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ON THE BEATING OF REINFORCEMENT PULP

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ABSTRACT

The aim of this work was to gain a better understanding of the effect of reinforcement pulp beating on the strength of mechanical pulp-dominated paper. The main purpose of reinforcement pulp beating is to improve the runnability of paper.
The first objective of this study was to maximize the runnability related strength properties by beating. It was assumed that the flaw-resisting ability of paper correlates with the runnability of the dry paper web. In-plane fracture properties were assumed to describe the flaw-resisting ability. The second objective was to understand the mechanism how beating affects paper strength and structure (e.g. the size of the fracture process zone).

It was found that the reinforcement pulp ranking and optimisation of beating depend on the criteria used. The selection of critical strength properties affects the pulp ranking – sometimes different methods give opposite results (e.g. neutral sulphite vs. kraft). It was found that beating does not increase the in-plane fracture energy of mechanical pulp-dominated paper. Fracture toughness and elastic breaking strain increase only a little. However, tensile strength, elastic modulus and z-directional strength properties increase. It was concluded that beating does not significantly improve the flaw-resisting ability of mechanical pulp-dominated paper, while the strength of unflawed paper does increase.

It was found that the fracture properties of mixture sheets cannot be estimated based only on fracture properties of pure components. The additivity behaviour of fracture properties was strongly non-linear. The fracture energy of pulp mixtures seems to correlate mainly with the average fibre length (and not with the beating level). Beating increases the fracture energy per fracture zone area in the pure chemical pulp sheets but not in the mixture sheets. Evidently beating does not increase the energy needed to open interfibre bonds in mechanical pulp-dominated paper.

The results can be explained by the beating induced increase in both interfibre bonding and fibre segment activation. It was also found necessary to divide bonding into in-plane and out-of-plane components. The results largely support the activation theory originally presented in the 1960's. However, no direct measurement of activation was used in this study. Low-freeness mechanical pulp seems to have as high (in-plane) interfibre bonding as but lower activation than beaten reinforcement pulp. Therefore reinforcement pulp beating improves bonding-related properties such as elastic breaking strain or in-plane fracture energy only marginally. On the other hand, properties strongly dependent on activation, such as elastic modulus and tensile strength, are increased by reinforcement pulp beating.

Keywords: beating, refining, reinforcement pulp, runnability, strength, fracture toughness, bonding, activation
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PREFACE

This study was carried out at the Laboratory of Paper Technology of Helsinki University of Technology (HUT) during the years 1996-2001. This work was part of a technology program "New challenges in chemical pulping" by the National Technology Agency, TEKES, Finland. That program was later part of Finnish Forest Cluster Research Programme (WOOD WISDOM 1998-2001). The study has also received financial support from the following foundations: Tekniikan edistämissäätiö, Walter Ahlströmin säätiö and Teknillisen korkeakoulun tukisäätiö. The financial support of each of these is very much appreciated.

I would like to express my gratitude to my supervisor, Professor Hannu Paulapuro, for his supports and advice during the course of this work. He has created the high-level research "infrastructure" to the Laboratory of Paper Technology and furthermore has allowed me to work here for more than a decade. Special thanks go to Professor Jaakko E. Laine who was my boss during the project. His advices, which were on both principal and practical level, helped me to get through this.

I wish to thank my co-authors for the great co-operation in the project work: Heikki Kettunen, Gongde Zhang, Yongzhong Yu, Atsushi Tanaka and Kaarlo Niskanen. It is difficult to imagine a better team for such a task. The staff of the Department of Forest Products Technology at HUT is thanked for all kinds of help.

Finally, I would like to thank Leena for all the love and encouragement and especially for Ilmari.

Otaniemi, January 2003

Eero Hiltunen

LIST OF PUBLICATIONS

The thesis consists of this summary and of eight publications.

I. Hiltunen, E., Laine, J.E., Zhang, G., The effect of refining chemical softwood pulp on the strength properties of a sheet made from a mixture of mechanical and chemical pulp. 52nd Appita Annual General Conference Proceedings. Brisbane, Australia, May 11-14, 1998. Appita, Carlton, 1998. Vol 1, 239-245.

II. Hiltunen, E., Kettunen, H., Laine, J.E., Paulapuro, H., Effect of softwood kraft refining on a mechanical-chemical mixture sheet. Tappi Journal, 83(2000)10, p 67, full text version in http://www.tappi.org, 9 pages.

III. Hiltunen, E., Kettunen, H., Laine, J.E., Paulapuro, H., Effects of softwood kraft pulp refining on fracture behaviour of TMP-Based paper. Paper Technology 43(2002)7, 35-39.

IV. Hiltunen, E., Kettunen, H., Laine, J.E., Paulapuro, H., Behaviour of Reinforcement Fibres in TMP-Based Paper. Paperi ja Puu 84(2002)4, 269-273.

V. Zhang, G., Hiltunen, E., Laine, J.E., Paulapuro, H., Kettunen, H., Niskanen, K., Comparison of the effects of wet straining and refining on the fracture properties of paper. Nordic Pulp and Paper Research Journal 17(2002)1, 45-49.

VI. Tanaka, A., Hiltunen, E., Kettunen, H., Niskanen, K., Fracture properties in Filled Papers. The Science of Papermaking, Transactions of the 12th Fundamental Research Symposium. Oxford, England, 17-21 September 2001. FRS, Frecheville Court, 2001. Vol 2, 1403-1421.

VII. Yu,Y., Kettunen, H., Hiltunen, E., Niskanen, K., Comparison of abaca and spruce as reinforcement fibre. Appita Journal 53(2000)4, 287-291.

VIII. Hiltunen, E., Laine, J.E., Paulapuro, H., Effect of fibre charge on refining of softwood kraft pulp. PTS-Symposium 1999 PAPIERFASERSTOFF-TECHNIK: Brachliegende Potentiale beim Einsatz von Faser- und Hilfstoffen. Dresden, Germany, April 14-15, 1999. PTS, München, 1999. Pages 19,19E and 19-1 to 19-10.

The roman numerals are used in this summary when the publications are referred to.

AUTHOR'S CONTRIBUTION

The author's role in each of the publications has been the following:

I	main part of experiments, main part of analysis, first version of the manuscript,
II	main part of experiments, main part of analysis, first version of the manuscript,
III	all experiments, main part of analysis, first version of the manuscript,
IV	main part of experiments, main part of analysis, first version of the manuscript,
V	experiments in part, analysis in part, manuscript in part,
VI	experiments in part, analysis in part, manuscript in part,
VII	experiments in part, analysis in part, manuscript in part,
VIII	all experiments, all analysis, first version of the manuscript.

LIST OF SYMBOLS AND ABBREVIATIONS

The following symbols and abbreviations are used in this summary:

ß	Geometry factor
ρ	Density
ε	Strain
σ	Stress, breaking stress
σ_{app}	Apparent tensile strength
a	Crack length, defect size, empirical parameter
b	Empirical parameter, basis weight
A	Specimen size
E	Elastic modulus, specific elastic modulus, tensile stiffness
F	Force
G, G_C	Fracture release rate, critical energy release rate or fracture energy
K_C	Fracture toughness
R	Critical energy release rate, fracture energy
w_d	Damage width
w_p	Pull-out width
W	Specimen width
W_{nip}	Nip-peeling energy, z-directional fracture energy
CD	Cross direction
CMC	Carboxymethyl cellulose
CSF	Canadian standard freeness
EW	Escher-Wyss refiner
EWF	Essential work of fracture
FSP	Fibre saturation point
IPT	In-plane tear
L&W	Lorentzen&Wettre
MD	Machine direction
PGW	Pressurized groundwood
RBA	Relative bonded area
SEL	Specific edge load
TCF	Totally chlorine free (bleached chemical pulp)
TMP	Thermomechanical pulp
TMP R200	TMP fibre fraction retained on a 200 mesh screen
UB	Unbleached
UBSK	Unbleached softwood kraft
WRV	Water retention value

1. INTRODUCTION

Reinforcement pulp is used in order to improve the runnability of the paper web. In this study chemical softwood pulp is added to reinforce a mechanical pulp-based paper. This is expected to increase the critical strength properties of paper and thereby to improve the runnability of the web. Reinforcement pulp beating is done in order to further increase the critical strength properties of paper.

However, it is not clear what strength property, if any, correlates with web runnability. The problem has been discussed in several publications /Niskanen 1998a, Uesaka 2001a/. Actually, it seems likely that different paper properties are critical in different converting operations, e.g. the printing press vs. coating station /Niskanen 2001a/.

Several studies have indicated that web breaks originate from small flaws in paper /Sears 1965, Palsanen 1979, Page 1982, Roisum 1990, Swinehart 1996, Moilanen 1996/. However, analyses also show that the rheological behaviour of unflawed paper is important for pressroom breaks /Uesaka 2001a/. It is known that in addition to paper properties other factors like pressroom tension control have a major effect on runnability /Uesaka 1999, Parola 2000, Moilanen 2001/. Nevertheless, in this study it is assumed that the flaw resisting ability of the (dry) paper is a significant runnability factor. It is also considered that the rheological behaviour of unflawed paper can be important e.g. for pressroom runnability.

Traditionally out-of-plane tear tests like the Elmendorf method have been used to evaluate the flaw resisting ability of paper. However, already for some time, in-plane fracture properties have been considered to be more relevant /Page 1982, van den Akker 1967, Niskanen 1998a/.

The development of the in-plane properties of pure chemical pulp during beating is well known. In-plane fracture energy behaves differently during beating than out-of-plane tear /van den Akker 1967, Seth 1975, Johnson 1983, Uesaka 1984/. It seems possible that in-plane properties have a different optimum for reinforcement pulp beating (mechanical-chemical pulp mixture optimum vs. pure chemical optimum).

Strength properties of pulp mixtures are known to be non-linear in relation to component properties /Parsons 1969, Mohlin 1984, Retulainen 1992/. Chemical pulp seems to behave in mixtures as it would have been refined less than it actually is /Parsons 1969, Mohlin 1984/. Mohlin et. al. /1984/ have proposed that this is because of different drying shrinkage of chemical and mechanical pulp, which distorts the interfibre bonding between chemical and mechanical pulp fibres. On the other hand, it has been proposed /Retulainen 1992, Retulainen 1997/ that the non-linearity occurs because of a lack of activation of chemical pulp fibres in a mechanical pulp-dominated, low-density network.

1.1. Objectives and structure of the study

The main objective of this study was to find out how chemical softwood pulp refining affects the structure and strength of a mechanical pulp-based paper. The main objective was divided into the following sub-objectives:

1. Review current knowledge of reinforcement pulp beating in relation to mechanical pulp-dominated paper.

2. Determine the potential of reinforcement pulp beating in improving both the flaw resisting ability of paper and the rheological properties of unflawed paper. Apply novel methods in characterizing the flaw resisting ability and fracture process of reinforced paper.

3. Determine the effect of beating on critical strength properties of paper in comparison to certain other relevant control variables like wet straining and starch or filler addition.

4. Analyse the role of softwood beating and other control variables on the bonding, activation and structure (e.g. the size of the fracture process zone) of paper

This study was originally based on certain hypotheses:

1. The in-plane fracture properties of paper are more relevant than out-of-plane tear for the flaw resisting ability of paper. The behaviour of these properties against reinforcement pulp beating is different. If reinforcement pulp beating is optimised based on in-plane properties vs. out-of-plane tear, the needed amount of beating energy is different.

2. Mixtures of mechanical and chemical pulp do not show linear additivity behaviour. Therefore optimisation of reinforcement pulp beating should include testing of mixture sheets.

3. The specific bond strength cannot be reliably measured from paper. This is because interfibre bonds cannot be examined separately from the network in which they are located. Therefore novel methods to evaluate the bonding and microscopic fracture of paper have to be applied.

During the course of the study it became evident that the effect of beating on the properties of paper could not be explained simply by changes in interfibre bonding. At least one further mechanism was needed for the explanation. Therefore, a fourth hypothesis was formulated:

4. Beating affects the strength and structure of paper by two separate mechanisms 1. Increased fibre segment activation 2. Increased interfibre bonding.

This thesis study was part of a larger study, the KUMOUS project which had as its objective the characterisation and modification of reinforcement pulp fibres /Niskanen 2001a/.

The structure of the study is shown in Figure 1-1.

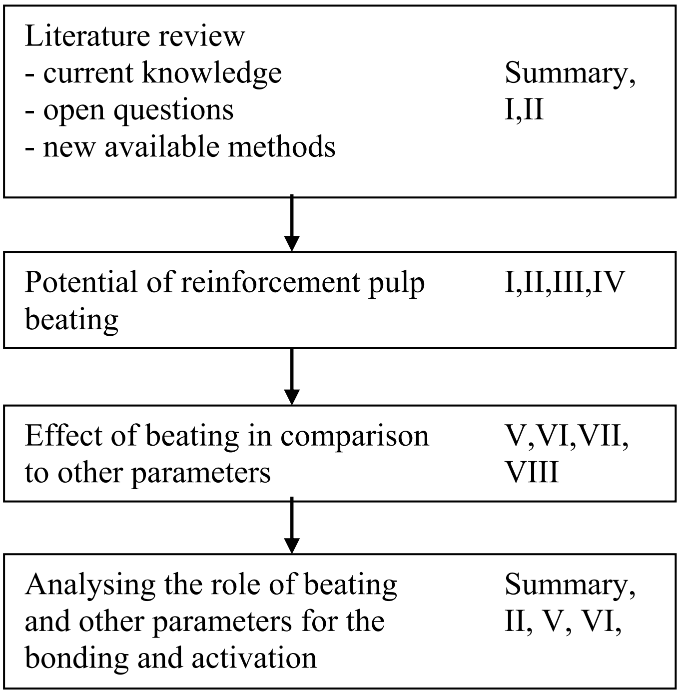

Figure 1-1. Structure of the study. The roman numerals refer to the publications, which deal with the objective in question.

1.2. Outline of the study

For mechanical pulp-dominated printing paper – like LWC base paper – runnability is a critical property. In this study runnability is assumed to be connected to the in-plane fracture properties and in-plane stress-strain behaviour. The study concentrates on the strength of *dry* paper.

Chapter 2 discusses the previous studies as a background for the work. Subjects discussed are interfibre bonding, activation, beating of reinforcement pulp, runnability of dry paper web, fracture mechanics and mixtures of mechanical pulp and chemical pulp.

Experimental methods used are described in Chapter 3. Further details are found in Publications I-VIII. Several mechanical tests were made according to standard procedures but a few non-standard methods were applied. Perhaps the most interesting new method used was the microscopic damage analysis (silicone impregnation) developed by Kettunen and Niskanen /Kettunen 2000a/. Another non-standard method applied was in-plane tear (to measure fracture energy) /Unger 1960, Kettunen 2000d/. In addition to standard methods z-directional strength properties were evaluated by the nip-peeling method /Tanaka 2000/. The FSP method introduced by Stone, Scallan et. al. /Stone 1968/ was applied to measure fibre swelling.

Relatively high proportions of chemical reinforcement pulp were used (30%-50%). This corresponds to the typical range used in the LWC base paper furnishes.

2. BACKGROUND

2.1. Interfibre bonding

Here the term "interfibre bonding" means the zone where two fibres are so close that chemical bonding and/or van der Waals interaction, occur or molecular entanglement may occur /Retulainen 1998/.

Specific bond strength and relative bonded area (RBA) are often used to describe the interfibre bonding in paper. However, the concept of specific bond strength and the measurement of both specific bond strength and RBA are problematic.

The concept of specific bond strength is based on the assumption that bond strength is directly proportional to the bond area /Niskanen 1998a/. However, Button /Button 1979/ has modelled the bond between two fibres as a lap joint and come to the conclusion that the stress distribution within a bond is nonuniform when loaded in shear. The highest stresses occur at the edges of the bond. Button /Button 1979/ also confirmed by experiments with cellophane strips that bond strength (nominal axial stress at failure) is almost independent of bond area.

According to Retulainen /Retulainen 1997, Retulainen 1998/, the specific bond strength cannot be measured reliably from paper. This is because interfibre bonds cannot be examined separately from the network in which they are located. According to Retulainen /Retulainen 1997/ there are two reasons for this:

1. The strength of a fibre bond is not only the strength of the interface but also the strength of the whole bonding system. This includes the internal strength of the fibre wall and axial elastic modulus of fibres.
2. The interfibre bond, fibre and network properties are interrelated. This is because during drying the properties of fibre bonds develop in continuous interaction with the fibre network.

According to Retulainen /Retulainen 1997, Retulainen 1993/ this means that the specific bond strength cannot be measured unambiguously, but only in relation to a certain structure and a certain loading mode (e.g. in-plane vs. out-of-plane).

In a recent article Page /Page 2002/ concludes that in certain conditions Nordman bond strength is not a relative index of the "real" specific bond strength. The reason is that the biggest part of the work of straining the sheet is not consumed by bond breakage but it is consumed by the fibres as they deform plastically.

The RBA characterizes the bonding degree of paper. By definition, RBA is the bonded surface area of fibres divided by their total surface area /Niskanen 1998b/. RBA has been measured directly with microscopical methods but there have been problems with the quality of the images of the paper cross sections. Cross sections have to be very thin to obtain a sharp image of the bonded interface /Uesaka 2001b/. In practice RBA is often measured indirectly as the light scattering coefficient of paper: the lower the light scattering, the higher the RBA. Another indirect method is gas absorption /Uesaka 2001b /. However, measurement of unbonded area is not straightforward by the indirect methods /Uesaka 2001b/. The use of the light scattering coefficient relies on the fact that a fibre surface element appears bonded if there is another fibre surface at a distance smaller than half the wavelength of light.

However, this does not guarantee that the two fibres are bonded mechanically, as the bonding distance is probably shorter /Niskanen 1998b/. Especially for mechanical pulp-dominated papers light scattering seems problematic as it has been shown that mechanical pulp fines increase both bonding and light scattering /Nieminen 1994, Retulainen 1997/. One approach is to assume that the apparent density of paper describes RBA. This method has a certain empirical justification though its theoretical validity is doubtful /Niskanen 1998b/.

It seems that bonded fibre segments (~bonding system), not the bonds themselves, are the relevant structural units that control the mechanical properties of paper. Because bond and fibre segment (and network) properties are interrelated through drying stresses, any change in bond properties will probably also effect fibre segment properties /Retulainen 1998/.

In this study the terms “interfibre bonding” or “bonding” are understood as the mean bonding strength of bonded fibre segments in paper. In a rough way, this could be understood as the combined effect of the specific bond strength and the RBA. The amount of bonding level is concluded based on indirect measurement, as no direct one was known.

2.2. Activation

Giertz /Giertz 1964, Giertz 1979/ has argued that the tensile strength increases during drying restraint because of increased fibre segment activation. When a bonded fibre shrinks laterally, compressive forces are transmitted to the neighbouring fibre via the bond area. Therefore, the neighbouring fibre is compressed axially, which, under restricted drying, will straighten, extend and strain the unbonded fibre segments. This causes the elastic modulus of the paper to increase, and from the strength point of view, the shrinkage activates the fibres /Giertz 1979/. Drying restraint increases drying stress, which then straightens “slag” fibre segments, that is, activates them.

According to Giertz and Rødland /Giertz 1979/ the local elongation during straining of paper is not constant; it can vary between 4 and 14 %. According to Retulainen /Retulainen 1997/, the local elongation can be up to 30 %, the average elongation of the test piece being 5-6 %. According to Giertz /Giertz 1979/, drying restraint decreases this variation in local elongation.

Van den Akker et. al. /van den Akker 1966/ have suggested that drying stresses not only affect activation but also increase the axial elastic modulus of fibres in the network. This idea is based on the results of Jentzen /Jentzen 1964, van den Akker 1966/ according to which drying under tension increased the elastic modulus of fibres.

According to Giertz /Giertz 1964/, increased fibre swelling by beating increases shrinkage during drying and therefore drying stresses increase and activation gets stronger and tensile strength increases.

2.3. Beating of reinforcement pulp

In this study the terms beating and refining of chemical pulp are considered to be synonyms.

Page /Page 1989/ has defined the beating effect as follows:
The changes that take place in the structure of pulp fibres, leading to changes in fibre properties and consequently pulp and sheet properties, as a result of subjecting the wet pulp fibres to a mechanical action.

Beating of reinforcement pulp aims to improve the runnability-related strength properties of paper. Typically, the aim is to increase the bonding ability of fibres so that most paper strength properties improve. However, certain paper properties, e.g. optical parameters, decrease during beating. Out-of-plane tear and in-plane tear strength typically first increase, reach a maximum and then decrease /Seth 1975, van den Akker 1967/. Refining also affects the papermaking process – with increased refining water removal on the paper machine becomes more difficult. The amount of refining is always a compromise between the desired and undesired effects.

2.3.1. Effect on fibres

Refining has several structural effects on fibres. Page /Page 1989/ has listed the following effects (simplified version here): fibre cutting, fines production, external fibrillation, internal fibrillation (swelling), fibre straightening (or curling), effect on kinks and microcompressions, dissolving of colloidal material, redistribution of hemicelluloses and formation of gelatinous surface.

Chemical pulp fibres swell as a result of beating. Fibre swelling is often called *internal fibrillation* /Page 1989/. Internal fibrillation can be detected e.g. by microscopy /Page 1967/. Probably the best quantitative measure of single fibre swelling is the fibre saturation point (FSP) method developed by Stone and Scallan /Stone 1968/. FSP increase by beating is caused by a random breakage of the crosslinks in the interfibrillar matrix, causing a loosening of the structure /Page 1989/.

It has been reported that beating increases the volume of so-called macropores in the fibre cell wall /Stone 1968, Maloney 2000/. Macropores are originally created during pulp cooking when lignin is removed from the cell wall /Stone 1967/. During pulp drying most macropores and some micropores are closed in such a way that they do not open during rewetting (~hornification) /Maloney 2000/. Beating of dried chemical pulp reopens macropores /Maloney 2000, Wang2002/.

Measurements have shown that beating increases fibre flexibility /Mohlin 1975, Kerekes 1985, Hattula 1988, Paavilainen 1993/. This is explained by the loosening of the cell wall structure during fibre swelling. This loosening causes a decrease in cell wall elasticity /Paavilainen 1993/. It seems logical that internal fibrillation increases fibre flexibility.

Mechanical pulping does not increase the amount of macropores in the fibre wall (amount of micropores increases) /Maloney 2000/. Internal fibrillation appears to be characteristic of chemical pulp fibres.

External fibrillation can be defined as the partial removal of the fibre wall, leaving it still attached to the fibre /Page 1989/. External fibrillation can be easily detected in microscopic studies. The amount of fibrillation has been quantified e.g. by measuring the increase in the specific surface of the long fibre fraction /Ingmanson 1959/. However, this does not answer the question how much external fibrillation affects paper strength.

Secondary *fines* are apparently generated by the abrasion of fibres against each other or against beater bars /Page 1989/. Fines consist mostly of fragments of P_1 and S_1 layers /Page 1989/.

Fibres are typically *straightened* during low-consistency beating /Page 1985, Mohlin 1991, Seth 2001/. This is believed to happen especially with dried pulps and because of fibre swelling and mechanical stress /Page 1985/. *Fibre shortening* occurs in a significant amount with high intensity beating and at high beating energy levels in low-intensity beating. In the beginning of (low-intensity) beating the measured fibre length (projection) can increase because of fibre straightening /II/. According to Page /1989/, fibres fail upon beating not by a cutting action but in tensile mode.

Beating increases *fibre collapse* /Paavilainen 1993/. However, it has been reported that most fibres collapse during papermaking, irrespective of the beating degree /Paavilainen 1993/ or already as unrefined /Jang 2002/. Therefore, fibre collapse does not seem to be a critical beating effect.

Low-intensity beating seems to affect *fibre strength* only a little /Paavilainen 1991, Thuvander 2001/. The slight increase in zero-span strength at low beating energy levels has been explained by fibre straightening /Mohlin 1991, Seth 2001/ and by better equalization of stress distribution within each fibre /Hardacker 1970/ and by additional intrafibre hydrogen bonding /Britt 1964/. However, fibre strength might increase because drying stress increases the elastic modulus of single fibre segments through the “Jentzen effect” /Jentzen 1964, van den Akker 1966, Retulainen 1998, Niskanen 2000/. With continued beating zero span starts to drop probably because of increased damage to the fibre cell wall and decrease in fibre length /VIII/.

2.3.2. Effect on paper strength

Internal fibrillation is often considered as the most important beating effect. Positive experience of the compression refining methods (creating mostly internal fibrillation) /Goncharov 1980, Hartman 1985/ support the view of the importance of fibre swelling (internal fibrillation). In addition, the similar effect achieved by refining-induced and charge induced-swelling /Scallan 1979, Lindström 1982, Laivins 2000/ supports the importance of internal fibrillation. Also, results showing the benefits of low-intensity beating support the same view. In several studies low-intensity beating of softwood has given the highest results for tensile strength /Seth 1996, Zhang 1997, Croney 1999/. On the other hand, opposite results have been reported for tensile strength (softwood) /Levlin 1980/. However, an obvious downside of high-intensity beating is fibre shortening. For property pairs like out-plane-tear vs. tensile /Levlin 1980/ and in-plane fracture properties vs. tensile /Zhang 1997, I/, low beating intensity seems to give the best results.

It is known that swelling makes fibres more flexible and it is assumed that because of that the sheet densifies during drying under the forces of surface tension. The fibre-fibre bonded area is increased and this accounts for the increase in e.g. tensile strength /Page 1985/. According to Page the theory was first proposed by Campbell /Campbell 1933/.

However, also other views have been expressed, e.g. Page /Page 1985/ has concluded that (for dried pulp) fibre straightening is the most important refining effect. It is known that if sheet density is increased by wet pressing instead of beating, the tensile strength and elastic modulus differ at constant density (beating gives higher strength) /Giertz 1964, Page 1979, Page 1985, Niskanen 1998a/. The conclusion /Page 1985/ is based on the idea that when fibre segments in the sheet are straighter and more ready to support a load and, the fibres as a whole are straighter, the stress distribution in the sheet is better. Niskanen /Niskanen 1998b/ has suggested that beating affects elastic modulus mostly by fibre segment activation, only a minor effect would be due to bonding. Niskanen bases his theory to a certain extent on earlier publications /Jentzen 1964, Giertz 1964, Giertz 1979, Page 1979/. Beating-induced fibre swelling could both straighten fibres /Page 1985/ and increase activation through drying stresses /Giertz 1979, Niskanen 2000/. The activation concept is discussed in more detail in chapter 5.

Page /Page 1985/ has concluded that external fibrillation has only a limited effect on the tensile strength or elastic modulus. This conclusion was based on the notion that beating gives a clearly higher elastic modulus at the same density compared to wet pressing. According to Page this could not be due to external fibrillation, because increased fibre-fibre bond strength does not have an effect in the elastic regime (where bonds are not broken). The above is based on the assumption that external fibrillation affects only bond strength and not the RBA. Hartman /Hartman 1985/ studied the effect of external fibrillation with a special laboratory device designed to produce external fibrillation on fibres. His results indicated that external fibrillation does not much effect the tensile strength.

Hartman /Hartman 1985/ also studied the effect of secondary fines during beating on the strength properties. He concluded that internal fibrillation causes 75 % of the increase in tensile strenght during beating and secondary fines 25 %, where as external fibrillation has practically no effect. Page /Page 1985/ has concluded that less than 20 % of the beating effect on tensile strength is caused by fines generation. The amount of secondary fines depends e.g. of the beating energy. In a study at HUT a moderate beating energy level of 100 kWh/t (conical laboratory refiner) resulted in about 3 % secondary fines for softwood kraft /Zhang 1997/. At the same time tensile strength increased by 96 %.

Howard, Poole and Page /Howard 1994/ did a factor analysis on a wide range of laboratory beating data (various equipment). They mathematically derived that there were only three factors that accounted for 94 % of the total variance. It was concluded through logical reasoning that the three factors are bonding, fibre length + fines and microcompressions /Howard 1994/. It seems that only a limited number of factors cause the major effect on paper properties /Waterhouse 1997/. A large number of refining effects on fibres are true but the importance of many of these effects for paper strength seems small.

It should be remembered that in many industrial cases the optimal refining for paper strength has to be a compromise because of e.g. drainage properties or because of production capacity.

2.4. Runnability

The runnability of the dry paper web is a critical factor in paper manufacturing, in finishing and converting operations and in printing. Several studies have indicated that web breaks are often initiated by small flaws in paper (shives, holes etc.) /Sears 1965, Palsanen 1979, Page 1982, Roisum 1990, Swinehart 1996, Moilanen 1996, Linna 2001, Koskinen 2001/. This being the case, fracture mechanics should give useful information about the flaw-resisting ability and therefore runnability of the paper web.

Because of certain practical problems (e.g. web breaks are very infrequent) extensive statistical data have been presented only in few cases in the literature to show what, if any, strength property actually correlates with paper runnability. However, there are a few studies which a show certain correlation between in-plane fracture parameters and press room runnability like the classical paper by Page and Seth /Page 1982/. Swinehart and Broek /Swinehart 1996/ reported a correlation between fracture toughness and coater runnability and Moilanen and Lindqvist /Moilanen 1996/ reported a correlation between fracture energy and printing press runnability. In other studies tensile strength correlated with coater runnability /Palsanen 1979, Swinehart 1996/ and press room runnability /Moilanen 1996, Uesaka 2001a/. In all cases the correlation to any average strength property has been weak. Especially when analysing the correlation between out-of-plane tear (e.g. Elmendorf) and breaks, the results are mostly poor /Palsanen 1979, Page 1982, Uesaka 2001a/. In addition to the experimental results, out-of-plane tear is considered irrelevant from the logical point of view that the paper web is almost always subjected to in-plane loads /Seth 1975, Page 1982, Niskanen 1998a/.

Uesaka et. al. /Uesaka 1999, Uesaka 2001a/ have criticised some of the pilot studies of the flaw-effect, because in them web tension is increased until a break occurs, and because there is no quantitative data from pressrooms indicating such a strong effect of shives or flaws. On the contrary, some studies show that only a small fraction of breaks start from holes /Frye 1994, Moilanen 1996/. Pressroom studies show that the tension values at printing presses are at much smaller levels; with the exception of short tension peaks during roll changes etc. Therefore, pilot experiments done at high web tension levels may not simulate the relevant phenomenon at the printing press /Uesaka 1999/. It is also known that because of formation variations small enough flaws or holes do not affect the average strength of paper (critical defect size is about 1-5 mm) /Donner 1997, Rosti 2001, Hansen 2001, Wathen 2001/. A parametric study of press room runnability suggests that even with a centre crack of 40 mm paper rolls can run with modest break frequency, if web tension is well controlled /Uesaka 1999/.

In order to characterize the strength of the paper web in press rooms, Uesaka et.al. /Uesaka 1999, Uesaka 2001a/ considered the tensile index and elastic stretch (tensile strength divided by elastic modulus). These two rheological factors are crucial for web breaks, if web breaks are not initiated by flaws in the paper, but instead by "random" tension peaks that in most cases occur on unflawed paper. Then the stress-strain behaviour of paper controls the maximum tension peak that the web can endure without a break. In this study instead of the term *elastic stretch* the term *elastic breaking strain* is used /Niskanen1998a, VI/. Elastic breaking strain is the elastic strain *recoverable* at failure – not the "stretch" at failure /Uesaka 1999/.

It seems plausible that breaks e.g. on a blade coating unit can depend on other factors than press room breaks /Niskanen 2001a/. Coating unit breaks probably depend, among other factors, on the flaw-resisting ability /Koskinen 2001/ and tensile strength /Palsanen 1979/ of (dry) paper. Even though paper is wetted at coating, part of the paper may remain dry, with the dry part carrying the load.

On the other hand, it seems that other reasons like paper strength non-uniformity /Uesaka 1999, Uesaka 2001a/, variations in the press room tension /Larsson 1984, Uesaka 1999, Uesaka 2001a/ and variations in the tension profile of paper (in CD) /Parola 2000, Linna 2001, Moilanen 2001/ can cause breaks. Air humidity variations between winter and summer affect breaks so that there are more breaks during low humidity /Page 1982/. It seems that different reasons cause breaks at pressroom: press room conditions, reel condition, variation of paper properties /Roisum 1990/ and the average level of paper strength. Therefore it seems understandable that a selected average paper strength property can, at best, have a limited correlation with pressroom breaks.

A laboratory study is, however, limited to the average level of paper strength. *In this study,* it is assumed that the flaw-resisting ability of dry paper correlates with runnability in certain converting operations like coating. Furthermore, it is assumed that the flaw-resisting ability can be described by in-plane fracture properties of paper, especially by fracture toughness. On the other hand, it is considered that tensile strength and elastic breaking strain correlate e.g. with printing press runnability /Uesaka 1999, Uesaka 2001a/.

2.5. In-plane properties

2.5.1. In-plane fracture properties

The in-plane fracture properties (fracture energy, fracture toughness and apparent tensile strength) are discussed in the following sections.

According to the classical paper by Griffith /Griffith 1920/, a crack (or flaw) in a material can propagate only if the stored elastic strain energy for crack growth is sufficient to overcome the fracture resistance of the material. For linearly elastic material the condition for crack propagation is /Niskanen 1998a/:

$$G = \beta\sigma^2 a/E \geq G_C = R \quad (1)$$

where G is the energy release rate, σ the applied stress, E the elastic modulus, a the crack length and β a geometry factor.

At the onset of crack propagation, $G = G_C = R$. This is the critical energy release rate, or the *fracture energy* of the material.

According to the linear elastic fracture mechanics, *fracture toughness*, K_C, (material property) can be calculated from fracture energy (G_C) and elastic modulus (E) /Niskanen 1998a/:

$$K_C = \sqrt{G_c E} \quad (2)$$

Fracture toughness is directly proportional to the critical web tension or the apparent breaking stress of a flawed paper web /Niskanen 1998a/. Fracture energy may also have a direct bearing on sudden disturbances in web tension, e.g. during printing.

The analysis of fracture energy is often considered appropriate even though paper is not a linearly elastic material. For some paper grades, plastic deformation close to the crack tip is large. This makes it difficult to experimentally test the fracture energy of paper and therefore a widely recognized fracture energy test has not been established. Basically all fracture energy tests should give the same results, but in reality the results vary depending of the method /Kärenlampi 1998, Yu 2001/. Different details of each test affect the results. The methods measure somewhat different quantities /Kärenlampi 1998, Yu 2001/ and many of the methods suffer from geometry-dependency /Yu 2001/. Even though no ideal method has been found, many fracture energy measurements like J-integral vs. in-plane tear /Kettunen 2000c, Yu 2001/ and short span test vs. in-plane tear /Yu 2001/ show linear relations with samples of large variation. Also EWF and STFI's J-integral reflect fibre length and the proportion of chemical pulp in the furnish similarly, but the two methods differ to a certain extent in the effect of beating /Kärenlampi 1998/.

The *apparent tensile strength* (measured from a notched specimen) is directly proportional to fracture toughness /Niskanen 1998a/. In this study the fracture toughness was sometimes approximated by apparent tensile strength measurements. The value of apparent tensile strength depends on the sample dimensions and the size of the pre-cut notch (flaw) in the test piece (it is not a material property like fracture energy and fracture toughness are).

It should be noted that there are little statistical data correlating fracture toughness to runnability and also the terms used are inconsistent (e.g. fracture toughness, fracture energy, fracture resistance, tenacity) /Seth 1975, Swinehart 1996, I, IV/. For example in some of our group's earlier publications, fracture energy (e.g. in-plane tear and J-integral) was misleadingly called fracture toughness /I, Hiltunen 1999/. SCAN standard for Fracture toughness (SCAN-P 77:95) is actually a standard for fracture energy measured by the J-integral method.

2.5.2. Breaking stress of paper web

According to Uesaka /Uesaka 1999, Uesaka 2001a/, the ordinary tensile test gives useful information on web breaks if clear defects are absent. However, there is always formation like variability present in a paper specimen and this gives variation to the measured breaking stress value. Gregersen /Gregersen 1998/ has tested the size dependency of breaking stress (σ) from tensile test. It seems to follow this theoretically derived approximation:

$$\log(\sigma) = a - b \log(A) \qquad (3)$$

where A is specimen size and *a*, *b* are empirical parameters. From equation 3 it can be seen that the breaking stress of the web is essentially smaller than the ordinary tensile strength. This is because σ depends on the weakest point in the sample – not on the average strength of the sample like fracture energy or elastic modulus /Niskanen 1998a/. This makes the measurement of the breaking stress of the web (~tensile strength of large sample) difficult.

2.6. Mixture of mechanical and chemical pulp

2.6.1. Additivity behaviour

In many cases, paper properties have been found to be non-linear in relation to pulp mixture proportions /Parsons 1969, Mohlin 1984, Retulainen 1997/. According to Retulainen /Retulainen 1992/, only in a few special cases does linear development occur.

The reason for the non-linear additivity behaviour has been explained in different ways. Mohlin et. al. /Mohlin 1984/ have proposed that mechanical and chemical pulp form two separate networks. This was based on certain results showing non-linear additivity behaviour while mixing chemical and mechanical pulps. In several cases the chemical pulp bonding potential did not seem to be fully utilized in mixture sheets. According to Parsons /Parsons 1969/ and Mohlin et al. /Mohlin 1984/, chemical pulp behaves in mixtures as if it would have been refined less than it actually is. The increase in tensile strength was smaller and in out-of-plane tear higher than expected from the linear additivity relationship. Mohlin /Mohlin 1984/ explained the low bonding of mechanical and chemical pulp fibres by the different drying shrinkage behaviour of chemical and mechanical pulp fibres. Kazi and Kortshot /Kazi 1996/ found a drop in fracture energy at small reinforcement pulp additions and they explained this by poor bonding of kraft and TMP.

On the other hand, Retulainen's /Retulainen 1992/ results indicate that chemical and mechanical pulp bond quite well. The plybonding of mechanical-chemical sheets was stronger than that of mechanical-mechanical sheets (though lower than chemical-chemical sheet bonding). Retulainen's /Retulainen 1992/ results show that the drying method (restraint vs. free) did not significantly affect the additivity behaviour of mixtures. Also in a tensile fracture of a mechanical pulp-based paper, a fairly large percentage of chemical pulp fibres are broken /Retulainen 1992/. In a recent publication, Koskinen, Kosonen and Ebeling /Koskinen 2001/ conclude that chemical and mechanical pulp bond well.

Retulainen /Retulainen 1992/ explains the non-linear additivity behaviour by lack of activation of chemical pulp fibres in a mechanical pulp-dominated, low-density network. According to Retulainen /Retulainen 1992/, in mechanical-chemical pulp blends mechanical pulp has a dominant role in determining the structure of the sheet (in his experiments the proportion of mechanical pulp was 50% or over). This seems to be because the stiff mechanical fibres form a low network density, where flexible chemical pulp fibres might form slack segments /Retulainen 1992/. Experimental results show that in mechanical-chemical pulp mixtures stiff mechanical pulp fibres debond first /Kettunen 1999a/. It has also been suggested that in the pulp mixture the amount of fines drops below a "limiting state" as chemical pulp contains little fines /Retulainen 1997/.

Kazi and Kortshot /Kazi 1996/ found that, with a small proportion of reinforcement pulp, beating had no effect on the fracture energy of mixed pulp. According to the linear rule of mixtures, the mixture sheet properties would be linearly proportional to the component properties. Kärenlampi et al. /Kärenlampi 1997a/ have reported that the fracture energy of a mixture of beaten softwood kraft (high proportion) and pressure groundwood pulp is linearly proportional to the relative pulp amounts. On the other hand, Shallhorn /Shallhorn 1994/ found the fracture resistance of a mixture of unbeaten softwood kraft and groundwood pulp to be non-linear: the fracture resistance was higher than predicted from the linear rule of mixtures. It seems that reinforcement pulp beating and the proportions of components affect the additivity behaviour of the mechanical-chemical pulp mixture.

2.6.2. Optimal morphology for reinforcement pulp

According to Levlin /Levlin 1991/ reinforcement pulp fibres should be long, slender and flexible. These ideas were based on, among other things, empirical experiments where out-of-plane tear was assumed to describe runnability. Slender fibres (low coarseness) give a higher number of fibres per unit weight of pulp. Lower coarseness is thought to give a greater contact area between fibres and therefore a chemical pulp network is formed with a low proportion of chemical pulp. The concept of the chemical pulp network is similar to the suggestion by Mohlin et. al. /Mohlin 1984/ that mechanical and chemical pulp form two separate networks. If we accept the idea of two separate networks, it means that the properties of both components could be optimised separately. Based on this idea Ritala /Ritala 1987/ calculated the percolation thresholds for the amount of chemical pulp (minimum amount for chemical pulp network to be formed). However, the concept of a separate chemical pulp network is in contradiction with Retulainen's /Retulainen 1992/ report of good bonding between chemical and mechanical fibres.

Paavilainen /Paavilainen 1991, Paavilainen 1994/ and Seth /Seth 1996/ have tested the effect of fibre coarseness on pure chemical pulp sheets. The results support the ideas of the benefits of low fibre coarseness. Low coarseness gives higher tensile strength /Seth 1996, Paavilainen 1991/, higher elastic modulus /Seth 1996/ and higher Scott-bond /Paavilainen 1994/. According to Seth /Seth 1996/ also fracture energy increases with low coarseness. The benefit of low coarseness is explained by a greater contact area between fibres /Paavilainen 1994, Seth 1996/.

On the other hand, in one experiment with softwood kraft and PGW, kraft coarseness was not critical for the in-plane fracture energy of the pulp mixture /Kärenlampi 1997a/. It has also been reported that low coarseness decreases the out-of-plane tear of pure chemical pulp /Seth 1988, Seth 1996/.

Fibre strength is important for out-of-plane tear /Seth 1988, Seth 1996/. Also in-plane fracture energy decreases when fibre strength decreases /Seth 1996, Kärenlampi 1997b, Kärenlampi 1998, Kettunen 2000b, Yu 2001/.

Fibre length has a large effect on in-plane fracture energy /Seth 1996, Kärenlampi 1997b, Yu 1999, Kettunen 2000c/. It has been shown that even low-bonding viscose fibres with sufficient length improve the fracture energy of softwood kraft-based paper /Yu 1999/.

3. EXPERIMENTAL

3.1. Sample material

The details of the experimental sample sets used in this summary are reported in Publications I – VIII.

* Sets of softwood reinforcement pulp with different beating levels /I,II,III,IV,V,VI,VII,VIII/
* A set of softwood reinforcement pulp with different cooking methods (e.g. kraft vs. neutral sulphite) and beating levels /IV/
* A set of softwood kraft pulp with different fibre charge levels and beating levels /VIII/
* A set of softwood kraft pulp at different wet straining levels and different beating levels /V/
* A set of softwood kraft pulp with filler and starch addition and at different beating levels /VI/
* Mixtures of TMP with a softwood reinforcement pulp of different beating levels /I,II,III,IV,VI/
* Mixtures of TMP with a softwood reinforcement pulp of different cooking methods /IV/
* Mixtures of TMP with softwood kraft pulp at different beating levels with filler /III,VI/, starch /VI/ and TMP without fines /III/
* Mixtures of TMP with coarse and fine abaca pulps /VII/

Both industrial and laboratory pulps were used. TMP was in all cases industrial never-dried (freezer stored) and made from Norway spruce. Industrial reinforcement pulp was unbleached never-dried /II,VIII/ or bleached dried pulp /I,II,III,IV,V,VI,VII/. Laboratory pulps were never-dried bleached pulps /IV/. Reinforcement pulp was either spruce /III,IV,V,VI,VII/ or a mixture of Scots pine and spruce /I,II,VIII/.

3.2. Refining of reinforcement pulp

Softwood reinforcement pulp refining was done in most cases with an Escher-Wyss conical laboratory refiner /I,II,III,IV,V,VIII/. In some cases, a Valley hollander beater /I,VI,VII,VIII/ was used.

3.3. Mechanical testing

60 g/m^2 handheets were made according to SCAN 26:76 (with restraint drying). The only exception in drying is the set of different wet straining levels in paper V. TMP containing sheets were made with a white water circulation.

Fracture energy has been evaluated mainly by using the in-plane tear (IPT) test. Because the results of fracture energy tests vary /Yu 2001/, two different methods should be used when possible. In this study in-plane tear was selected as the primary method because it was suitable for the damage analysis method. As a reference for IPT, J-integral fracture energy was used (SCAN-P77:95). In the beginning of this study /I/, one additional method was used to measure fracture energy. However, the use of this method was not continued because of a fear that the results include also elastic energy.

The elastic modulus (E) is inversely proportional to paper thickness, and has the units GPa. For thin materials like paper, however, accurate measurement of thickness is sometimes problematic. The specific modulus of elasticity or modulus divided by density (ρ), $E/\rho = F/Wb\varepsilon$, is often more appropriate because no thickness measurement is necessary /Niskanen 1998a/. F/W is the measured force over specimen width for a given strain ε, b being basis weight. In this study the specific elastic modulus (i.e. tensile stiffness index) is used unless otherwise stated (units kNm/g).

Apparent tensile strength is measured from a test strip with a prepared transverse cut. The specimen geometry has a strong effect on the measurement results. Because results depend on e.g. the size of the cut, this ranking predicts break frequency only if the papers compared have the same distribution of defect size and shapes. In this study, the apparent tensile strength was measured with a L&W Fracture toughness tester. The test strip widths were 15 mm for the uncut sample and 50 mm with a 20 mm pre-cut notch in the middle for the cut sample /SCAN-P77:95/.

For out-of-plane fracture characterization was done**,** the Scott bond test /V,VI,VII/, nip-peeling test /VI, Tanaka 2000/ and z-directional tensile test were performed /VI/.

3.4. Damage analysis

The size of the fracture process zone was measured by “damage analysis” from silicone-impregnated samples /Kettunen 2000a, Kettunen 2000c, Niskanen 2001b/. The damage analysis gives “damage width”, which is a measure of the size of damage zone (fracture process zone). The fracture process zone is the area where plastic deformation during paper fracture occurs /Seth 1993/. Plastic deformation in the damage zone consists of fibre and bond breakages and other microscopic ruptures during the fracture of paper. Silicone enhances the contrast of newly revealed fibre surfaces enough to allow the interfibre bond openings and fibre breakage to be detected /Korteoja 1996/. Examples of images of siliconized samples can be seen at /V/. Silicone impregnation has only a minor effect on the interfibre debonding process /Kettunen 2000c/. After in-plane tearing, the samples were scanned with an ordinary desktop scanner (Mustek). The images were analysed using an appropriate program /Kettunen 2000a/. Two parameters are measured: Damage width (w_d) characterizes the extent of fibre debonding from the crack line. Pull-out width (w_p) characterizes the distance of the sticking fibre ends from the crack /Kettunen 2000a/. Independent thermograchic measurement of the damage zone agrees with damage analysis results /Kettunen 2000c/.

4. RESULTS

This chapter presents the experimental results of the study. Both pure chemical pulp and corresponding mechanical-chemical pulp mixture results are analysed.
First, the connection of beating and internal fibrillation is discussed briefly.
Second, the effect of reinforcement pulp beating on in-plane strengths and z-strength is discussed.
Third, the effect of beating on the size of the fracture process zone is analysed. Based on this, conclusions regarding interfibre bonding are drawn.
Fourth, the effect of beating and fibre morphology on additivity of mixture properties is analysed.

4.1. On the beating and internal fibrillation

The results in Figure 4-1 compare the property pare in-plane fracture energy vs. tensile strength at two beating intensities. Generally, the lower the beating intensity, the higher seems to be the achieved strength combination for pure chemical pulp (e.g. fracture energy vs. tensile /Levlin 1980, Zhang 1997, I, II/. Presumably the reason is that high-intensity refining causes more fibre damage than low-intensity refining. In deed, high intensity refining gave lower fibre length and strength /I,II/ and also a bigger amount of fibre wall dislocation zones /VIII/ at given refining energy.

Also in a mixture of chemical and mechanical pulp low refining intensity seems to give a little higher fracture energy at given tensile strength (Figure 4-1) /II/. However, refining intensity does not seem to affect the trend of the mixture fracture energy against increased kraft refining /I,II/.

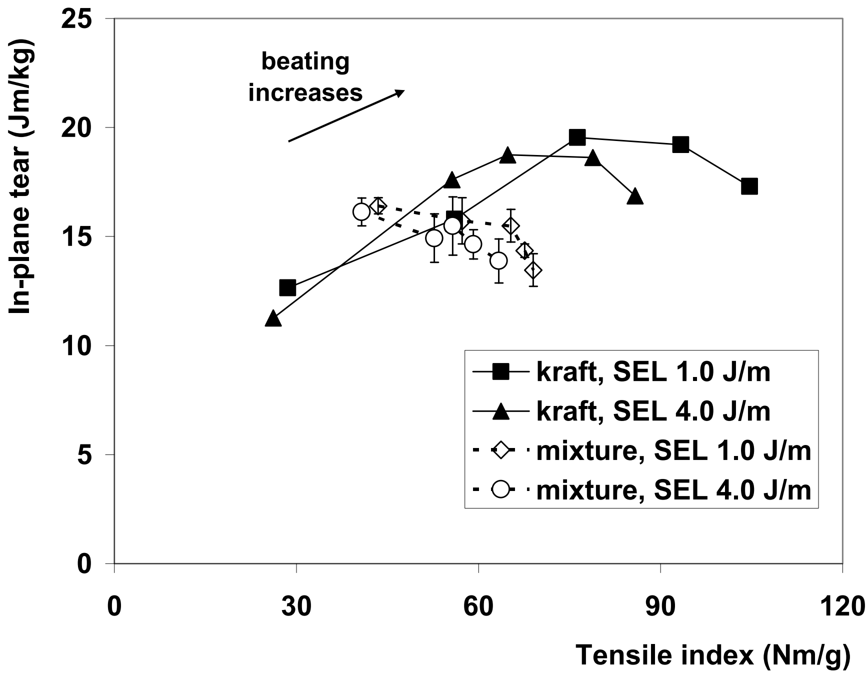

Figure 4-1. In-plane tear against tensile index – two refining intensities and two pulp furnishes. Pulps are from publication II (unbleached softwood kraft) - the mixture has 50 % TMP. Tensile index is changed by kraft refining. 95 % confidence levels are shown for mixture series.

In Figure 4-2 charge-induced (total charge of fibres) fibre swelling and beating are compared. It is known that a Valley beater has a low intensity compared to industrial refiners /Eskelinen 1969/. In Valley beating the result curves of the two charge levels concur, even though high-charged pulp has a higher tensile strength at given beating time /VIII/. Increased fibre charge and low-intensity beating seem to affect tensile strength and density mainly through the same mechanism. Laivins and Scallan /Laivins 2000/ have recently reported similar results. It seems that charge-induced swelling and low-intensity beating have similar effect, which supports the view that internal fibrillation (fibre swelling) is the most important beating effect.

The result curves do not fully concur at high-intensity beating, which is possibly due to other beating effects like external fibrillation and/or the "buffer effect". It has been suggested in the literature that higher swelling protects the fibre cell wall from refining impacts thereby reducing effects like fibre cutting and other fibre damages /Wultsch 1964, Hietanen 1990/. Therefore high-intensity refining gives higher tensile strength at given density for high-charged pulp /VIII/. In support of this theory it was found out that the amount of dislocation zones in the fibre cell wall increases with beating and that high-charged pulp has a smaller amount of dislocation zones at given beating energy, Figure 4-3 /VIII/.

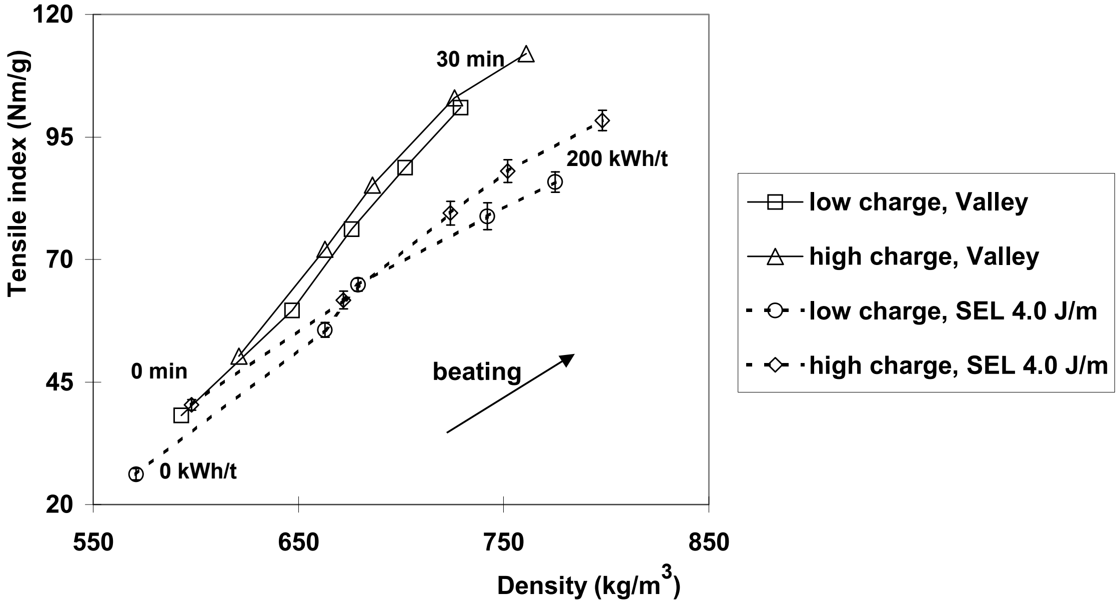

Figure 4-2. Tensile strength against density - two charge levels and two beating methods /VIII/. In Escher-Wyss beating the SEL was 4.0 J/m. First point of each curve is unbeaten sample.

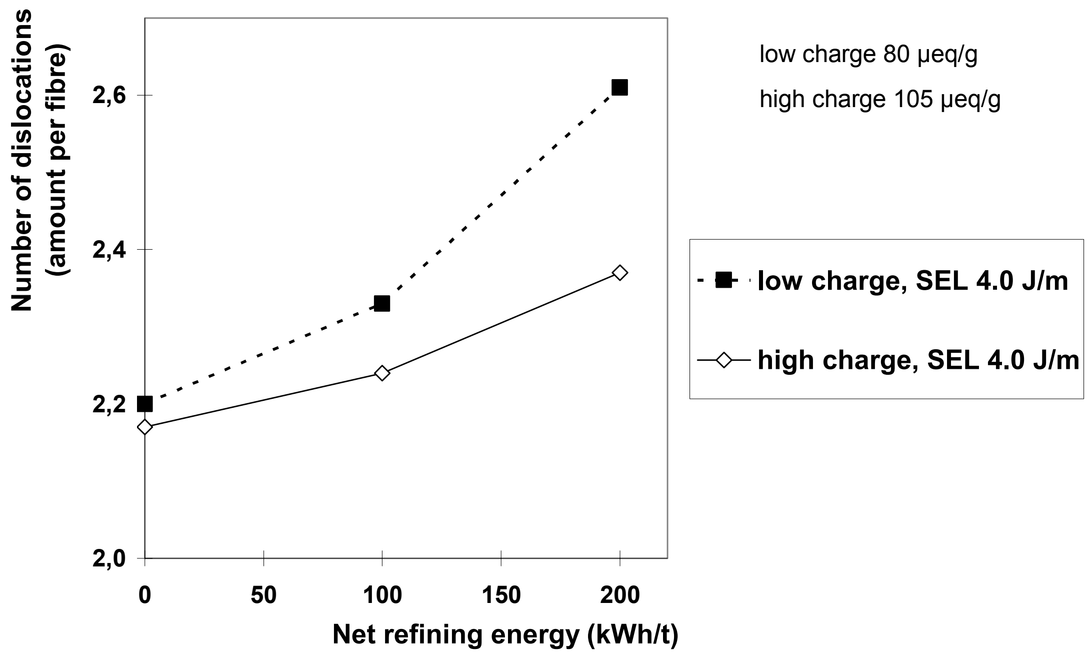

Figure 4-3. Number of dislocations of early wood fibres during high-intensity Escher-Wyss laboratory refining - two charge levels /VIII/.

4.2. Reinforcement pulp beating vs. strength

According to the linear elastic fracture mechanics, in-plane fracture energy and elastic modulus define fracture toughness and therefore the flaw resisting ability of paper /Niskanen 1998a/. On the other hand, Uesaka et al. /Uesaka 1999, Uesaka 2001a/ have concluded that tensile strength and elastic breaking strain are important factors for pressroom runnability (they describe the rheological behaviour of unflawed paper).

4.2.1. In-plane rheology of unflawed paper

Figure 4-4 shows that the specific elastic modulus of both pure softwood kraft sheets and that of mixture sheets increases during kraft refining. In mixture series, however, the increase in specific elastic modulus slows down at high levels of kraft refining. Tensile strength shows a similar trend as elastic modulus during refining.

The elastic breaking strain of pure softwood kraft increases during refining, as shown in Figure 4-5. Corresponding elastic breaking strain of mixture sheets first slightly increases but soon levels off. The elastic breaking strain seems to depend less on reinforcement pulp refining than the elastic modulus or tensile strength does. Because of definition (tensile/elastic modulus) elastic breaking strain is considered to be more dependent on bonding than the elastic modulus /VI/.

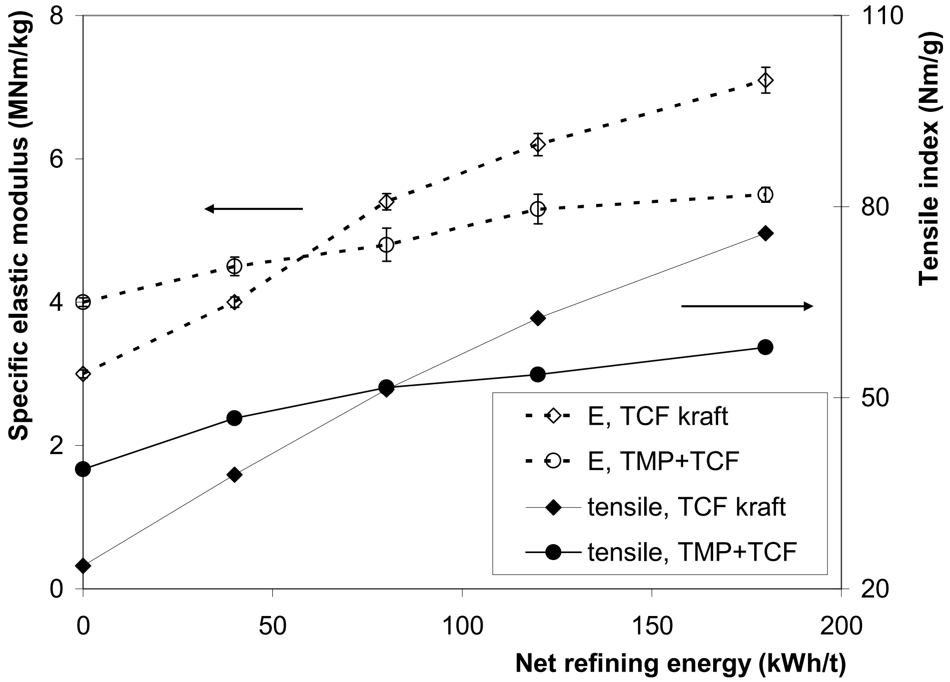

Figure 4-4. Effect of kraft refining on tensile index and specific elastic modulus (E) of pure TCF bleached kraft sheets and mixture sheets (kraft + TMP) /II/.

4.2.2. In-plane fracture properties

It is known that the in-plane fracture properties behave differently during refining than the out-of-plane tear (Figure 4-6). Earlier studies have shown that the in-plane fracture energy of pure chemical pulp sheets has a similar maximum against refining as the conventional out-of-plane tear, however, the maximum occurs at a higher level of refining energy /van den Akker 1967, Seth 1975, Johnson 1983, Uesaka 1984/.

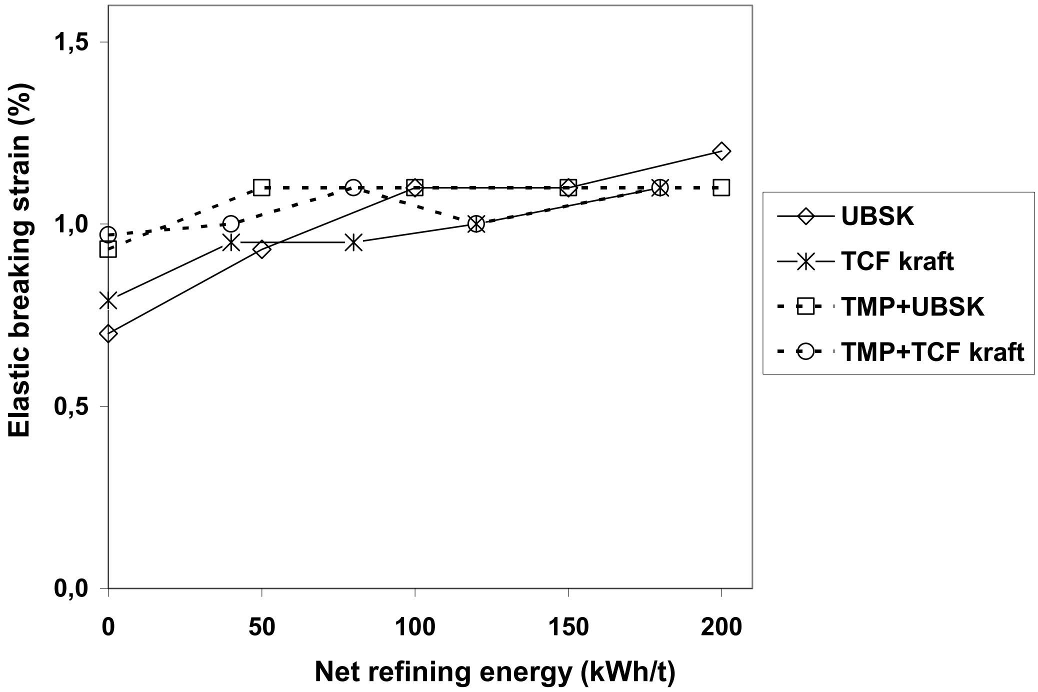

Figure 4-5. Effect of kraft refining on elastic breaking strain of pure kraft and mixture sheets, elastic breaking strain calculated by dividing the tensile index with specific elastic modulus. Data from /II/. Mixture ratios: unbleached softwood kraft (UBSK) 50 % and TCF-kraft 40 % /II/. TCF was dried and UBSK was never dried.

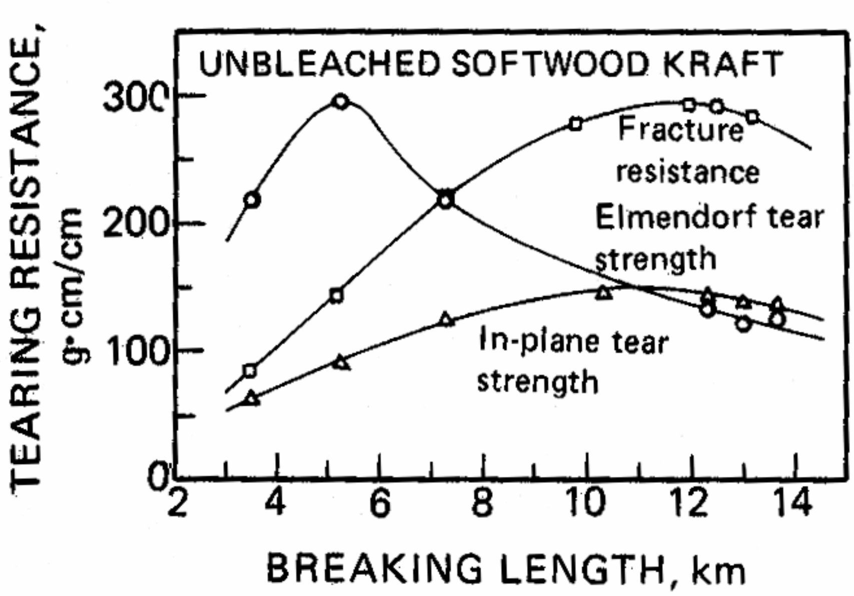

Figure 4-6. In-plane tear, out-of-plane tear and fracture resistance against beating /Seth 1975/.

The refining effect on fracture energy is caused by an increase in bonding (and possibly activation) and an increase in the number of fibre failures that take place during the fracture process /Niskanen 2001a/. The first effect increases fracture energy and the latter effect decreases it. At a certain level of refining, the increasing amount of fibre failures begins to dominate, thus decreasing fracture energy. This is a similar interpretation as that offered for the well-known behaviour of ordinary out-of-plane tear energy in refining /Parsons 1969/. The difference in the testing mode (in-plane vs. out-of-plane) causes the shift in the location of the maximum /Seth 1975/. It has also been reported that the out-of-plane tear of pulp mixtures is higher than expected from the pure component values /Parsons 1969, Mohlin 1984/. It seems that chemical pulp behaves in mixtures as if it would have been refined less than it actually is /Parsons 1969, Mohlin 1984/

The original *working hypothesis* of this study was based on the above observations. It was hypothesised that refining the chemical pulp component would increase the fracture energy of a mixture of mechanical and chemical pulps. It was assumed that in-plane fracture energy would show a maximum at higher beating energy level than out-plane tear also for mixture sheets.

The in-plane tear (IPT) of pure kraft series reached a maximum at a rather high refining degree compared to out-of-plane tear (Figure 4-7) /I,II,III,IV/. As expected the in-plane tear work of the pure chemical pulp series developed differently from that of the mixture series during chemical pulp refining, as shown in Figures 4-1 and 4-8. However, the mixture series did not show a expected maximum - the in-plane tear work actually decreased against kraft refining /I,II,III,IV/.

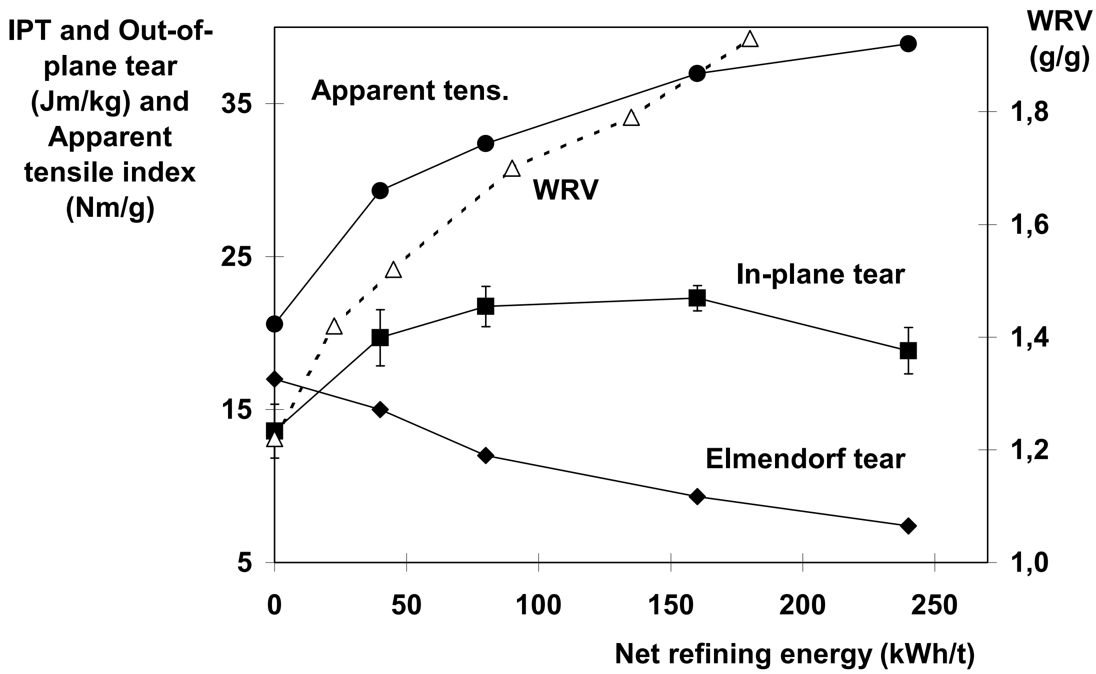

Figure 4-7. In-plane tear index (IPT), out-of-plane tear index (Elmendorf), apparent tensile index and WRV of kraft vs. refining energy (SEL 1.0 J/m) /III/. Typical 95 % confidence levels shown for one curve.

The behaviour of IPT against refining did not change with refining intensity /II/ nor with different cooking methods for reinforcement pulp /IV/ nor with different drying conditions /Zhang 2002/ nor with different reinforcement pulp proportions (60 % or less chemical pulp) /III, Zhang 2002/. On the other hand, results did not depend on the measurement method for fracture energy as the trend lines by in-plane tear work differ only a little from the L&W J-integral results (J-integral method gave lower absolute energy values than the in-plane tear) /II/. The J-integral-IPT comparison is in agreement with other reports /Yu 1999, Kettunen 2000c, Yu 2001/.

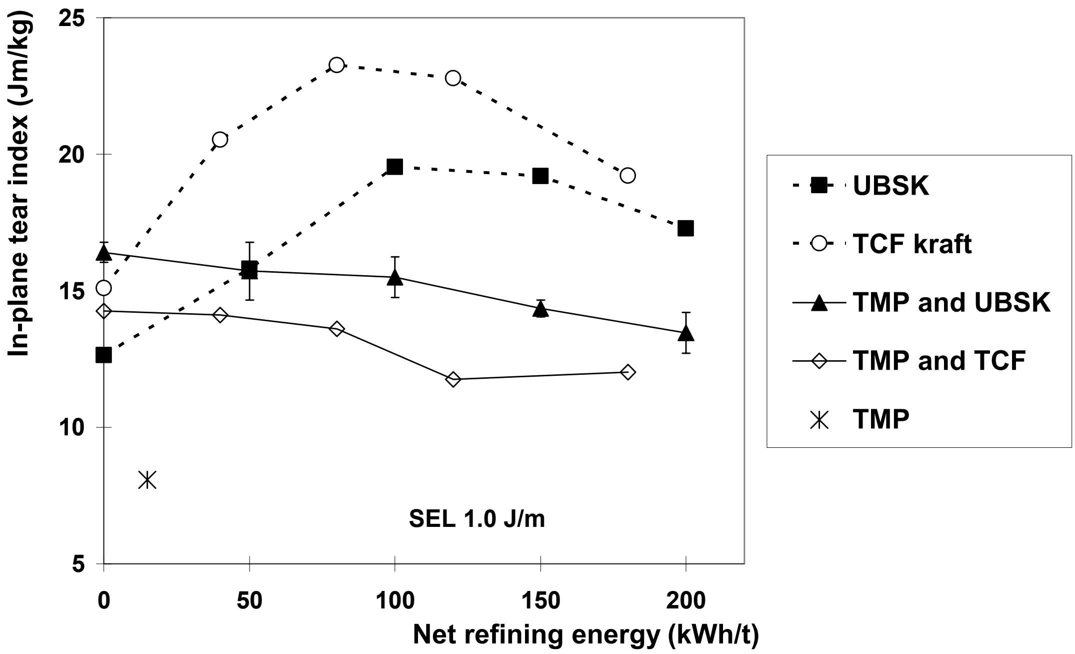

Figure 4-8. In-plane tear index of the kraft and the mixture series vs. refining energy of softwood kraft /II/. Mixture ratios: unbleached softwood kraft (UBSK) 50 % and TCF-kraft 40 % /II/. TCF was dried and UBSK and TMP were never-dried. Typical 95 % confidence levels shown in one curve. The position of the pure TMP on the x-axis is arbitrary.

Clearly the working hypothesis of fracture energy behaviour during reinforcement pulp refining was shown to be false /I,II,III,IV/. One of the main targets of the study became to understand the reasons for this behaviour. This question is tackled in chapters 4.3, 4.4 and 5.

Figure 4-9 shows that the in-plane fracture toughness of both pure kraft and that of a mixture increases during kraft refining. The fracture toughness (K_C) was calculated from specific elastic modulus and fracture energy according to (2). Both in chemical pulp series and in mixture series the fracture toughness levels off with continued refining. However, in the mixture series, the increase in fracture toughness is reasonably small and fracture toughness levels off earlier than with chemical pulp series. At a high level of refining the increase in elastic modulus seems to compensate for the decrease in fracture energy, as the fracture toughness does not decrease even though fracture energy does (Figures 4-7, 4-8 and 4-9). It was noted that sulphite gave as high or higher fracture toughness as kraft pulps – a clear difference to the ranking based on Elmendorf tear /IV/.

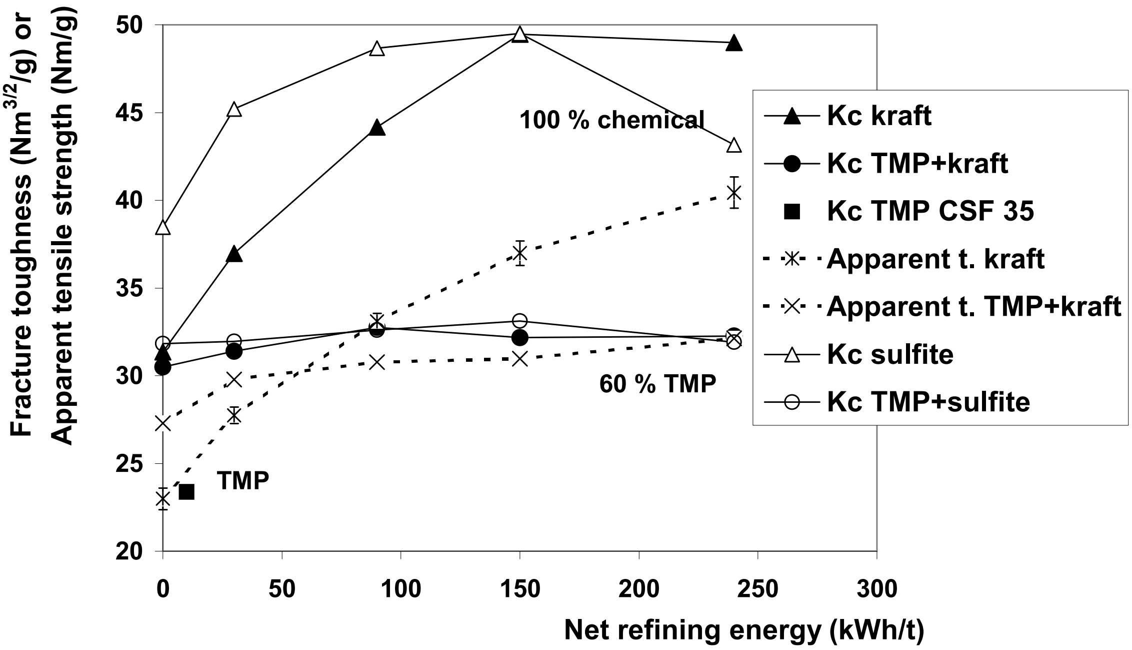

Figure 4-9. Effect of kraft refining on fracture toughness (K_C) of softwood kraft and softwood neutral sulphite and corresponding mixture sheets against refining. Fracture toughness calculated based on Equation (2) /IV/. Also apparent tensile strength shown for kraft and TMP+kraft mixture. Typical 95 % confidence levels shown in one curve.

Apparent tensile index should correlate with fracture toughness even though the former is not a material property but depends on test piece geometry /Niskanen 1998a/. According to Figure 4-9, this is the case here – the trend against refining is similar.

Apparent tensile strength depends among others on the flaw size of the test specimen. On the other hand, it is known that small enough flaws do not affect average strength of paper /Donner 1997, Hansen 2001/. Therefore, Niskanen et. al. /Niskanen 2001b/ have proposed a formula for calculating the apparent tensile strength (σ_{app}) of paper with flaws of different sizes. The new feature of Equation 4 is the use of damage width (size of fracture process zone) in the place of theoretical correction term /Niskanen 2001b/.

$$\sigma_{app} = \frac{\sqrt{G_C E}}{\beta\sqrt{2\pi(a + w_d)}} \tag{4}$$

Where G_C is the fracture energy, E the specific elastic modulus (or tensile stiffness), and w_d the damage width of paper. The factor β depends on the defect size, a, and on the specimen width. It can be noticed from Equation 4 that if the size of the flaw is smaller than the damage width, flaw has only small effect on the apparent tensile strength of the paper. In this study the damage widths of TMP-kraft mixtures were typically in the range of 1.5 to 3 mm.

The effect of reinforcement pulp beating on the breaking stress and breaking strain at different flaw sizes is shown in Figure 4-10. No flaws correspond to ordinary tensile strength and the flawed samples to apparent tensile strength (in SCAN-P 77:95 for Fracture toughness the flaw size is 20 mm). It can be seen that reinforcement pulp beating has a limited effect on the breaking stress of TMP-based paper with flaws over a certain size.

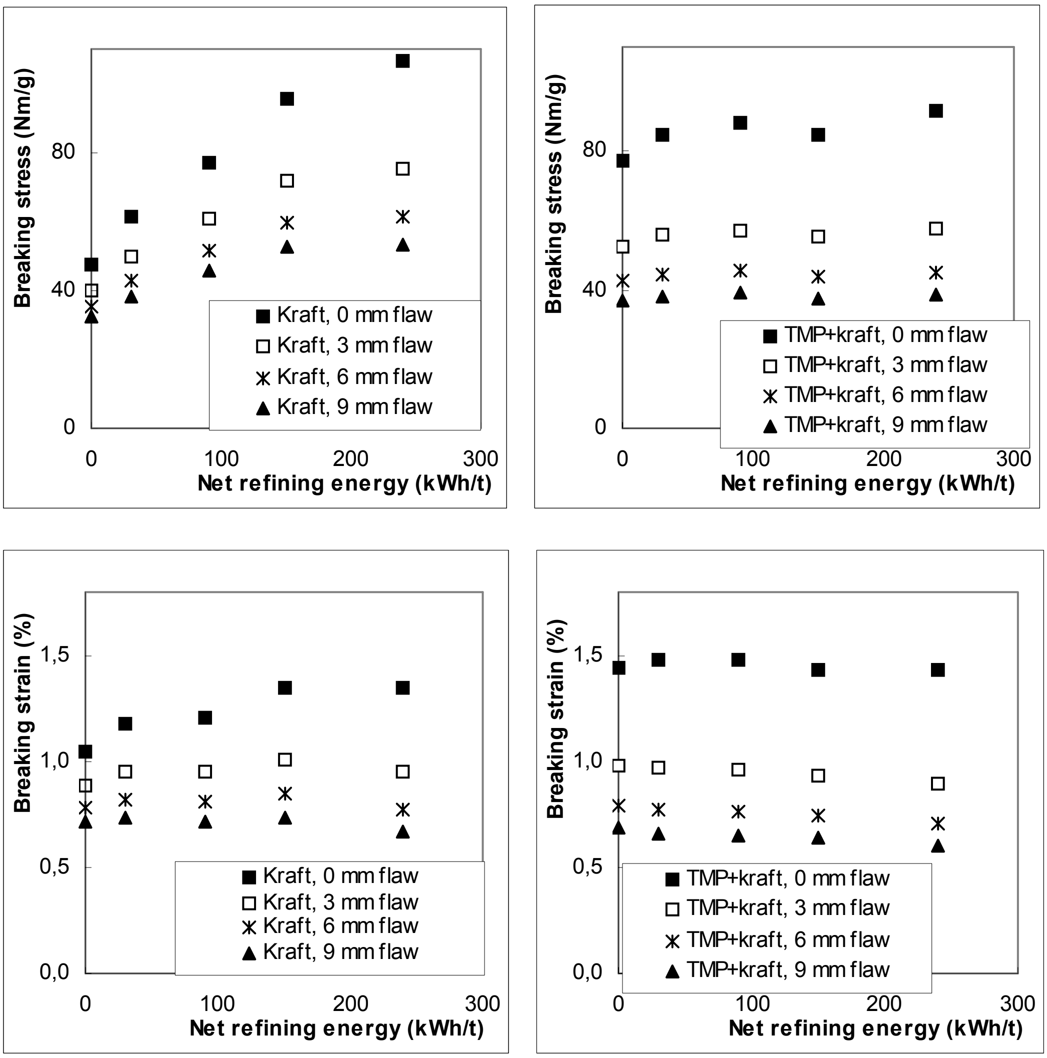

Figure 4-10. Breaking stress and strain vs. flaw size for conventional kraft pulp and corresponding mixture with TMP. Original data from /IV/ - values for breaking stress and strain calculated according Equation 4. In this figure the breaking strain is the elastic breaking strain component (σ_{app} / E).

If the behaviour of a running web is assumed to be linearly elastic then the elastic breaking strain component (σ_{app} / E) gives an approximation for the total breaking strain of the web. On the other hand while measuring the apparent breaking strain according to SCAN-P 77:95 stress-strain curve is often not linear. Therefore, the use of elastic breaking strain seems more relevant (even though our group used apparent breaking strain in /IV/).

The elastic breaking strain of flawed TMP-based paper even decreases with increased beating (Figure 4-10). For pure chemical pulp the elastic breaking strain increases slightly with small flaw sizes, but decreases in cases with a large flaw. The case with no flaw corresponds to the “ordinary” elastic breaking strain of Uesaka /Uesaka 1999/ apart from the fact that equation (4) is not always theoretically valid for the relatively narrow (15 mm) ordinary tensile specimen.

It is interesting to note that Fellers et. al. /Fellers 1999, Fellers 2001/ have reported that straight, beaten reinforcement fibres gave the highest critical force but they did not give the highest critical elongation. Curly fibres gave highest critical elongation (for test pieces containing a well-defined crack).

4.2.3. Z-directional strength

Z-directional strength properties of chemical pulp clearly increase during beating – the trend was the same for three different test methods: Scott-bond, z-directional fracture energy (W_{nip}) and z-directional tensile strength /VI/. The role of beating for the z-directional strength of the TMP/kraft mixture sheets is qualitatively similar to the pure kraft sheets behaviour, only the range of variation is much smaller in the mixture sheets /VI/.

4.2.4. Optimal beating

The optimal beating level seems to depend on the optimisation criteria, i.e. which strength property is considered to describe runnability. When beating is optimised either based on out-of-plane tear or in-plane tear or fracture toughness or tensile strength, often all of these give a different “optimum”.

If the flaw-resisting ability described by fracture toughness or apparent breaking strain is assumed critical for runnability, then reinforcement pulp should be beaten only lightly. On the other hand, the rheology of unflawed paper can be improved by reinforcement pulp beating, as tensile strength clearly increases.

This can also be interpreted so that the optimum beating for reinforcement pulp varies depending on the type of converting operation, which is considered to be most critical for the paper grade in question (e.g. printing press vs. coating unit).

4.3. Size of fracture process zone vs. beating

The damage width (w_d, ~size of the fracture process zone) decreased during reinforcement pulp refining, both with the pure kraft and the mixture series, as shown in Figure 4-11. However, the change was much smaller in the mixture series than in the pure kraft series. It is noted that with unrefined chemical pulp the damage width is large – fracture process zone seems to be much wider than the average fibre length. This indicates that unrefined chemical pulp sheet has a much looser structure than “normally” bonded paper.

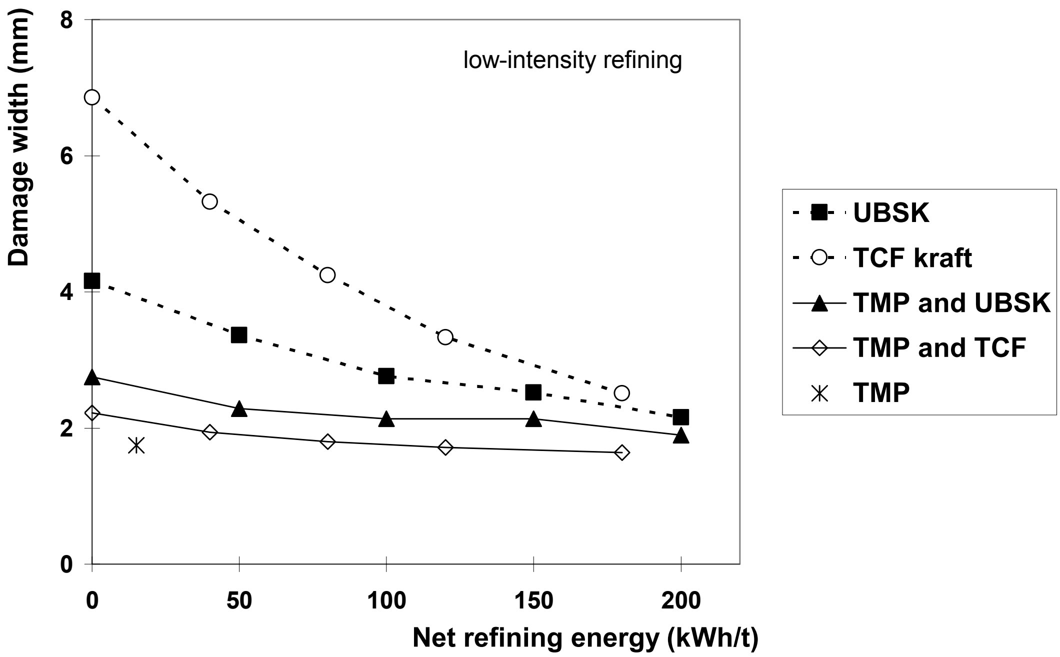

Figure 4-11. Damage width of the kraft and the mixture series vs. refining energy of softwood kraft (UBSK unbleached softwood kraft).

Figure 4-12 shows the change in pull-out width (w_p) with increased refining. The trend lines indicate that the proportion of broken fibres increases (compared to fibres being pulled-out as a whole) both in the pure kraft and the mixture series right from the beginning of kraft refining.

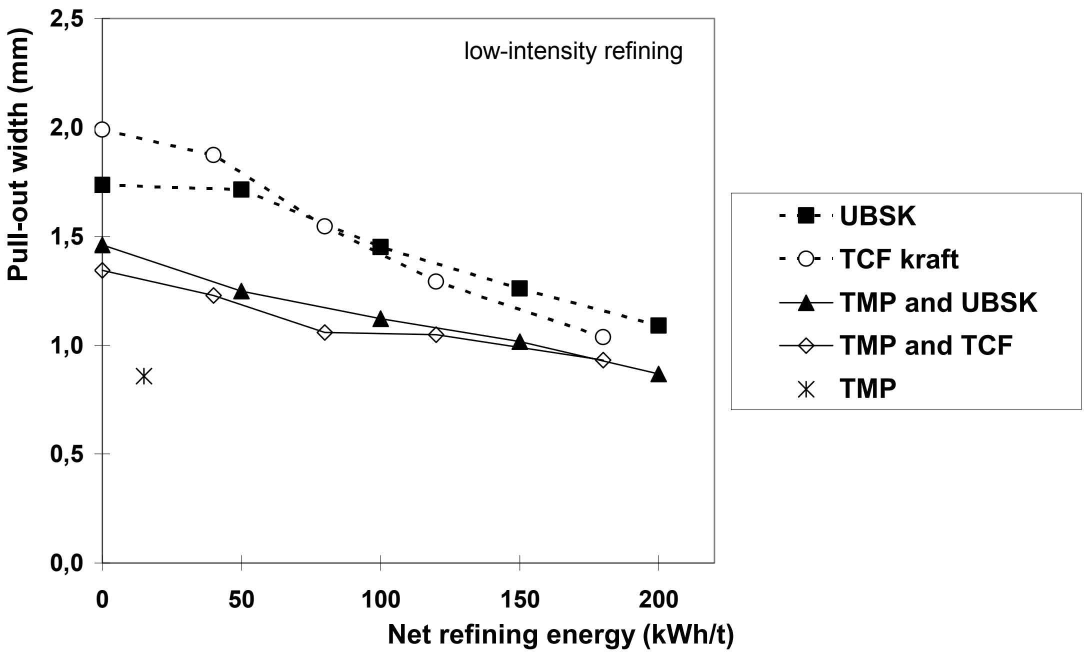

Figure 4-12. Pull-out width of the kraft and the mixture series vs. refining energy of softwood kraft.

It can be seen that fibre breakage alone does not explain the difference between the fracture energy behaviour of the pure chemical pulp and the mixture series. Though also with pure chemical pulp sheets fibre breakage (~w_p) decreased right from the start of refining, the fracture energy increased in the beginning of refining. In mixture series both fibre breakage and fracture energy decreased right from the start of refining.

The fracture energy per damage width (energy per area of the fracture process zone) of the kraft series increased clearly with kraft refining, while that of the mixture series was almost constant, as shown in Figure 4-13. In the mixture sheets, kraft refining apparently does not increase the energy needed to break the bonded fibre segments. The energy which is consumed during the fracture of bonded fibre segments goes mostly for plastic elongation of fibre cell wall material and related opening of interfibre bonds /Kettunen 1999a/. In summary: In the mixture series, the in-plane tear work per damage width remains constant even though the damaged area becomes smaller. This could be interpreted to mean that reinforcement pulp beating does not increase the interfibre bonding in mixture sheets /II/. The behaviour of in-plane bonding-dependent elastic breaking strain and fracture energy during beating supports the conclusion (Figures 4-5 and 4-8). However, strongly bonding-dependent z-directional strength does increase to a certain extent /VI/.

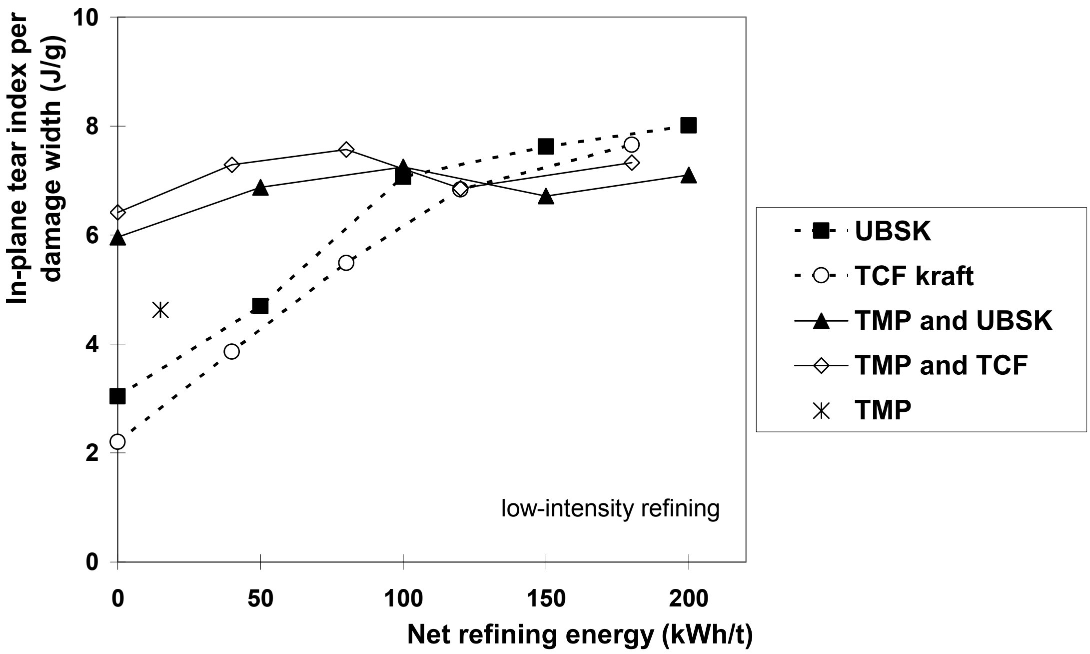

Figure 4-13. Energy density index (in-plane tear index/damage zone width) of the kraft and the mixture series vs. refining energy of softwood kraft /II/.

4.4. Additivity of pulp mixtures

4.4.1. Beating

According to the results, the fracture properties of a TMP-reinforcement pulp mixture are not linearly dependent on the component values. The fracture energy and fracture toughness of a mixture can be even higher than the corresponding values of the components, see Figures 4-8 and 4-9. The non-linearity of fracture properties seems to depend on the chemical pulp beating level so that the behaviour is strongly non-linear with unbeaten chemical pulp /II, III/.

According to Kettunen et. al. /Kettunen 2000c/ the damage width of "reasonably well-bonded" sheets is equal to about twice the arithmetic mean fibre length (Figure 4-14a). Furthermore, the fracture energy of reasonably well-bonded paper is almost uniquely related to the damage width /Kettunen 1999a, Kettunen 1999b/ - the trend line (y = 12x - 9) in Figure 4-14b illustrates this relation. Kettunen analysed a wide range of data and the only exceptions to the general correlation of fracture energy and damage width and fibre length were poorly bonded sheets /Kettunen 2000c/. Kärenlampi /Kärenlampi 1996/ has reported a similar correlation between fracture energy and fibre length, also there lightly beaten pure kraft was the only exception. The points that fall clearly on the right hand side of the trend line are considered poorly bonded /Kettunen 2000c/ (Figure 4-14b). It seems that in many practical cases involving mechanical pulp, interfibre bonding is sufficient, so that additional bonding does not increase the fracture energy.

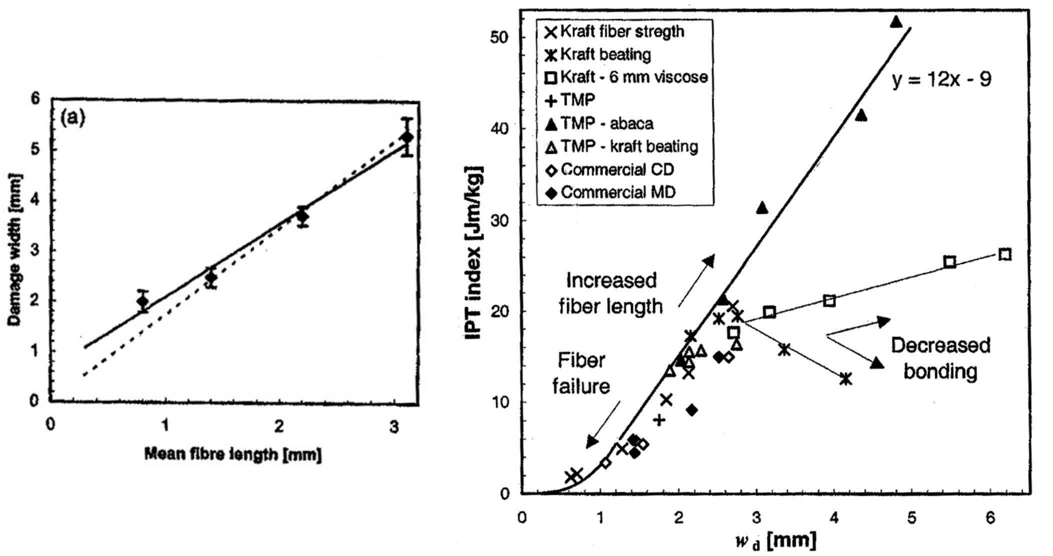

Figure 4-14a Damage width vs. mean fibre length /Kettunen 2000b/.
4-14b In-plane tear index vs. damage width /Kettunen 2000c/.

According to Kettunen /Kettunen 2000c/, cases which are not reasonably well-bonded are e.g. pure unbeaten softwood kraft or mixtures of poorly bonded viscose fibres and kraft. One additional poorly-bonded case seems to be the addition of TMP-long fibres to TMP, also there the correlation of average fibre length and fracture energy breaks /Lehto 2002/. Though the classification into well-bonded and poorly-bonded is qualitative, the classification seems to be a useful indirect measure of poor bonding.

Based on the above discussed classification, it can be seen that unrefined pure chemical pulp is poorly bonded in all the tested cases, see Figure 4-15. On the other hand mixtures of low-freeness mechanical pulp and unrefined chemical pulp are well bonded. The relatively high bonding ability of mechanical pulp partially explains the small effect of chemical pulp refining on the fracture energy and fracture toughness of mixture sheets. A suitable combination of long fibres (reinforcement pulp) and high-bonding-capacity fines (TMP) is likely to lead to this non-linear phenomenon /II/.

It seems that the fracture energy of chemical-mechanical pulp mixture primarily follows the average fibre length. Therefore, the fracture energy depends more on reinforcement pulp proportion and average fibre length than the beating degree. For example when four differently cooked reinforcement pulps (same raw material) were compared, it was seen that the beating level has only a minor effect on fracture energy and damage width of pulp mixtures, as shown in Figure 4-15. The clear effect of the pulp proportion on fracture energy can also be seen e.g. from Figures 4-8 and 4-15 and from /III/.

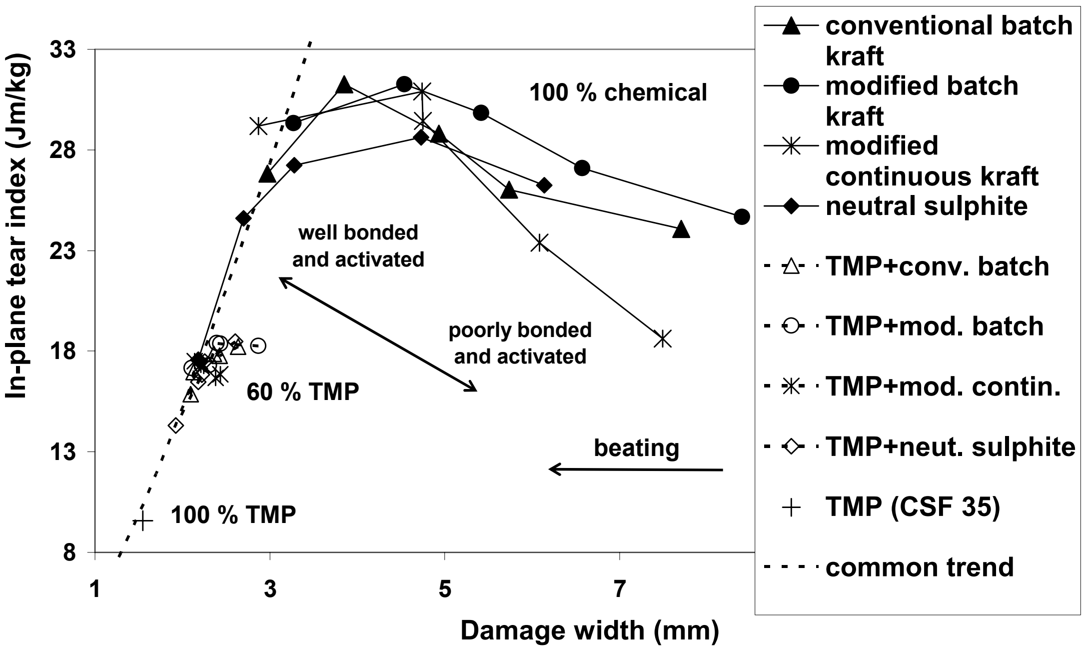

Figure 4-15. In-plane tear index vs. damage width for six chemical softwood pulps at different refining energy levels, corresponding mixtures with TMP and pure TMP /IV/. Chemical pulp refining varied from 0 to 240 kWh/t.

It seems that the fracture properties of mixture sheets cannot be predicted based only on the pure reinforcement pulp properties. Therefore the evaluation of the reinforcing ability of certain reinforcement pulp also requires information on the properties of the mechanical pulp in question.

The fracture energy and the damage width of mixtures could be estimated based on fibre length information of the pulp mixture, but the elastic modulus depends a lot on drying conditions (see chapter 5).

4.4.2. Fibre morphology

Fibre length seems to have a large effect on the in-plane fracture energy. Even low-bonding viscose fibres with sufficient length improve the fracture energy of softwood kraft-based paper /Yu 1999/.

It was found that too low fibre coarseness reduces reinforcement capacity if fibre failures increase in frequency /VII, Niskanen 2001a/. Also Kärenlampi et. al. /Kärenlampi 1997a/ have found that the fracture energy of pulp mixtures depends mostly on reinforcement pulp fibre length and not on the coarseness. An explanation for this could be that the in-plane fracture energy of reasonably well-bonded sheets arises entirely from the plastic elongation of fibres and related opening of bonds /Kettunen 1999a/. The general trend of Figures 4-14a and 4-14b would be difficult to understand without the importance of the strain of fibres /Niskanen 2001a/.

Low coarseness increases the contact area between fibres (number of fibres per gram of paper increases). However, because of the importance of the plastic elongation of fibres during fracture of paper, the effect of the increased fibre contact area on the fracture energy can be small. Interfibre bonding interfaces are just a small fraction of the total material (and hence of the molecular bonds) in the fibres /Niskanen 2001a/.

These results appear to disagree with those of Seth /Seth 1996/ who, however, used quite coarse fibres compared to the fibres used in this study (e.g. abaca). From fracture energy measurements on pure kraft handsheets Seth /Seth 1996/ concluded that fine softwood fibres are superior, irrespective how the comparison is made. On the other hand, it has been reported that high coarseness in beneficial for out-of-plane tear /Seth 1988, Seth 1996/.

Lower levels of bonding (very mild beating, filler addition) may naturally alleviate the problem of excessive fibre failures of low-coarseness fibres. On the other hand, high coarseness may also be detrimental. If nothing else, more beating may be required than at lower coarseness to achieve sufficient tensile strength and elastic modulus /VII/.

Retulainen /Retulainen 1997, Retulainen 1998/ has concluded that the main reason for non-linear additivity behaviour of pulp mixtures is not the weakness of interfibre bonds but the lack of activation to bear load for the chemical pulp fibres. In support of that it has been concluded /Niskanen 2001a/ that the points on the right hand side of the trendline on Figures 4-14b and 4-15 (e.g. unbeaten kraft) are not just poorly bonded but also poorly activated. Bonding vs. activation is discussed more thoroughly in chapter 5.

It has also been reported for reinforced mechanical pulp-dominated paper that fracture energy and damage width do not show a percolation transition at low grammages /Niskanen 2001a/. Percolation argument says that the reinforcement fibres themselves have to form a continuous network in order that they contribute to paper strength /Ritala 1987/. Based on the above discussion, it has been concluded in KUMOUS project /Niskanen 2001a/ that the percolation argument of the threshold content of reinforcement pulp is not seem valid.

Above all, high fibre length seems much more important a factor than coarseness for improving the flaw-resisting ability of paper /VII/. On the other hand, Paavilainen /Paavilainen 1994/ has reported that low coarseness (low cell-wall thickness) gives higher Scott-bond and higher tensile strength /Paavilainen 1991/. Therefore low coarseness can have a positive effect on interfibre bonding and rheological properties of unflawed web.

4.4.3. Fibre length vs. fibre strength

When fibre strength is decreased by acid vaporization of paper, the fibres may suffer from serious failures during paper fracture, and consequently the out-of-plane tear decreases /Seth 1988/. Also in-plane fracture energy decreases by acid vaporization /Seth 1996, Kärenlampi 1997b/. Both the proportion of broken fibres and damage width decrease in acid treatment, demonstrating an increase in the amount of fibre failures /Yu 2001/.

In many practical cases, especially in pulp mixtures, the differences in fibre strength are not as large as those induced by acid treatment. In one experiment the cooking method was varied in order to achieve different levels of fibre strength and bonding ability /Lindholm 1999, Niskanen 2001a/. The results of the damage analysis suggest that refined neutral sulphite fibres break easier and are less ductile than kraft fibres (pure chemical pulps) /IV/. However, this did not seem critical for the in-plane fracture properties of paper. Sulphite pulp gave a little higher or the same fracture toughness (Figure 4-9) and fracture energy as the kraft pulps (pure chemical) /IV/. In contrast to the pure chemical pulps, the TMP-based sheets showed no significant differences between the fibre properties of sulphite pulp and kraft pulps, see Figure 4-9 /IV/.

In kraft pulping, the bleaching method influenced interfibre bonding, fibre segment activation and fibre strength of pure chemical pulps /Niskanen 2001a/. However, in a mixture with TMP the differences in the interfibre bonding ability disappeared. The cooking or bleaching method giving highest fibre length improved the fracture energy of mixture sheets best /Niskanen 2001a/. Fibre length seems more important than fibre strength. However, fibre strength has to be high enough.

5. MECHANISM OF BEATING EFFECT

Often the effect of beating on strength (e.g. tensile, elastic modulus, Scott-Bond) is explained simply by increased interfibre bonding. Most frequently it is assumed that bonding improves because beating increases fibre flexibility, this explanation was suggested already in the 1930's by Campbell /Campbell 1933, Page 1985/. However, it is known that if sheet density is increased by wet pressing instead of beating, the tensile strength and elastic modulus differ at constant RBA – beating gives higher tensile and elastic modulus /Giertz 1964, Page 1979, Page 1985/ (Figure 5-1) and higher elastic breaking strain /Niskanen 1998a/. This can be interpreted as evidence that it cannot be just the increase in RBA that explains the beating effect. In addition to increased bonding, there should be (at least) one other mechanism explaining the behaviour. Based on the literature review (see Chapter 2) it is assumed that in low-intensity beating the effect of both external fibrillation and secondary fines is small.

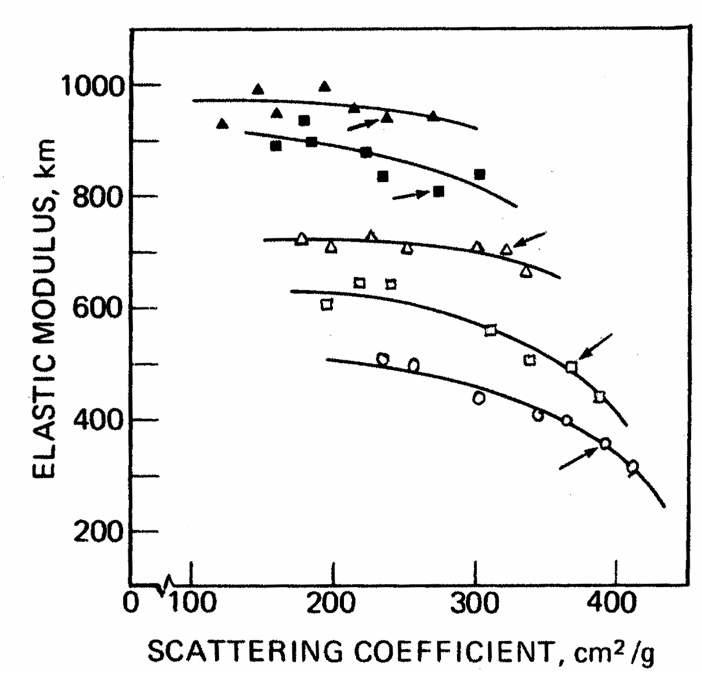

Figure 5-1. Elastic modulus vs. scattering coefficient for sheets made of bleached kraft pulp of eastern Canadian white pine /Page 1979/. The lines are for constant beating but varying wet-pressing pressures (arrows indicate data for the standard pressure 345 kPa). Beating increases from bottom to the top (from 0 to 59 minutes).

It is known that straining during drying increases the tensile strength and elastic modulus /Htun 1980, Zhang 2001/ but not Scott bond, actually z-directional strength decreases /Parsons 1972, Wahlström 2000, V/. This has been explained by a decrease in the RBA /Wahlström 2000/. Therefore, the increase in tensile strength and elastic modulus because of straining during drying seems to be because of some other effect than bonding (as z-directional strength properties decrease). On the other hand beating increases tensile strength and elastic modulus and also Scott bond /V/. Other z-directional measurements (z-fracture toughness and z-tensile) show a similar trend as Scott bond during beating /VI/. It seems clear that wet straining and beating affect strength through different mechanisms.

Here it is assumed that the other effect is the activation, as suggested originally by Giertz /Giertz 1964, Giertz 1979/. He has argued that the increase in tensile strength during restraint drying is caused by increased fibre segment activation. During drying, fibres shrink and through the microcompression effect (suggested by Page and Tydeman /Page 1962/) create drying stresses in the network. These stresses straighten slag fibre segments enabling them to carry load more "actively" /Giertz 1979/. Beating increases fibre swelling and therefore also fibre shrinkage during drying, which makes the activation effect stronger /Giertz 1964/.

Another explanation for the effect of beating on elastic modulus is given by van den Akker et. al. /van den Akker 1966/ and that is the increased *axial* elastic modulus of fibres ("Jentzen-effect"). Jentzen /Jentzen 1964/ has shown that when single fibres are dried under straining their elastic modulus increases. Beating increases fibre swelling which then increases drying stresses. Therefore beating would increase the axial elastic modulus of fibre segments in paper /van den Akker 1966/.

Page, Seth and De Grace /Page 1979/ have suggested that the elastic modulus of paper depends only on three factors: 1. Elastic modulus of fibres 2. Degree of bonding 3. The presence of curl, kinks, crimps or microcompressions. For effect of beating they suggest that first beating swells fibres, thereby removing crimps and kinks and second beating also increases activation during drying. Third, beating increases the bonding degree. Later Page /Page 1985/ has somewhat modified his view about activation. He argues that tensile strength develops during beating mainly because of the straightening of fibres (during beating itself) – not because of activation. Fibre and fibre segment straightening makes them more ready to take load and improves the stress distribution in the fibre network and therefore both the elastic modulus and the tensile strength increase /Page 1985/. According to direct experimental results the elastic modulus of well-bonded paper with straight fibres comes from the elastic modulus of component fibres /Page 1980/.

An additional view is given by Retulainen et. al. /Retulainen 1998/: paper does not consist of homogeneous fibres whose mechanical properties are single-valued; instead the properties of individual fibres vary (in addition to natural variation) because of local network geometry and the drying stresses during drying. He has concluded that the main reason for non-linear additivity behaviour of chemical-mechanical pulp mixture is not the weakness of interfibre bonds but the lack of activation of the chemical pulp fibres /Retulainen 1997/.

Niskanen /Niskanen 2000/ has suggested that beating affects *elastic modulus* mostly by activation and only a minor effect is due to increased bonding (in the dry paper). He argues that beating increases fibre swelling which then increases the drying stresses during restraint drying. Increased drying stress straightens fibre segments, that is, activates them. Based on his concept, the bonding in *wet paper* is critical for activation – not the bonding in dry paper. Page and Tydeman /Page 1962/ have presented the idea of "wet bonding" and have connected fibre shrinkage during drying to drying stresses. According to Niskanen /Niskanen 2000/ beating in addition to activation also increases the axial elastic modulus of single fibre segments through the Jentzen-effect. In normal grammage range of paper fibre length seems to have only a small effect on elastic modulus /Yu 1999, Niskanen 2000/.

In this study it is hypothesized based on the above discussion that the factors that contribute to *all the mechanical properties of dry paper* are drying stresses and inter-fibre bonding in the dry paper /VII/, see Figure 5-2.

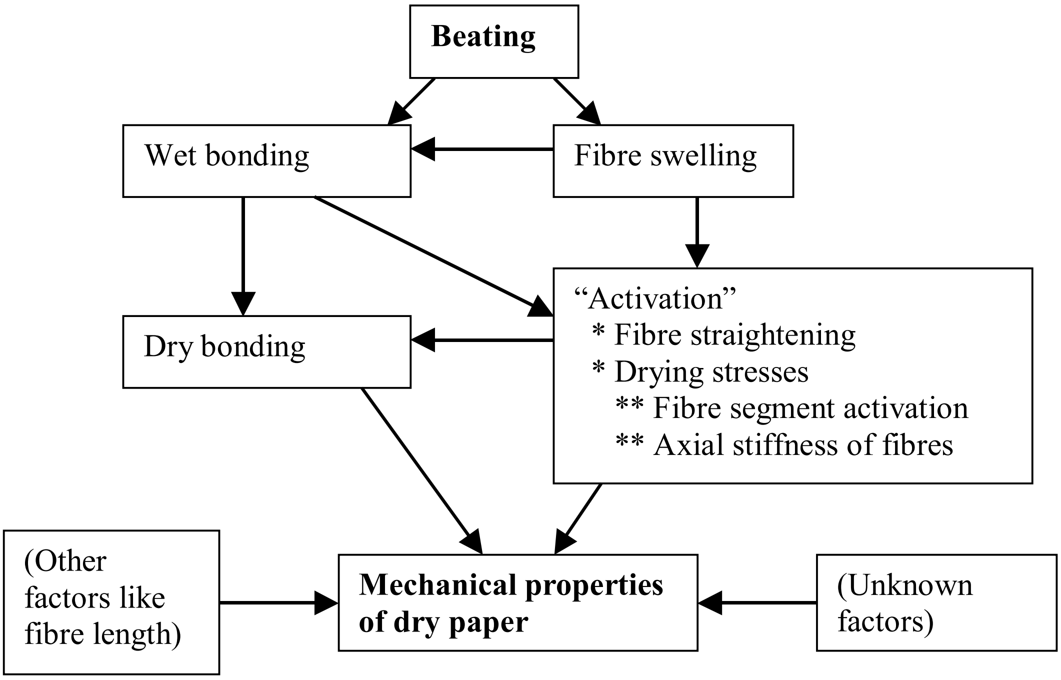

Figure 5-2. Simplified scheme of the mechanism how beating is assumed to affect the mechanical properties of dry paper. Arrows show the direction of the assumed effect.

Drying stress improves fibre segment activation and increases axial fibre stiffness in restraint-dried handsheets. Increasing beating should contribute to the elastic modulus through both increased swelling and increased bonding. The first effect influences the drying stresses and activation /VII/. For example dry strength agents like starch affect only bonding and not fibre segment activation.

In the following, the hypothesis that beating affects paper strength both through increased bonding and increased activation is compared to experimental data.
The effect of beating is compared to selected other papermaking variables. The analysis concentrates first on the pure chemical pulp sheet properties in order to understand the mechanisms through which beating affects paper strength. Softwood chemical pulp beating is compared to straining during drying (wet straining), filler addition and starch addition in order to better understand the effect of beating on paper strength. Second, the effect of beating on TMP-containing sheets is studied.

5.1. Straining during drying vs. beating

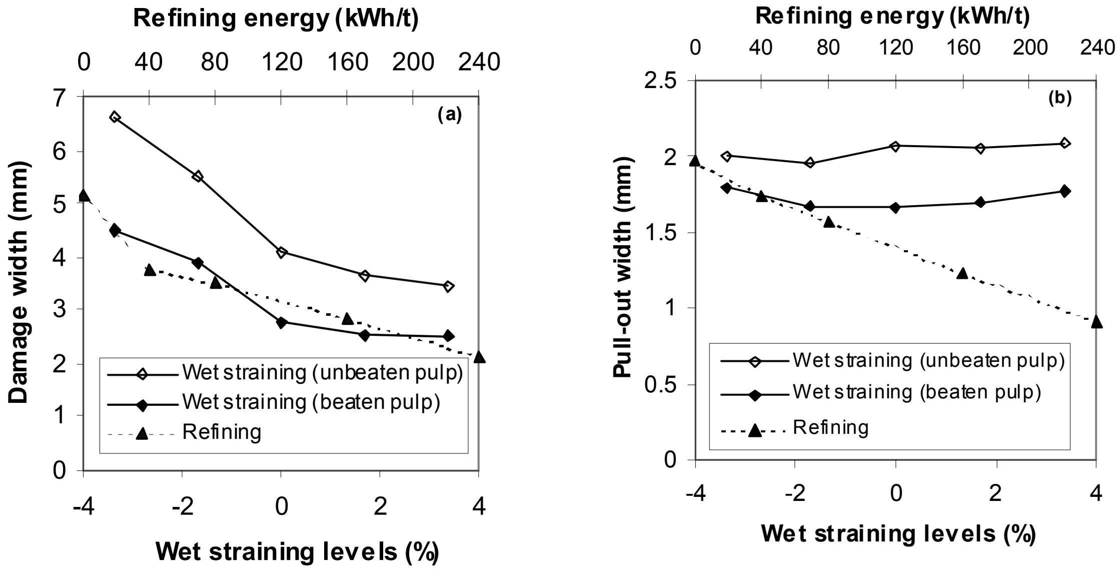

Figure 5-3. Effect of wet straining levels and refining (dried pulp) on the damage width (a) and pull-out width (b). For solid lines (wet straining), use the bottom x-axis. For dashed lines (refining) use the top x-axis /V/.

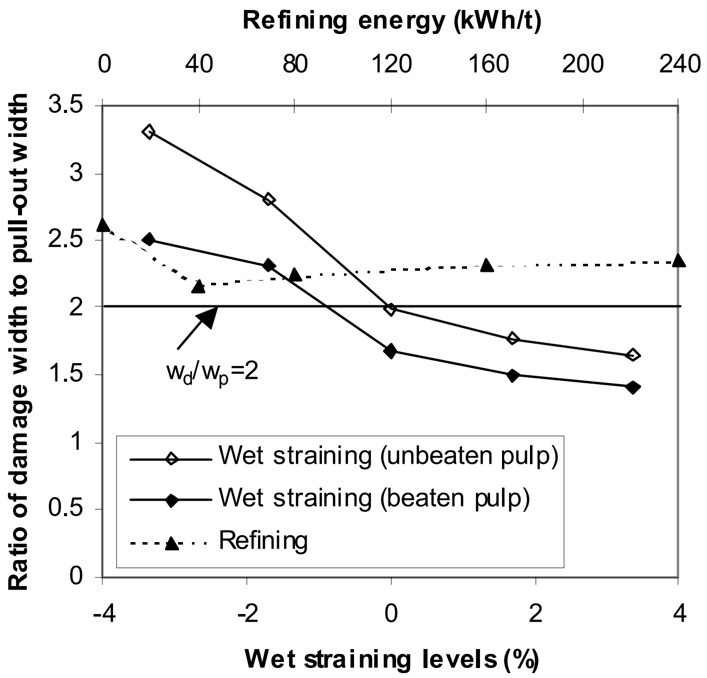

Figure 5-4. Effect of wet straining and refining (dried pulp) on the ratio of damage width to pull-out width. For solid lines (wet straining), use the bottom x-axis. For dashed lines (refining) use the top x-axis /V/.

Both damage width and pull-out width decrease during beating. However, increased wet straining was found to decrease damage width whereas pull-out width remained almost constant, as shown in Figure 5-3. Figure 5-4 shows the ratio of damage width to pull-out width during wet straining and beating (~ratio of the size of the damaged area to the amount of single fibre debonding). During increased wet straining, the ratio drops, while during beating the ratio first drops but soon levels off.

If drying shrinkage is hindered /Seth 1993/ or wet straining applied /V/, then the in-plane fracture energy decreases. Although increasing wet straining reduces bonding (e.g. G_C/w_d), its effect on the fracture energy, G_C, seems to come primarily through a reduction in the area of the fracture process zone (damage width, w_d) /V/. This could be interpreted to mean that activation decreases the size of the fracture process zone. As the pull-out width, w_p, does not decrease simultaneously with the damage width, w_d, it indicates that fibre breakage does not increase during wet straining.

If we assume that the main mechanism during wet straining is activation, then the ratio w_d/w_p may serve as an indication of the activation of fibre segments /V/. Based on Figures 5-4 and 5-5 showing the w_d/w_p ratio during beating, it could be concluded that beating increases fibre segment activation especially in the beginning of beating (pure chemical pulp sheets).

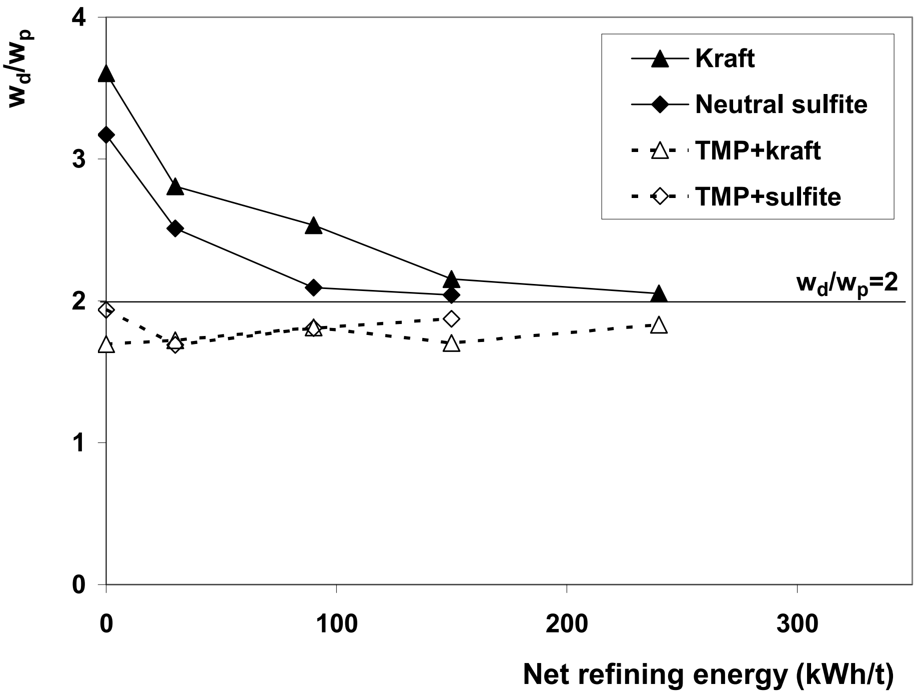

Figure 5-5. w_d/w_p (damage width/ pull-out width) ratio for conventional kraft pulp and neutral sulphite pulp during beating and corresponding mixtures with TMP (50 % TMP). Data from /IV/.

5.2. Filler and starch vs. beating

When comparing the effect of filler and starch addition to beating, the first assumption is that both filler and starch affect only bonding and not drying stresses or activation.

In this analysis paper strength properties were compared against fibre network density, i.e. specific elastic modulus and in-plane tear (IPT) index are expressed using only the mass of the fibres and not the mass of the whole sheet grammage as in publication /VI/. Mass of starch is included in the “mass of the fibres”.

The elastic breaking strain (tensile/elastic modulus) during beating, filler addition and starch addition on kraft is shown in Figure 5-6 /VI/. Elastic breaking strain is considered to depend on inter-fibre bonding /VI/. Increasing beating and increasing filler content seem to have an exactly opposite effect on elastic breaking strain against network density. On the other hand, increasing starch content has a clearly different effect, Figure 5-6. An increase in starch dosage does not affect network density but elastic breaking strain increases. The tested z-directional properties: fracture energy (W_{nip}), Scott-bond and z-directional tensile strength all show similar behaviour as elastic breaking strain when comparing beating, filler and starch effects /VI/. The effect of beating clearly differs from the effect of starch addition, suggesting that beating affects also by some other mechanism than starch.

It is somewhat surprising to note that the effect of filler addition resembles that of decreasing beating and not that of decreasing starch addition as it would seem logical that starch increases bonding while filler decreases bonding and that neither affects activation. Both elastic breaking strain and z-directional fracture energy should mostly depend on interfibre bonding and not on activation. Starch seems to increase bonding without changing the network density, which is natural, as starch does not change fibre swelling and therefore not drying stresses either. However, also filler addition does not change fibre swelling but still decreases network density. Possibly filler located between fibres disturbs the network structure thereby decreasing density. In addition to that decreased bonding caused by filler could decrease the compressive shrinkage forces on the crossing fibres during drying (Page-Tydeman effect /Page 1962/). This decreases the drying stresses. A problem with this explanation is that accordingly starch addition could increase drying stresses because of additional bonding. On the other hand, maybe starch affects bonding only in dry paper so that the drying stresses have already had their effect, while filler disturbs bonding already in wet state (wet bonding). In support of this idea it has been reported /Retulainen 1997/ that starch increases the drying stresses only marginally.

From Figure 5-7 it is seen that starch addition had only a small effect on specific elastic modulus. This could be interpreted to mean that elastic modulus depends more on activation/drying stresses than on bonding. The observation that straining during drying has a large effect on elastic modulus /Htun 1980, Zhang 2001/ supports the idea that activation (instead of bonding) dominates elastic modulus. An indirect evidence of the role of activation on elastic modulus is that elastic modulus correlates with fibre swelling (measured by FSP method) when comparing several beaten softwood pulps /Mäkinen 2000/. As beating increases elastic modulus also in the mixture series where bonding does not increase, it seems that beating affects mixture series mainly through activation.

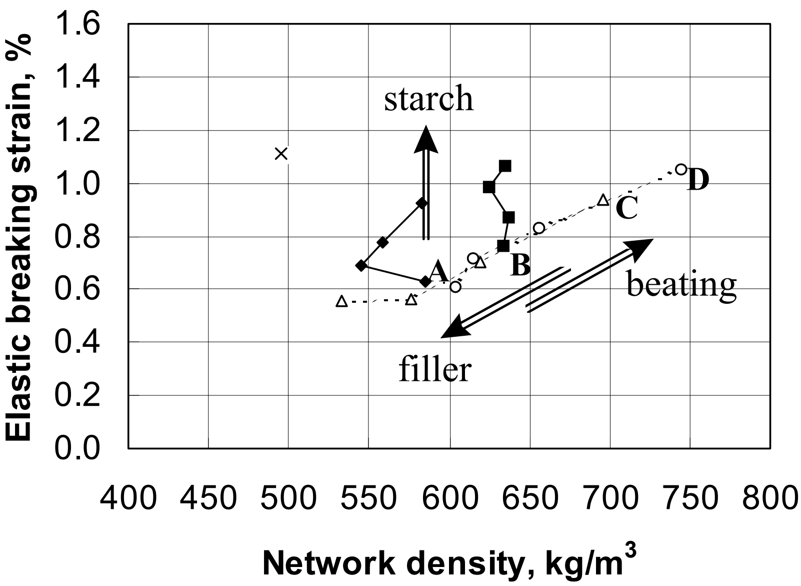

Figure 5-6. Elastic breaking strain against network density for pure TMP (x-symbol) sheet and pure softwood kraft sheets; unbeaten with different content of starch (close diamond), lightly beaten with starch (close square), medium beaten with filler (open triangle), and heavily beaten with filler (open circle). Increasing beating without starch/filler corresponds to points **A—D** /VI/.

Since filler does not alter fibre swelling, it also should have a somewhat different effect compared to beating. It is therefore reasonable that elastic modulus did not decrease during filler addition as rapidly against network density as during decreased beating, Figure 5-7 /VI/. This supports the hypothesis that increasing beating contributes to the specific elastic modulus also through swelling and drying stresses.

5.3. Charge-induced swelling vs. beating

It seems that whether certain level of fibre swelling is achieved by modifying fibre charge or by increasing low-intensity beating, the effect on paper strength is very similar (the curves concur), see Figure 5-8. This supports the importance of fibre swelling (internal fibrillation) as a beating effect. It seems that the interfibre bonding (~G_C/w_d) and activation (~elastic modulus) are primarily controlled by fibre swelling independent of charge. Fibre swelling by beating increases drying stresses /Zhang 2001/ and also straightens fibres /Seth 2001/.

As charge-induced fibre swelling is close to the beating effect, it indirectly supports the idea that fibre swelling-induced activation is an important beating effect.

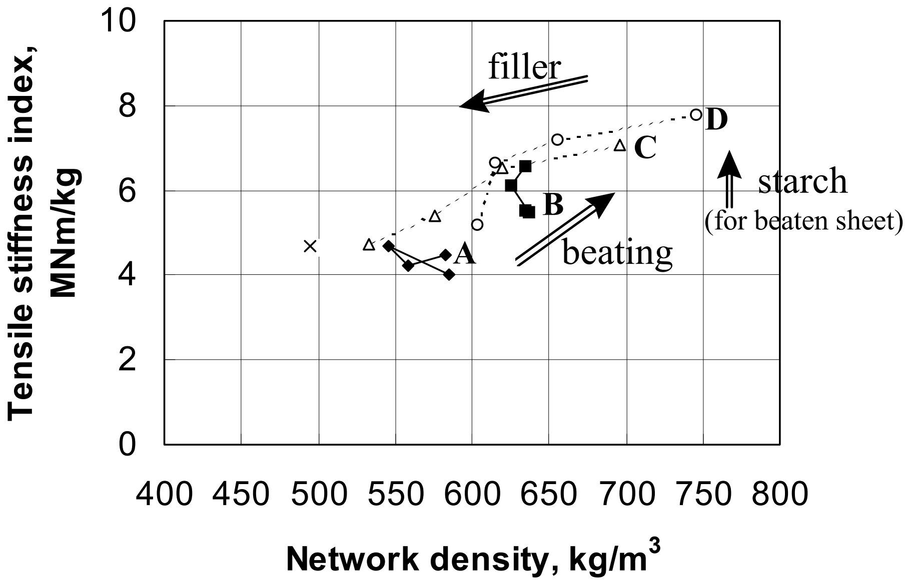

Figure 5-7. Tensile stiffness index (specific elastic modulus) against network density for pure TMP (x-symbol) sheet and pure softwood kraft sheets; unbeaten with different content of starch (close diamond), lightly beaten with starch (close square), medium beaten with filler (open triangle), and heavily beaten with filler (open circle). Increasing beating without starch/filler corresponds to points **A**—**D** /VI/.

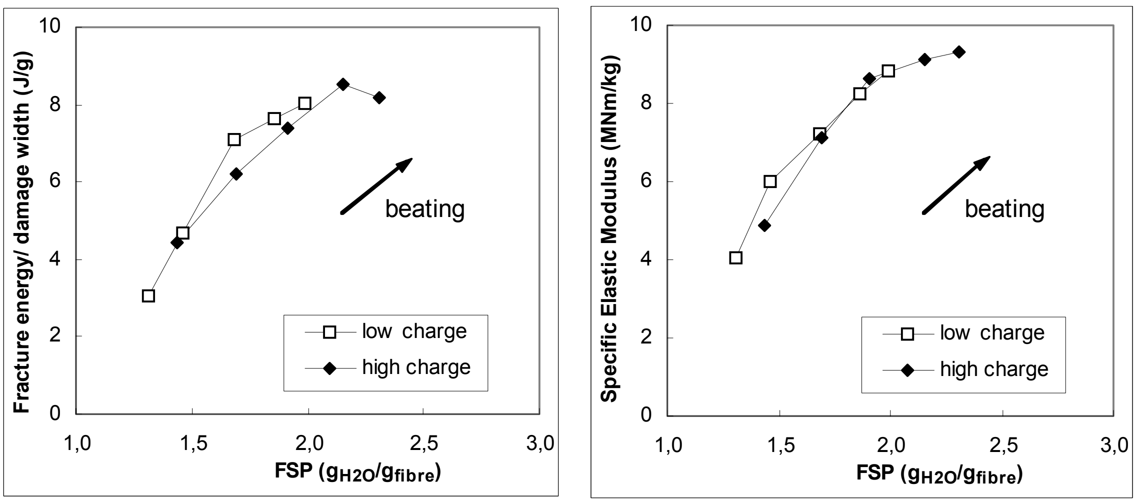

Figure 5-8. Fracture energy per damage width (G_C/w_d) and elastic modulus vs. fibre swelling (measured by FSP method) at two charge levels and several beating levels. Low-intensity Escher-Wyss beating (SEL 1.0, from 0 to 200 kWh/t).

5.4. Mixture sheets

Also with mechanical pulp-dominated sheets drying restraint increases the elastic modulus /Htun 1980, Zhang 2001/ and decreases the fracture energy /Zhang 2002/ and z-directional strength. However, the free shrinkage of mechanical pulp-containing sheets is clearly smaller than that of chemical pulp sheets /Mäkinen 2000/. Swelling (FSP) of mechanical pulp fibres is low /Maloney 2000/. On the other hand, fibre shrinkage behaviour (trend of shrinkage against water content) of mechanical pulp fibres is similar to that of beaten chemical pulp fibres /Nanko1991, Weise 1997/. Reports about the amount of fibre shrinkage vary – either mechanical pulp fibres shrink less /Nanko 1991/ or the same amount /Weise 1997/ as beaten chemical pulp fibres. The drying stress of low-freeness TMP sheets is between the level of unbeaten and beaten softwood krafts /Zhang 2001/. The relatively high drying stress of mechanical pulp could be because of high RBA (fines).

Considering the role of inter-fibre bonding for the mixtures sheets, it was noted that the pure TMP sheets have almost the same elastic breaking strain as the pure softwood kraft sheets at high beating level or at a high starch content /VI/. In comparison, the specific elastic modulus of the pure TMP sheets is lower than that of beaten kraft sheets. This indicates that lower tensile strength values of TMP are caused by the low specific elastic modulus of the paper (~ low activation or low axial stiffness of fibres) and not by poor inter-fibre bonding /VI/. According to Page et. al. /Page 1977/ the axial elastic modulus of fibres is almost the same independent of pulp yield (mechanical pulp not measured). The elastic modulus of fibres is controlled by fibril angle /Page 1977/. Therefore, the explanation for the lower elastic modulus of TMP sheets seems to be the lower fibre segment activation of TMP compared to beaten kraft.

The effect of beating on (in-plane) bonding of mixture sheets can be seen from G_C/w_d (fracture energy/damage width, Figures 4-13 and 5-9) and elastic breaking strain (Figure 4-5). It seems that chemical pulp beating cannot much increase the G_C/w_d or elastic breaking strain of mechanical pulp-based paper. However, z-directional strength increases slightly /VI/. Also elastic modulus increases, as shown in Figures 4-4 and 5-9. It could be concluded that because of low activation of pure TMP, kraft beating does improve activation-dependent properties like elastic modulus.

On the other hand, the w_d/w_p (damage width/pull-out width) ratio of mixture sheets does not decrease during beating, Figure 5-5. It is below the “critical” value 2 already with unbeaten reinforcement pulp - this indicates good activation in all points. Possibly, the w_d/w_p ratio is not a suitable indicator of activation in mixture sheets? The analysis based on elastic modulus is in agreement with Retulainen´s /Retulainen 1997/ conclusion regarding the important role of activation in mixture sheets.

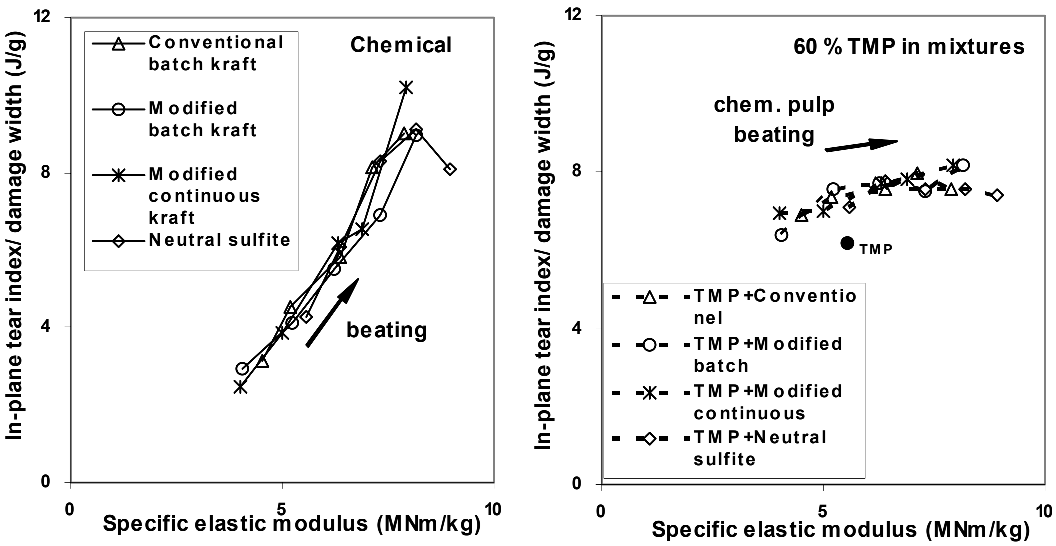

Figure 5-9. Fracture energy per damage width (G_C/w_d) vs. specific elastic modulus (both for pure chemical pulp and mixture) during beating /Niskanen 2001a/. Data from publication /IV/.

Starch addition to the chemical pulp leaves the fibre network density unchanged (opposite to refining) but leads to a clear improvement in all the mechanical properties considered. Starch addition improves the z-directional strength properties also in the mixture sheets, even though the in-plane properties do not change at all /VI/. The sensitivity of the out-of-plane properties to starch content should originate from the different role of interfibre bonding /VI/. This further supports the hypothesis that in order to achieve good in-plane strength (e.g. elastic modulus and tensile) for mixture sheets, fibre segment activation is needed.

When fines are removed from TMP, the fracture energy/damage width ratio, G_C/w_d, (~bonding) clearly decreases (Figure 5-10) /III/. However, fracture energy of pure TMP decreases only a little and damage width increases clearly /III/. This is because fracture energy is not monotonously correlated to bonding or activation. The increase in damage width is caused by decreased bonding and activation. Fines removal causes a bigger drop in bonding than 20 % filler addition – the drop in activation (~elastic modulus) of pure TMP is the same in both cases (Figure 5-10).

During kraft beating the G_C/w_d ratio of the mixture without TMP fines increased but did not quite reach the value of a "normal" mixture, Figure 5-10. It seems that beating cannot fully compensate for the drop in bonding caused by fines removal. According to Retulainen /Retulainen 1997/ and Nieminen /Nieminen 1994/ fines affect mostly the RBA (and not specific bond strength like starch). Retulainen /Retulainen 1997/ has speculated that TMP fines can better "fill" the interfibre bonds in the stiff mechanical pulp-controlled network than kraft refining does. These results support that idea.

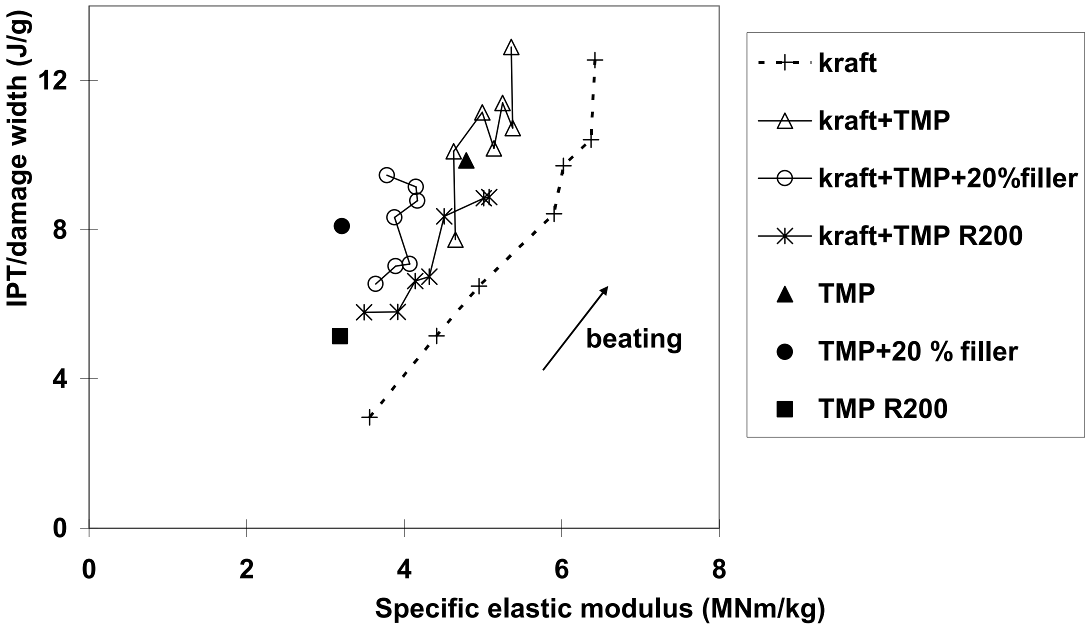

Figure 5-10. In-plane tear index (IPT) per damage width (G_C/w_d) against specific elastic modulus vs. during beating, filler addition and TMP-fines removal /III/. TMP fraction retained on a 200 mesh screen is expressed as TMP R200.

5.5. Conclusions of beating effect

The effect on RBA is not the only mechanism of beating, as RBA increased by wet pressing does not increase elastic modulus or tensile strength as much as beating-induced increase does /Giertz 1964, Page 1979, Page 1985/. The beating effect is also different from the effects of starch or filler or TMP-fines /III,VI/. On the other hand, the effect of beating seems similar to the effect of charge induced fibre swelling /VIII/. It seems clear that beating does not affect the strength and structure of paper simply by increasing interfibre bonding. It was concluded that beating affects paper mainly through three mechanisms: 1. Increased fibre segment activation 2. Increased interfibre bonding 3. Decreased fibre length, see Figure 5-11. Fibre straightening is considered as part of the activation effect. In low-intensity beating, significant fibre shortening occurs only at high refining energy levels. Therefore, in the earlier phases of beating the first two effects are the most important ones.

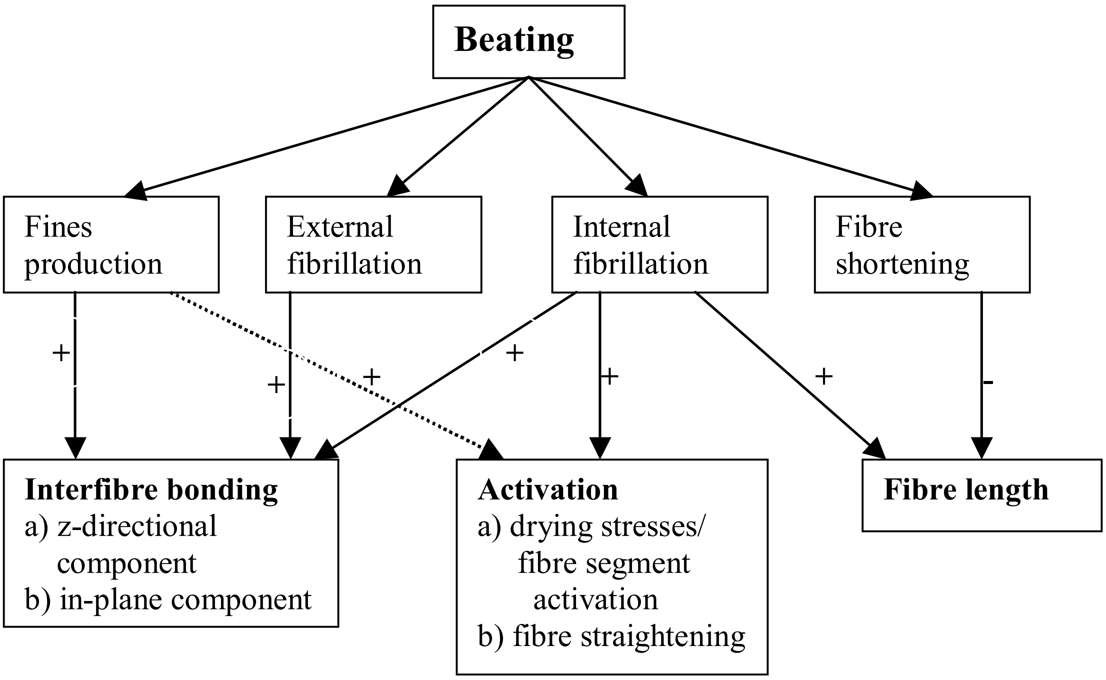

Figure 5-11. Simplified scheme of the beating mechanism. + signifies positive correlation and - negative correlation. Dotted line signifies a weak effect.

However, the analysis of activation is difficult, as there is no direct measurement for fibre segment activation. In this study, the elastic modulus was often used to indicate activation, though elastic modulus is known to depend also on bonding /Page 1979, Niskanen 2000/. However, it seems that the effect of bonding on elastic modulus reaches saturation level sooner than some other effects /Page 1979/, see Figure 5-1. The other analysed indicator for activation, the ratio of damage width to pull-out width (w_d/w_p), gave poorer results.

For analysing bonding, there seems to be reasonable indirect methods like certain z-directional strength properties and G_C/w_d (fracture energy/damage width) and elastic breaking strain. However, it is known that all z-directional measurements do not correlate with each other /Lundh 2001/. Despite certain practical and theoretical problems, z-directional measurements seem relevant at least for selected cases with laboratory sheets /Niskanen 2001a, VI/. It seems logical to assume that activation has a marginal effect on z-directional strength (see Figure 5-12).

One question is whether the in-plane bonding (e.g. G_C/w_d or elastic breaking strain) and z-directional bonding describe the same thing? As the loading mode is clearly different, it seems logical to assume that in-plane and out-of-plane bonding do not always correlate. Actually, in certain cases, in-plane and out-of-plane bonding gave different results (see chapter 5.2. and 5.4.), though in some cases they correlated /VI/. This agrees with Retulainen /Retulainen 1993/ who has reported that the results of different fibre-fibre bonding test methods depend on the testing mode (mode of loading).

The experimental results of this study can be rather well explained by beating-induced increase both in activation and in interfibre bonding. For example the small effect of beating on elastic breaking strain and fracture energy of mixture sheets is apparently due to the small effect on bonding. The effect on elastic modulus is bigger because of activation (Figures 4-4, 4-5 and 4-8).

It is recognized that the real phenomenon of the beating effect may be too complex to be explained by just two or three mechanisms, and that some effects are outside the bonding-activation-effective fibre length coordinates. On the other hand, Howard, Poole and Page /Howard 1994/ have concluded that there are three independent factors explaining most of laboratory beating effect: 1. Bonding 2. Fibre length and fines 3. Microcompressions. Based on this study, three factors seem suitable but factors 2. and 3. could be somewhat modified: 2. Fibre length 3. Activation. The effect of fines is then assumed to be caused mostly through bonding and possibly through wet bonding to activation. At low-intensity beating also fibre length could be excluded - this would leave just two independent factors: bonding and activation.

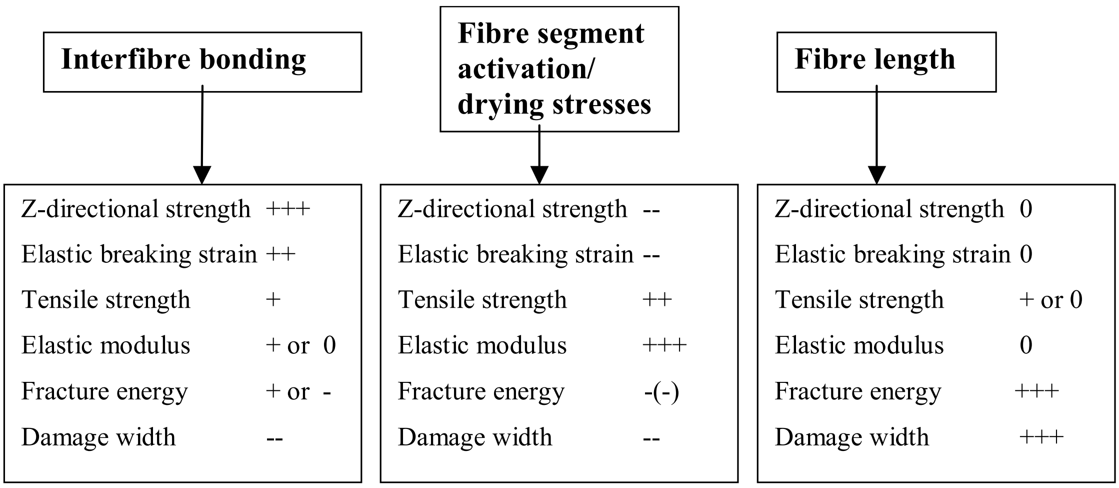

Figure 5-12. Simplified scheme of the effect of certain fibre level mechanisms on paper strength properties. + signifies positive correlation and - signifies negative correlation and 0 signifies no effect or a very small effect. A single + signifies a small effect and several +++ signify a strong effect.

5.6. Practical indications

Based on this laboratory study and related studies /Niskanen 2001a/ (this study was part of the KUMOUS-project), the practical implications of the results can be commented briefly. Some of these implications cannot be strictly scientifically based on the results. However, this could nevertheless give directions for future applications.

Z-directional properties of mechanical pulp based-paper seem to depend mostly on interfibre bonding and not on activation. This means that beating has only a limited effect on z-strength of mixture sheets. Dry strength additives like starch or possibly some novel application of CMC /Niskanen 2001a/ should have more potential.

Certain strength properties are connected to fibre morphology, e.g. in-plane fracture energy depends strongly on average fibre length. In mechanical pulp-based paper, neither bonding nor activation seems critical for the in-plane fracture energy, as long as there is reasonably good bonding (and activation). With mechanical pulp-based paper, there seems to be enough bonding so that the fracture energy does not improve by reinforcement pulp beating. However, increased average fibre length increases fracture energy (e.g. proportion of reinforcement pulp). Fibre strength and coarseness affect fracture energy a little but to a smaller extent than fibre length.

The in-plane tensile properties of mechanical pulp based-paper, like elastic modulus and tensile strength, seem to depend more on activation than on interfibre bonding. This implies that beating has a bigger effect on tensile properties than dry strength agents. Because mechanical pulps seem to have lower fibre segment activation than chemical pulps, kraft beating increases the elastic modulus of mixture sheets. The elastic modulus does not much depend on fibre length /Niskanen 2000/. However, elastic modulus depends a lot of drying conditions /Htun 1980, Zhang 2001/. Because of the importance of drying conditions, the relevance of standard laboratory sheets in the evaluation of the elastic modulus is questionable.

6. CONCLUSIONS

The aim of this work was to gain a better understanding of the effect of reinforcement pulp beating on the strength of mechanical pulp-dominated paper. The main purpose of reinforcement pulp beating is to improve the runnability of paper.

The first objective of this study was to maximize runnability-related strength properties by beating. It was assumed that the flaw-resisting ability of paper correlates with the runnability of the dry paper web. In-plane fracture properties were assumed to describe the flaw-resisting ability. However, the effect of refining on the rheology of unflawed paper was also analysed.

The second objective was to understand the mechanism how refining affects paper strength and structure. As a part of this analysis, the size of the fracture process zone was measured by the damage analysis method.

It was found that reinforcement pulp ranking and optimisation of beating depend on the criteria used. The selection of critical strength properties affects the ranking – sometimes different methods give opposite results (e.g. out-of-plane tear vs. in-plane tear). For example neutral sulphite pulp does well when ranking is based on the in-plane fracture properties. Also, whether the analysis is based on pure chemical pulp sheets or mixture sheets with mechanical pulp, clearly affects the conclusions.

It was found that beating does not increase the in-plane fracture energy of mechanical pulp-dominated paper. Fracture toughness and elastic breaking strain increase only a little. However, tensile strength and elastic modulus increase. Beating increases z-directional strength properties of mechanical pulp-dominated paper only slightly. It was concluded that beating does not significantly improve the flaw-resisting ability of mechanical pulp-dominated paper, while the strength of unflawed paper does increase.

It was found that the in-plane fracture properties of mixture sheets cannot be estimated based only on fracture properties of the pure components. The additivity behaviour at different beating levels was strongly non-linear. The fracture energy of pulp mixtures seems to correlate mainly with the average fibre length (and not with the beating level).

Beating increases fracture energy per fracture zone area (damage width) in the pure chemical pulp sheets but not in the mixture sheets. In other words, in the mixture series, the fracture energy per damage width remains constant, though the damaged area becomes smaller. Evidently, beating does not increase the energy needed to open interfibre bonds in mechanical pulp-dominated paper. However, at the same time, fibre breakage increases, causing a slight decrease in the fracture energy. Elastic breaking strain results support the view that beating does not significantly increase (in-plane) interfibre bonding of TMP-based paper.

The mechanism of beating was analysed both for pure chemical pulp and mixture sheets. It was found that beating affects paper structure and bonding-related strength properties differently compared to wet straining, starch, filler and TMP-fines removal. It seems clear that beating does not affect the strength of paper simply by increasing interfibre bonding. The results can be better explained by beating-induced increase in both interfibre bonding and fibre segment activation. In high-intensity beating also the decrease in fibre length should be taken into account. It was found necessary to divide bonding into in-plane and out-of-plane components. The results largely support the activation theory originally presented by Giertz. However, no direct measurement of activation was used in this study.

It is recognized that the beating effect may in reality be too complex to be explained by just two or three mechanisms. This means that some effects could be outside the bonding-activation-fibre length coordinates. However, bonding and activation were concluded to explain the results to a large extent.

Low-freeness mechanical pulp seems to have as high interfibre bonding as but lower activation than beaten reinforcement pulp. Therefore, reinforcement pulp beating improves bonding-related properties such as elastic breaking strain or in-plane fracture energy only marginally. On the other hand, properties strongly dependent on activation, such as elastic modulus and tensile strength, are increased by reinforcement pulp beating.

Some practical indications of this study were considered: Starch addition alone cannot secure in-plane strength, especially for mechanical pulp-dominated paper. However, in the out-of-plane direction, starch addition seems more effective than reinforcement pulp beating. In pulp mixtures, starch affects only the z-directional bonding component. A combination of moderate beating and starch addition is probably the best approach if z-strength properties are critical or if high levels of in-plane strength are required /VI/. Examining the effect of fibre morphology it was concluded that fibre length has a clearly stronger effect on in-plane fracture energy than fibre coarseness /VII/.

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