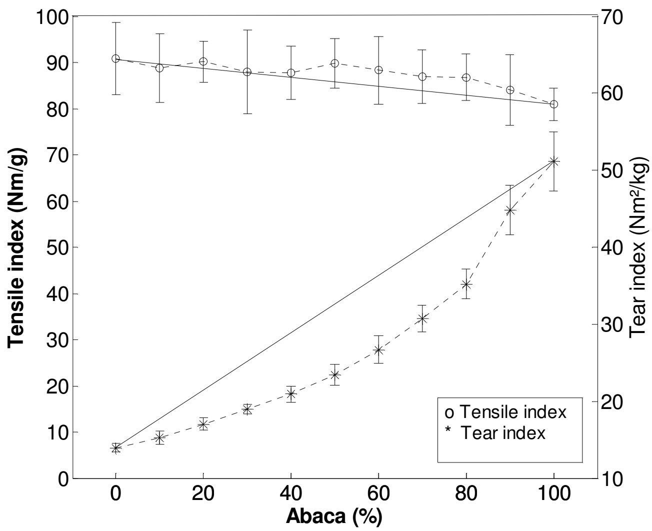


Figure 34. Tensile and tear indices plotted against abaca content for sheets consisting of abaca and softwood pulps. The lines represent linear mass fraction additivity. (Reprinted from Paper I).

Fracture toughness index, folding endurance and air permeance were found to increase with increasing abaca content, whereas tensile stiffness index and tensile energy absorption showed a slight decrease.

The most important conclusion of this study is that abaca fibres can be added to a softwood pulp to increase tearing resistance, fracture toughness, folding endurance and air permeance, but that the tensile strength, tensile stiffness and tensile energy absorption are slightly decreased. It is however possible to add up to about 60% abaca without any great loss in tensile strength. As an example, with the addition of 30% abaca, the tear index was increased by 36% while the tensile index was decreased by 8%. These main findings are based on data from isotropic as well as from anisotropic sheets produced in a dynamic sheet former. An increase in pressure in the press section could perhaps compensate for the decrease in density and thus prevent the decrease in the tensile properties. However, to obtain the positive effects of the fibre length of abaca, the density should not be too high.

7.2 Paper II

- *Handsheet former for the production of stratified sheets*

In this paper a handsheet former for the production of stratified sheets, the *LB Multilayer Handsheet Former*, was presented and evaluated. The utility of the sheet former was demonstrated and its performance in relation to conventional handsheet formers was studied. The operating function of the sheet former was optimized and the quality of the sheets produced analysed. The sheet former was originally developed at Åbo Akademi and has been used in previous studies (Beghello et al. 1996). The basic idea was to create a multilayer sheet former for laboratory use that is easy to handle and has a high sheet producing rate. The sheets produced are to be isotropic, so that paper properties can be studied in the absence of effects of fibre orientation. A certain degree of interlayer mixing should produce sheets with a high ply-bond strength.

The basic design of the multilayer handsheet former is shown in Figure 8, section 2.3.1. The pulp container can be divided into two, three or four compartments using sliding plates. Each compartment has a volume of 15 dm^3 and is equipped with a propeller for stirring the pulp. The former is drained downwards and the sheet is formed on a metallic wire screen supported by a 10 mm thick plate with drilled holes.

To start the production of handsheets, water should be added up to a level approximately 50 mm above the wire in order to prevent fibres becoming stuck on the wire screen. The bottom-layer pulp is then added in the top of the former and the lower compartment is filled with water. The first divider plate is pushed into the former to seal the bottom compartment. The second-layer pulp is added and the compartment is filled with water up to the next divider plate. For additional layers, the procedure is repeated. When the desired number of compartments are filled with pulp, the stirring is started. After stirring has been completed, the divider plates are steadily pulled out in succession from the top to the bottom and the valve to the drainage pipe is opened. To promote further dewatering of the sheet, after the drainage is finished, blotters are placed on top of the formed sheet and a couch-weight is placed on the top. The sheet can then be wet-pressed and dried under standardized conditions.

Single-layer sheets, stratified sheets and multilayer sheets composed of single-layer sheets combined in the press were produced in the LB Multilayer Sheet Former. The uniformity of the sheets was analysed and compared to that of sheets made in conventional sheet formers, and to that of commercially produced single-layer and two-layer papers.

To evaluate the uniformity of the sheets, several strips were cut from a single sheet, paper properties were measured and a within-sheet coefficient of variation was calculated. The variation of properties of single-layer and stratified sheets of hardwood is shown in Figure 35. For grammage and tensile strength, the uniformity of the sheets seems to be unaffected by the number of layers. However, for thickness and tensile stiffness, the variation increased with increasing number of layers in the sheet. Stretch-at-break showed a divergent result with the greatest variation in the single-layer sheets.

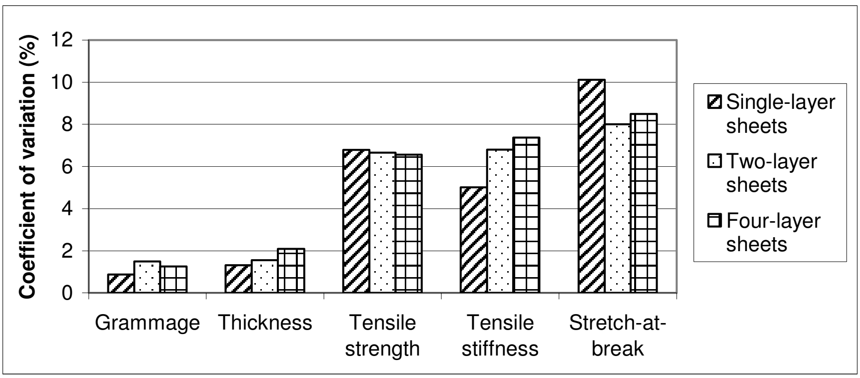


Figure 35. Coefficient of variation of properties of single-layer and stratified sheets of hardwood. (Reprinted from Paper II).

The β-formation index of sheets produced with different forming methods is listed in Table 4. ISO are isotropic sheets produced in a standard handsheet former. PFI are sheets produced in a PFI Sheet Former. Comm 1 and Comm 3 are commercially produced sheets. It can be seen that the LB Multilayer Handsheet Former produced sheets with a better formation than the other sheets, except for those produced in the fully automatic PFI Sheet Former.

Table 4. β-formation index for sheets produced with different forming methods. The handsheets were single-layer sheets produced with the forming concentration 0.15 g/l. (Reprinted from Paper II).

Sheet forming method	Fibre composition	β-formation index (√g/m)
LB Multilayer Handsheet Former	100% hardwood, HW	0.32
LB Multilayer Handsheet Former	100% softwood, SW1	0.47
ISO	100% hardwood, HW	0.38
ISO	100% softwood, SW1	0.70
PFI	100% hardwood, HW	0.26
PFI	100% softwood, SW1	0.42
Comm 1	50% softwood 40% hardwood 8% broke	0.84
Comm 3	Bottom layer: softwood Top layer: 60% hardwood 40% broke	0.79

Figure 36 shows the within-sheet coefficient of variation of properties of papers made in different handsheet formers and of commercial papers. In the case of grammage and thickness, the LB Multilayer Handsheet Former produced the most uniform sheets. In the case of tensile strength, tensile stiffness and stretch-at-break, the variation of the sheets made in the LB Multilayer Handsheet Former was somewhat greater than that of the sheets produced in the ISO former and in the PFI Sheet Former.

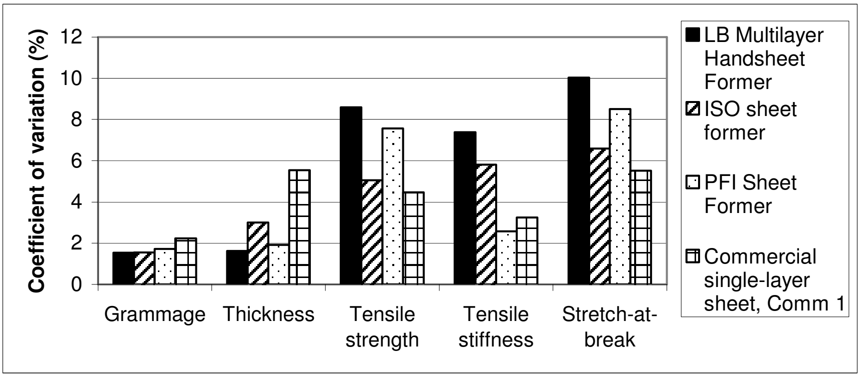


Figure 36. Coefficient of variation of properties of sheets produced by different sheet forming methods. The handsheets were all single-layer sheets produced of hardwood with the forming concentration 0.15 g/l. (Reprinted from Paper II).

The main conclusion from the study is that the LB Multilayer Handsheet Former is a suitable tool for studying the effects of sheet stratification on paper properties. The sheet former is easy to handle, and single-, two-, three- or four-layer sheets can be produced efficiently at a high sheet production rate. The sheet former produces isotropic sheets, which means that the effects of the fibre orientation are excluded when studying the paper properties. The sheet former produces sheets with good formation, and the variation in the properties of the sheets remained at a fairly constant level when the number of layers in the stratified sheets was increased. The variation in properties of the sheets made in the LB Multilayer Handsheet Former is generally at the same level as of those made in conventional sheet formers. The variation in the sheets can probably be reduced by better alignment of the wire.

7.3 Paper III

- ***Paper strength of both stratified and homogeneous sheets with selected fibres – Part I: Effect of fibre properties***

In Paper III, the effect on paper strength of placing different types of fibre in separate layers instead of homogeneously mixing the fibres was examined. The aim was to displace the tensile-tear relationship towards higher strength values. Two-layer laboratory sheets were made in the LB Multilayer Handsheet Former and compared to single-layer sheets made from a homogeneous mixture of the two pulps. Fibres with high coarseness (southern pine) and fibres with long fibre length (abaca) were added to a Swedish softwood kraft pulp. To intensify the special properties of the two selected fibres, i.e. the fibre length of abaca and the coarseness of southern pine, the amount of fines in the pulps was reduced by fractionation. The properties of the pulps are listed in Table 5. The compositions of the sheets used in the study are given in Table 6.

Table 5. Properties of the pulps used in the study. (Reprinted from Paper III).

Pulp	Swedish Softwood	Southern Pine	Abaca
Mean fibre length, mm	2.00	2.27	3.56
Fibre width, µm	27.5	30.8	16.2
Fibre wall thickness, µm	9.3	11.3	6.0
Coarseness, µg/m	165	206	146
Shape factor, %	85.1	84.5	80.9
Wet fibre flexibility index	65	58	61
Water retention value, g/g	1.98	1.58	1.39
Dewatering resistance, SR	31.5	16.0	16.0
Zero-span tensile index, Nm/g	126	115	156

As indicated by fibre width, coarseness and fibre wall thickness, the most collapsible fibres should be the Swedish softwood fibres, followed by the southern pine fibres. The wet fibre flexibility index indicates the conformability of the fibres, whereas the water retention value indicates the swelling ability, and a high degree of swelling promotes fibre-fibre bonding. Thus, the figures shown in Table 5 suggest that the softwood fibres should form a dense network with a high bond density, while networks of southern pine and abaca fibres should be less dense and exhibit a lower bond density. Further, the softwood pulp includes fines which increase the bond density.

Table 6. Sheet IDs and composition of the sheets used in the study. (Reprinted from Paper III).

Sheet ID	Pulp(s)	Nr of layers	Target grammage (g/m^2)
S30	Softwood	1	30
S60	Softwood	1	60
SP30	Southern Pine	1	30
SP60	Southern Pine	1	60
A30	Abaca	1	30
A60	Abaca	1	60
S+SP	Softwood and Southern Pine	1	60
S+A	Softwood and Abaca	1	60
S/SP	Softwood and Southern Pine	2	60
S/A	Softwood and Abaca	2	60

Figure 37 show the tensile index values for the different sheets. The tensile index was higher in the mixed sheets than in the stratified sheets. The mixed sheets containing abaca had a higher tensile index than those containing southern pine, while the two different kinds of stratified sheets had approximately the same strength. The tensile index of the pure softwood sheets was at the same level as that of the mixed sheets, while that of both the pure southern pine and the abaca sheets was somewhat lower than that of the stratified sheets. Thus, the less conformable fibres did not seem to impair the bonding between the softwood fibres. It is suggested that the load-bearing capacities of the less collapsed southern pine and abaca fibres are activated to a greater extent when they are mixed with softwood fibres. In the network together with the more flexible and conformable softwood fibres, the number of fibre-fibre contacts increased, increasing the number of bonds in the sheet. Moreover, the fines in the softwood pulp probably contribute to the bonding in the sheet. With the higher degree of activation of the southern pine and abaca fibres, their individual fibre strengths, indicated by the zero-span tensile index, should have a positive effect on the tensile strength of the sheets.

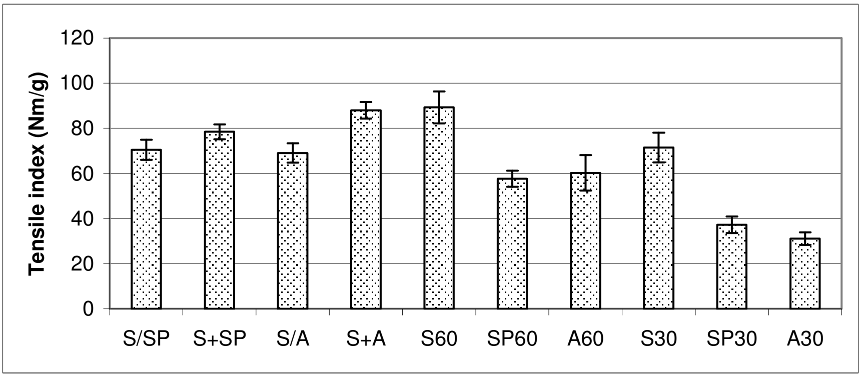


Figure 37. Tensile index for the single-layer and the stratified two-layer sheets. Sheet IDs are given in Table 6. (Reprinted from Paper III).

In the case of the tearing resistance, the relation between the mixed and stratified sheets was the reverse, as shown in Figure 38. The stratified sheets showed a higher tear index than the mixed sheets. Furthermore, the pure abaca sheets had a remarkably high tear index resulting in a high tear index for both the mixed and the stratified sheets containing abaca. Tearing resistance is, to a high degree, promoted by fibre length and the results clearly show that the long abaca fibres contributed to a high tear index. Tearing resistance has been shown to decrease with increasing fibre-fibre bonding (Yu 2001) and the lower tear index for the mixed sheets is most likely a result of the higher bonding level discussed above.

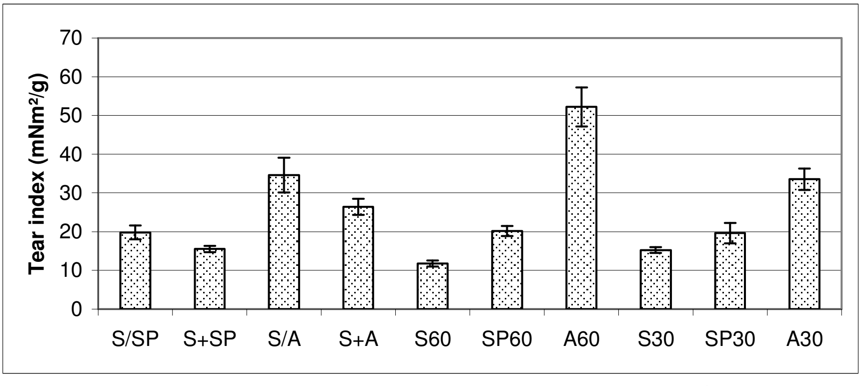


Figure 38. Tear index for the single-layer and the stratified two-layer sheets. Sheet IDs are given in Table 6. (Reprinted from Paper III).

The effect of stratification on the tensile-tear relationship is shown in Figure 39. The result predicted by linear mass fraction additivity is indicated. As could be deduced from the results reported above, the relation was shifted towards a higher tear index, but with a lower tensile index for the stratified sheets than for the mixed sheets. It can be seen that the sheets containing southern pine show strengths close to the predicted ones, although the stratified sheets showed a somewhat higher tear index than predicted. In the case of the sheets containing abaca, the predicted values lie within the limits for the stratified sheet, while the mixed sheets deviate somewhat showing a higher tensile index but lower tear index. The reason for the deviating result is probably to be found in the bond density. When the abaca fibres are activated, they contribute to the tensile strength resulting in a synergy, whereas they do not contribute to the tearing resistance, showing a lower resistance than predicted.

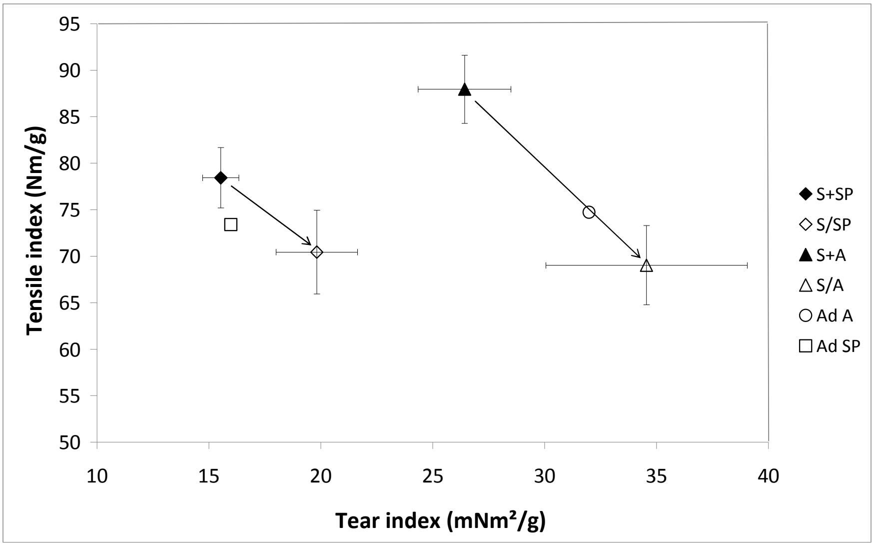


Figure 39. Tensile index plotted against tear index for the mixed and stratified sheets. Ad SP and Ad A indicate the results predicted by linear mass fraction additivity. Sheet IDs are given in Table 6. (Results from Paper III).

The main conclusion from this study is that by making sheets with one layer of long fibres and a lower degree of bonding, and a second layer with well-bonded fibres, the tensile-tear relationship can be shifted towards a higher tear index, but with a lower tensile index than in sheets of a homogeneous mixture of the two pulps.

7.4 Paper IV

- ***Paper strength evaluation both in stratified and in homogeneous sheets with selected fibres – Part II: Effect of refining***

The effect on the tensile strength and tearing resistance of refining one of the pulps in mixed and stratified sheets consisting of two pulps was examined. The aim was to see whether stratification has a positive effect on the tensile-tear relationship, or whether the same effect can be reached by adjusting the degree of refining of the pulps. Moderately refined, fractionated abaca (*Musa textilis*) pulp with a high tearing resistance potential was added to a Swedish softwood kraft pulp. The softwood pulp was refined to three different dewatering resistances: 16.5, 24.0 and 33.0 SR. Two series with different amount of abaca, 50% and 17%, were produced. A third series was also produced using the softwood of 24.0 SR, where the amount of abaca was increased in steps corresponding to 10 g/m^2.

In the case of the sheets composed of 50% of each pulp, the mixed and the stratified sheets showed different responses to the increased refining of the softwood pulp, as shown in Figure 40. The mixed sheets behaved as the pure softwood sheets, showing about the same gradient in the tensile-tear relationship with increased refining. The tensile index increased as a consequence of the increased bond density caused by the increased refining. However, the higher degree of bonding together with fibre shortening caused a decrease in tearing resistance. In the mixed sheets containing abaca, these long fibres contributed less and less to the tearing resistance, the higher the degree of refining of the softwood pulp. A synergy for the tensile index of the mixed sheets was observed, as in Paper III. In the sheets with the more well-refined softwood pulp, the abaca fibres are activated to a higher extent and thus contribute more to the tensile strength. The stratified sheets, like the mixed sheets and the pure softwood sheets, also showed an increase in tensile index with increased refining, but the tear index seemed to be unaffected. The tearing resistance of the abaca fibres was considerably higher than that of the softwood fibres, so that the abaca layer provided the greatest resistance to crack propagation and dominated the result. The tearing resistance of the stratified sheet was therefore expected to remain at the same level even if the tearing resistance of the softwood layer is decreased. There is no significant variation, at a 5% statistical level, in the tear index of the stratified sheets with increasing refining, although the decrease in tear index with increasing refining in the case of the mixed sheets was significant. Thus, by placing the abaca pulp in a separate layer, it is possible to increase the tensile strength by refining the softwood pulp while retaining the high tearing resistance provided by the abaca pulp.

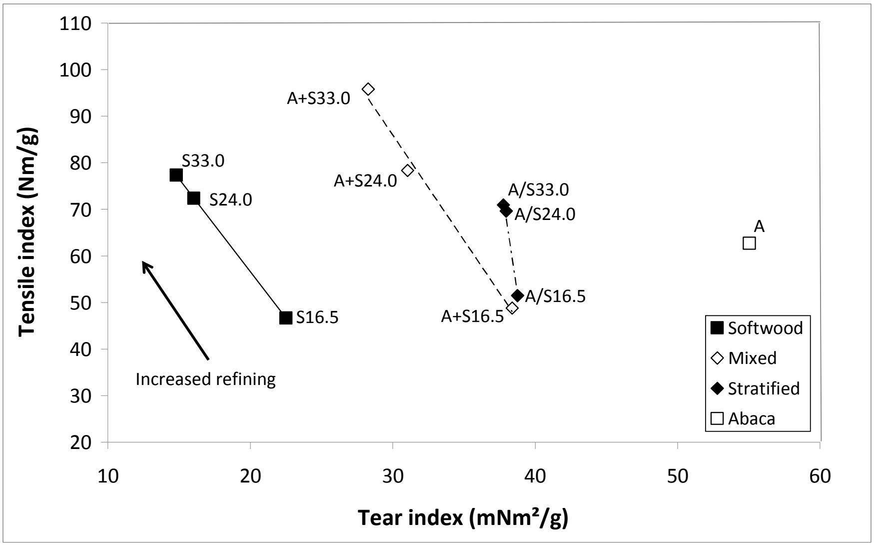


Figure 40. Tensile and tear indices of pure (100% of one pulp), homogeneously mixed and stratified sheets produced from softwood pulp with three different dewatering resistances and abaca pulp. The dewatering resistance of the softwood pulps is indicated with figures, i.e. 16.5, 24.0 and 33.0 SR. The amount of abaca in the mixed and stratified sheets is 50wt%. Trend lines are added for clarity. (Reprinted from Paper IV).

Figure 41 shows the tensile-tear relationship for the mixed and stratified sheets consisting of 83% softwood with different degree of refining and 17% abaca. There is no significant difference between the mixed and the stratified sheets, indicating that, for this amount of abaca, there is no positive effect of stratifying the sheets. The tensile index decreased and the tear index increased with decreasing refining in the stratified sheets, the pure softwood sheets and the mixed sheets. The values for the stratified sheets closely followed the prediction of linear mass fraction additivity, while the mixed sheets showed a slightly lower tear index and, for the intermediate and highest degrees of refining, a somewhat higher tensile index than predicted. It is suggested that the amount of abaca is so low that the fibres are more firmly bonded in the fibre network, i.e. each abaca fibre is surrounded by a larger proportion of the more flexible softwood fibres, and thus the average number of fibre-fibre bonds on each abaca fibre should be greater than in the sheets with a larger amount of abaca. This implies that the abaca fibres in the mixed sheets contribute less to the tearing resistance and correspondingly that the tensile index of the mixed sheets is higher than expected. The thin layer of abaca in the stratified sheets seemed to be insufficient to give a greater tearing resistance than that of the mixed sheets.

The tensile-tear relationship for the mixed and the stratified sheets produced from different compositions of softwood and abaca pulp is shown in Figure 42. Based on previous results, an increase in the tearing resistance and a decrease in the tensile strength when the sheet was stratified, i.e. when the pulps were placed in separate layers, were expected. The sheets with the lowest amount of abaca showed this behaviour, while the sheets with the highest amount of abaca showed the opposite behaviour. When the amount of abaca fibres is as large as 87% the fibre network is dominated by the abaca fibres and it is not expected that stratification will have a great effect on the paper strength properties.

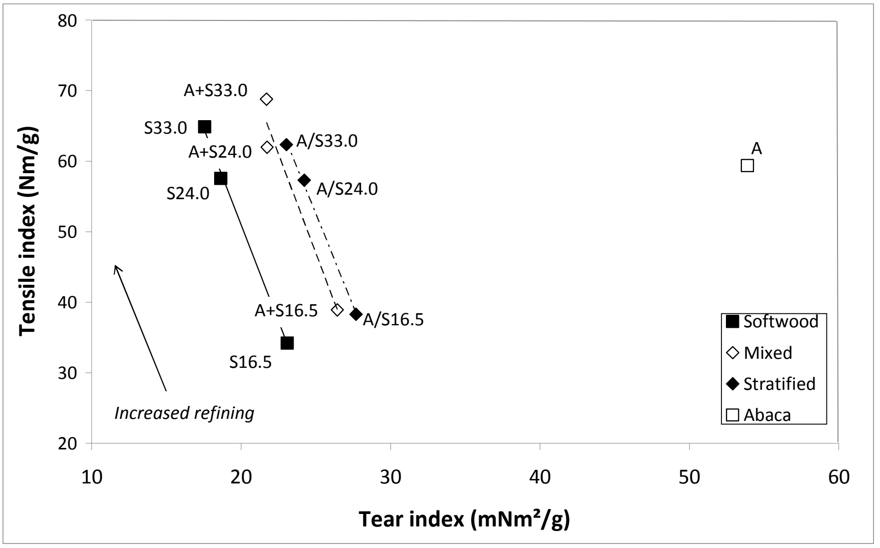


Figure 41. Tensile and tear indices for pure, homogeneously mixed and stratified sheets produced from softwood pulp with three different degrees of refining and abaca pulp. The dewatering resistance of the softwood pulps is indicated with figures, i.e. 16.5, 24.0 and 33.0 SR. The amount of abaca in the mixed and stratified sheets is 17%. Trend lines are added for clarity. (Reprinted from Paper IV).

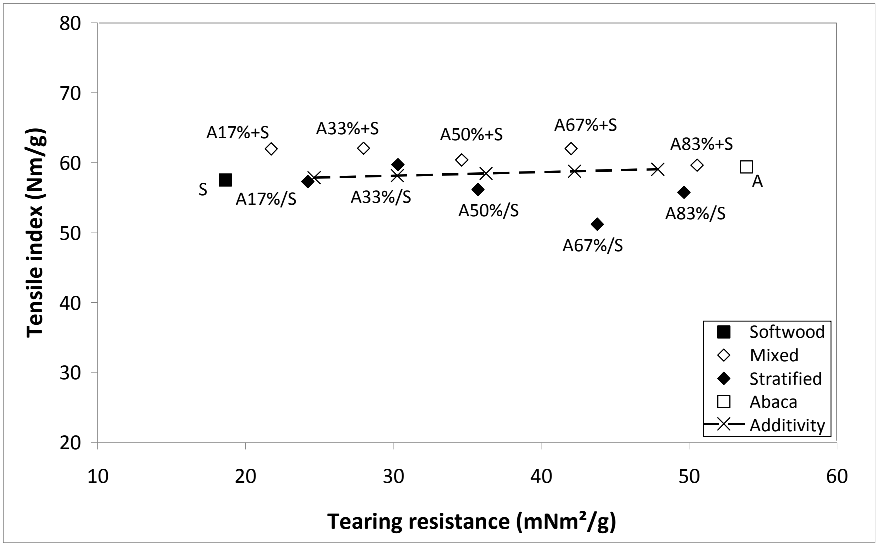


Figure 42. Tensile and tear indices for pure, homogeneously mixed and stratified sheets produced from different compositions of softwood and abaca pulp compared to the results predicted by linear mass fraction additivity. The studied amounts of abaca were 0, 17%, 33%, 50%, 67%, 83% and 100% and the dewatering resistance of the softwood pulp was 24.0 SR. The results predicted by linear mass fraction additivity are indicated by crosses. (Reprinted from Paper IV).

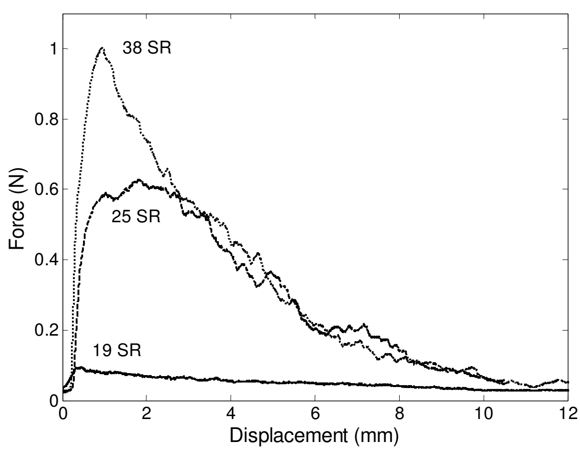
The main conclusions from this study is that, by stratifying a sheet composed of 50% of a pulp with a high tensile strength potential (softwood kraft) and 50% of a pulp with a high tearing resistance potential (abaca), it is possible to increase the tensile index by refining the first pulp while retaining the tear index. The stratification thus has a different effect on the tensile-tear relationship than the degree of refining.

7.5 Paper V

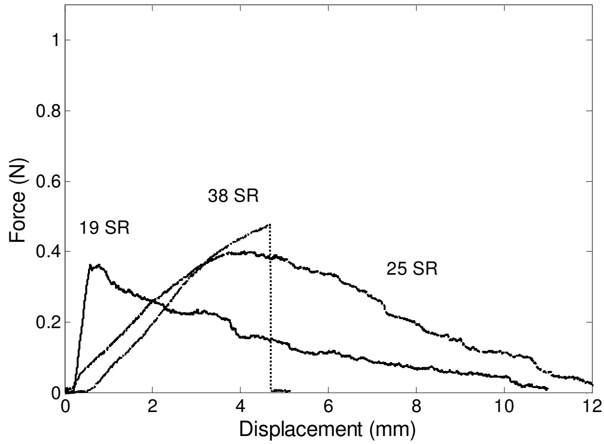
- ***The effect on paper strength of man-made fibres added to a softwood kraft pulp***

The effect on paper strength of incorporating man-made fibres with different abilities to create chemical and mechanical bonds with a softwood kraft pulp was evaluated. Three different man-made fibres were used: lyocell, polyester and glass fibres, and the bonding ability was evaluated by pull-out tests. The fibres were added to softwood pulp of different dewatering resistances: 19, 25 and 38 SR. The fibres were added in amounts of 1, 3 and 5%.

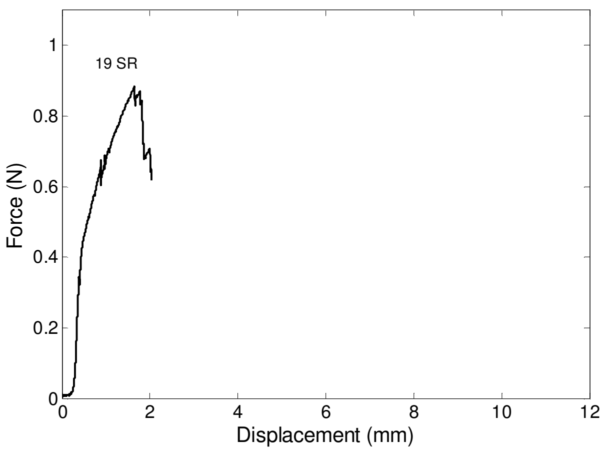
The pull-out tests were performed by pulling a long man-made fibre out of the softwood network in a tensile tester at low speed. Figure 43 show representative curves for the fibres embedded in handsheets produced from the three softwood pulps respectively. As expected increased refining of the softwood meant that a greater force was required to pull out the fibres. The glass fibres were pulled out intact from all sheets while about 50% of the polyester fibres were broken in the 25 SR sheets and 90% in the 38 SR sheets. All of the lyocell fibres were broken in the 19 SR sheets and no tests were performed for the pulps of higher resistances. This indicates that the lyocell fibres were most strongly bonded in the softwood network, while the glass fibres were most loosely bonded.



a



b



c

Figure 43. Force-displacement curves for single fibre pull-out tests of a: Glass fibre, b: Polyester and c: Lyocell. The dewatering resistance of the softwood pulp is indicated by figures. (Reprinted from Paper V).

Figures 44, 45 and 46 show the tear index and figures 47, 48 and 49 show the tensile index of sheets produced from the three softwood pulps with different amounts of man-made fibres added. The tensile index decreased slightly with the addition of man-made fibres for all three fibre types. Glass fibres seem to affect the tensile index least. This might partly be explained by the fact that the specific breaking load of glass fibres was the highest among the fibres evaluated. Further, the glass fibres have a higher coarseness meaning that a sample of a certain weight contains fewer fibres than a sample of polyester, or especially of lyocell fibres, of the same weight. This fact, along with the smaller fibre diameter of the glass fibre compared to that of the polyester, suggests that the glass fibres affect the softwood fibre network less than the other two fibre types and that the strength of the softwood network should to a great extent be retained.

In the case of the tear index, the effects of adding glass fibers and polyester fibres are similar, while the lyocell fibres made a considerably lower contribution to the strength. The reason for the lower tear index for the sheets with lyocell is probably a combination of the low breaking load of the fibres and their firm attachment in the softwood network. The low bonding ability and high specific breaking load of the glass fibres was expected to give sheets of higher tear index than those containing polyester. The glass fibres were however more brittle and less extensible than the polyester fibres and this might be one reason why they gave sheets with a lower tear index when added to softwood pulp of higher dewatering resistance. In these sheets, the fracture zone should be narrower and the out-of-plane forces are distributed on shorter segments of the fibres. Further the polyester fibres had a larger diameter than the glass fibres, and this should mean that a higher force is required to pull out a fibre from the softwood network. Also, the number of polyester fibres added to the sheet at a certain percentage was larger than that of glass fibres. The positive effect of the long fibres on the tear index is thus promoted in the case of polyester.

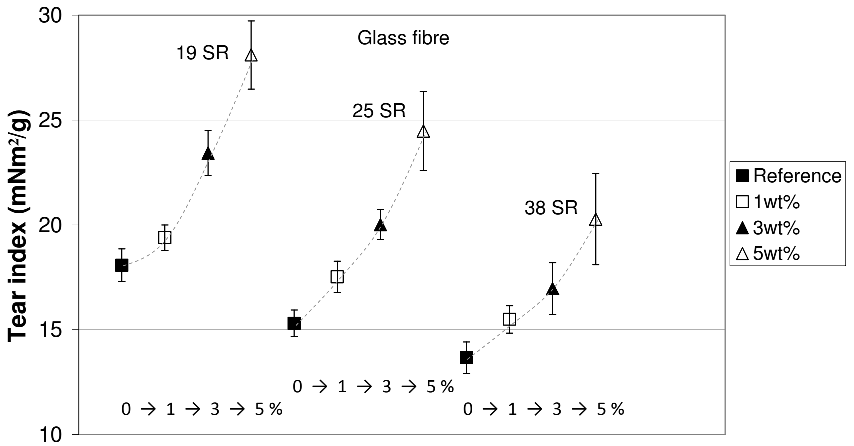


Figure 44. Tear index of handsheets made from softwood kraft pulp reinforced with glass fibres. Three different dewatering resistances were used for the softwood pulp: 19, 25 and 38 SR. (Reprinted from Paper V).

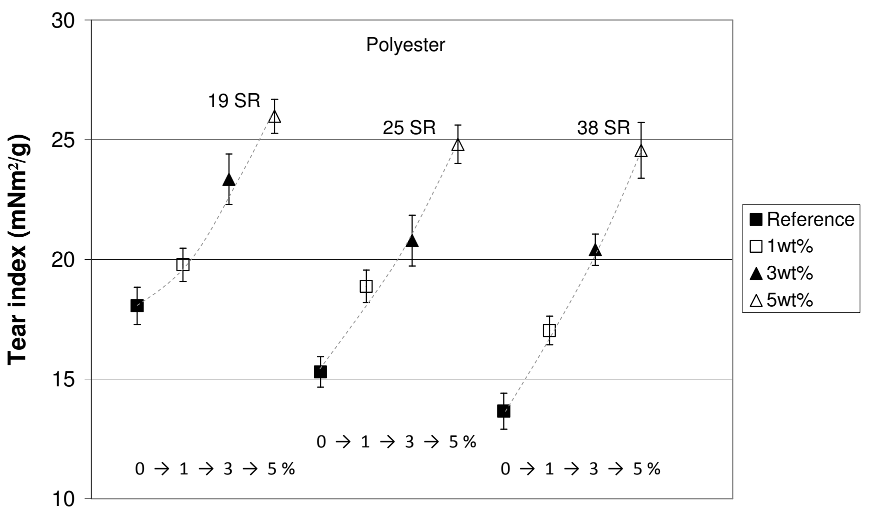


Figure 45. Tear index of handsheets made from softwood kraft pulp reinforced with polyester fibres. Three different dewatering resistances were used for the softwood pulp: 19, 25 and 38 SR. (Reprinted from Paper V).

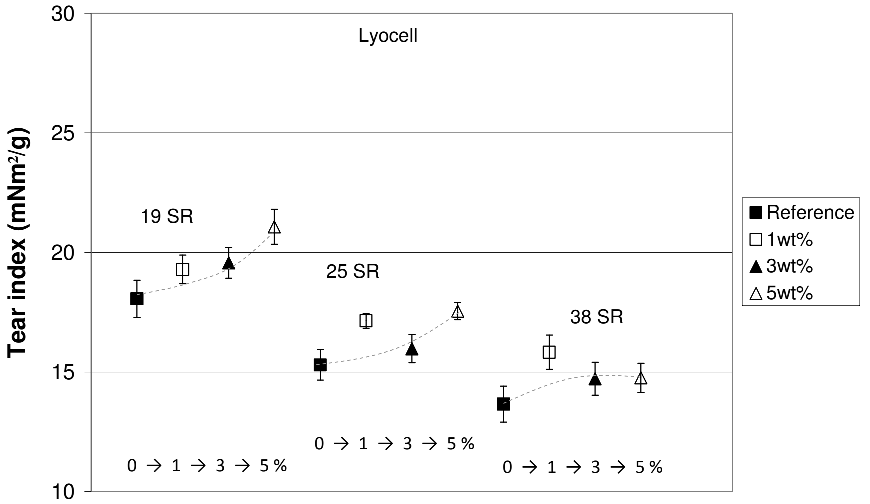


Figure 46. Tear index of handsheets made from softwood kraft pulp reinforced with lyocell fibres. Three different dewatering resistances were used for the softwood pulp: 19, 25 and 38 SR. (Reprinted from Paper V).

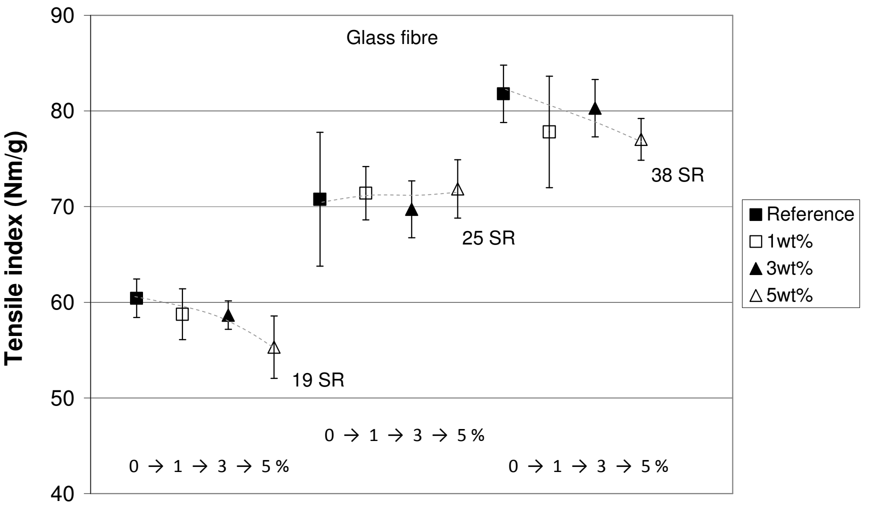


Figure 47. Tensile index of handsheets made from softwood kraft pulp reinforced with glass fibres. Three different dewatering resistances were used for the softwood pulp: 19, 25 and 38 SR. (Reprinted from Paper V).

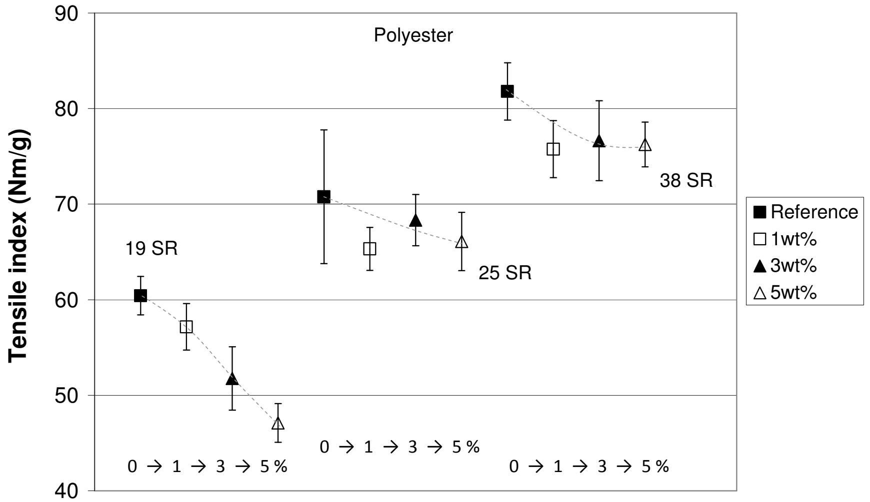


Figure 48. Tensile index of handsheets made from softwood kraft pulp reinforced with polyester fibres. Three different dewatering resistances were used for the softwood pulp: 19, 25 and 38 SR. (Reprinted from Paper V).

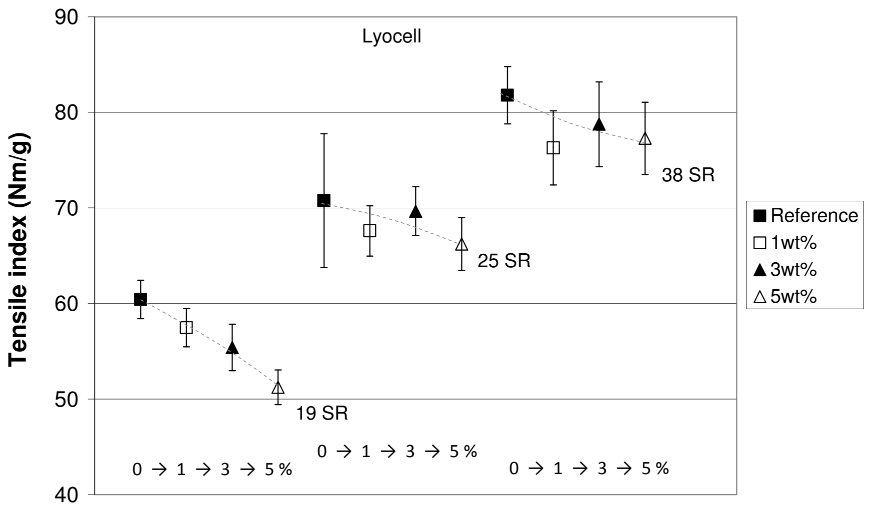


Figure 49. Tensile index of handsheets made from softwood kraft pulp reinforced with lyocell fibres. Three different dewatering resistances were used for the softwood pulp: 19, 25 and 38 SR. (Reprinted from Paper V).

The aim of this work was to move the tensile-tear relationship towards higher strength values. In Figure 50, this relationship is plotted for the softwood pulp with different degrees of refining. As expected, the tensile index was increased while the tear index was reduced with increasing refining. By adding 3% polyester the tear index of a softwood pulp refined to 25 SR is increased by 30% and the tensile index is reduced to a level corresponding to approximately 23 SR. Equally a sheet made from 38 SR softwood pulp will increase its tear index by 80% by adding 5% polyester while the tensile index corresponds to that of a pure softwood pulp refined to approximately 30 SR. It is clear from the figure that the trend is shifted from decreasing tear index to an increase in both tear and tensile indices. Thus the main conclusion from this study is that, by the refining of a softwood pulp and subsequent addition of long fibres with low bonding ability, the tensile-tear relationship can be moved towards higher strength values.

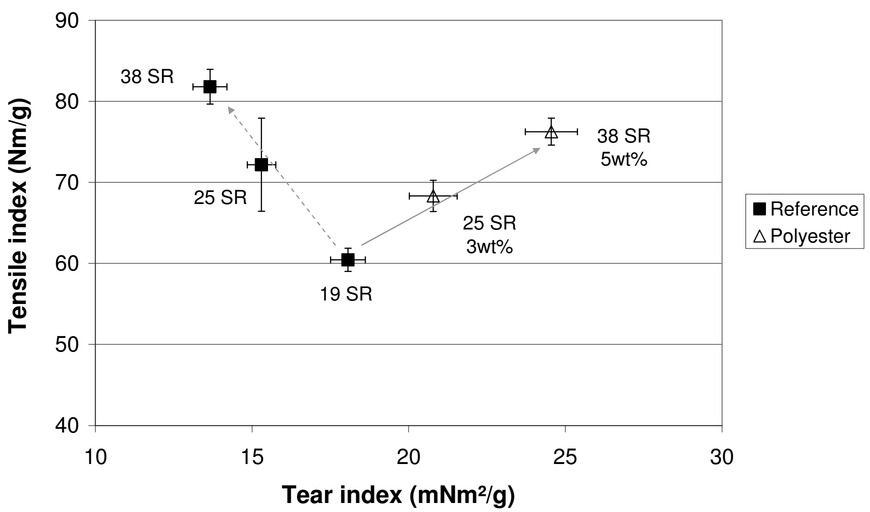


Figure 50. Tensile-tear relationship of the softwood kraft pulp with increased refining compared to increased refining and subsequent addition of polyester fibres. (Reprinted from Paper V).

8 Conclusions

- Abaca fibres can be added to a softwood pulp to increase the tearing resistance, fracture toughness, folding endurance and air permeance.

- It is possible to add up to about 60% abaca without any great loss in tensile properties as a result of the decrease in density.

- The LB Multilayer Handsheet Former can be used for studying properties of stratified sheets.

- The LB Multilayer Handsheet Former produces sheets with good formation and good uniformity in paper properties compared to conventional sheet formers and commercially produced sheets.

- By stratifying a sheet, so that fibres with a high tearing resistance potential and fibres with a high tensile strength potential are placed in separate layers, rather than by making sheets of a homogeneous mixture of the pulps, it is possible to increase the tear index of the resulting sheet by approximately 25%, whereas the tensile index decreases by 10-20%.

- By mixing a pulp with less collapsible and less conformable fibres and no fines with a pulp with fibres with higher collapsibility and higher conformability and fines, a synergism in tensile index is obtained, i.e. the mixed sheets have a higher tensile index than that predicted by linear mass fraction additivity.

- By stratifying a sheet composed of 50% of a pulp with a high tensile strength potential (softwood kraft) and 50% of a pulp with a high tearing resistance potential (abaca) it is possible to increase the tensile index while retaining the tear index by refining the first pulp.

- By refining a softwood pulp and reinforcing it with long fibres with low bonding ability, the tensile-tear relationship of sheets produced from the pulp can be moved towards higher strength values.

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