Quantitative Study of Paper Structure at the Fibre Level for Development of a Model for the Tensile Strength of Paper

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Submitted in fulfilment of the requirements for the Degree of Doctor of Philosophy

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July 2005

Declaration

I hereby declare that this thesis contains no material which has been accepted for the award of any other degree or diploma in any university, and to the best of my knowledge and belief contains no material previously published or written by another person, except where due reference is made.

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Date _____

Summary

A new combined technique of resin embedding and confocal laser scanning microscopy is developed for quantitative analysis of paper structure at the fibre level. Fibre dimensions, fibre orientation and fibre collapse have been measured simultaneously in paper by using the new technique and image analysis. This technique is validated by comparing the measured values of fibre wall areas by the new technique in paper with those measured by the routine confocal microscopy technique on freely dried fibres on glass slides. For the first time, a technique is available for quantitative analysis of paper structure at the fibre level.

This technique has been used to study the mechanisms of densification of the paper structure in wet pressing. New parameters are defined for the purpose of quantifying the changes in the transverse dimensions of fibres in paper in wet pressing. It was found that fibre twist, fibre collapse and gap closure are the major types of movement of fibres in paper in wet pressing. Fibre twist, fibre collapse and gap closure is the predominant mechanism in paper structure densification at low pressing pressures. Increasing pressure only increases the apparent density slightly and the density increase is mainly contributed by the additional twist and collapse of the fibres at the high pressing pressure.

Based on the above new technique, a new technique for measuring properties of fibrefibre contacts is developed. Properties of fibre-fibre contacts, including the free fibre segment length, number and nature of fibre-fibre contacts and out-of-plane angle of fibre segments are measured in paper. It is the first technique that can determine all of the parameters associated with the fibre-fibre contacts simultaneously in paper. For the first time, data of fibre-fibre contacts in 'real' paper is available for testing models of fibre-fibre contacts. This study shows that the out-of-plane deflection angles of the free fibre segments have no regular trend with the pressing intensity. However, the out-ofplane deflection distance has been reduced by wet pressing since the free fibre length has been reduced. The experimental data seems to fit a two-parameter Weibull probability density function, even if no theoretical basis is given for using the Weibull density function. This thesis presents a new model that relates the fibre cross-sectional dimensions and the apparent density of paper to the number of fibre-fibre contacts per unit length of fibre. It is the first model that considers the effects of fibre cross-sections on the fibre-fibre contacts and it is also the first time such a model has been fully verified with experimental data. The model is also converted into two expressions for *RBA*, which is verified with measured results by two different methods.

This thesis also presents a new analytical model for tensile strength of paper based on the assumption that the macroscopic fracture of paper is trigger by the failure of fibres lying in the direction of the applied load. The new model relates the tensile strength to the zero-span strength of the component fibres through a factor r. The value of r is the ratio of the peak load and the average load in the fibres. It is the first analytical model that attempts to predict the start point of paper failure under load. It is shown that the shear lag analysis does not seem to apply to the fracture of paper.

Model structures of a fibre of interest connecting the fibre network matrix were constructed by using the experimental data and the Weibull density function. Simulations of load distribution on the fibre of interest suggest that the value of r is a 'dynamic value', which is determined by the way that the fracture of paper is triggered. It is shown that bond breakage occurs before the sheet fracture, and it significantly affects the value of r. The simulation model is still at very preliminary stage. It can not predict the correct values of r. However, a very good correlation is shown between the tensile index calculated by the peak average load from the bond breakage model and the measured tensile index, although the predicted value is only 1/3 of the measured value. This indicates the promise of the bond breakage model. Further modifying the bond breakage model by including fibre fracture is expected to provide better prediction of the sheet fracture and therefore better calculation of r value for testing the simple fibre fracture model. This project has built up a solid base for further refining the bond breakage model in the future study.

Acknowledgements

I would like to thank my supervisor Dr. Warren Bachelor for his guidance, flexibility and great support which allowed me to make most of my time to finish this thesis and develop my career in the paper industry. Thanks also go to my co-supervisor Professor "Bob" Johnston. He has also been a great supervisor and one who is always willing to advise and to help.

Thank you to Richard Markowski for your assistance in many experiments including pulping, fibre fractionation and paper testing. Thank you also for your friendship.

I would like to thank Andrew Conn for his excellent assistance in writing those macros for image analysis. Thanks Loi Nguyen for providing wood chips and assistance in pulping. Thank you John Barnard and Gamini Ganegoda from the Chemical Engineering Department Workshop for helping build the heating rig for the Nitrogen Adsorption test. I would also like to thank Roy Harrip and Gamini for your assistance in solving computer associated problems.

Thank you to all APPI staff and students for your friendship. The good memories we shared will remain in my mind forever.

I want to thank my beautiful wife Ying and my son Eric for their unconditional love and patience during my long hours away and for the pure joy that they bring to me everyday.

This work is funded by the Australian Research Council (ARC). I gratefully appreciate the funding received from ARC. I also appreciate a small fund received from Visy Paper for part of this work.

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Jihong He, Warren Bachelor and Bob Johnston, *A new analytical model for fibre-fibre contacts in paper and expressions for RBA*, presented at the 2003 international paper physics conference, Canada, September.

Jihong He, Warren Bachelor and Bob Johnston, *Comparison of analytical models for the tensile properties of paper with experimental results*, presented at the 2004 international paper physics conference, Troudheim, Norway, June.

1 Introduction

Every paper grade used in daily life needs certain strength to satisfy not only the end uses but also the converting operations. Papermakers have been making great efforts to produce papers with greater strength to meet the increasing requirements of modern life. For example, the increase in the already very high speed of printers is one of the major driving forces.

A sheet of paper is a network made of millions of fibres bonded together through hydrogen bonds at the crossing points. The strength of the paper is determined by both the properties of the component fibres and the network. These properties include fibre length, fibre strength, fibre-fibre bond strength, bond area, dimensions of the fibre cross-section, fibre-fibre contacts and the sheet density. It is essential to understand the relationship between the strength of paper and the fundamental properties in order to be able to make stronger paper products. Fundamental studies for the tensile strength of paper always try to develop models to describe this relationship.

The difficulty for modelling tensile strength of paper is that paper has a complex structure and the component fibres have properties with certain statistical distributions. This is also evidenced by the enormous work attracted by this topic in the past 50 years. Today this topic is still of interest of researchers.

Two types of methods have been used for developing models for tensile strength of paper: the analytical method and simulation method. Correspondingly models developed by these two methods are called as analytical models or closed-form models and simulation models. Most of the models developed before 1980s are analytical models. In later studies, particularly in recent years, computer simulation has been increasingly employed for modelling the tensile strength of paper.

It has been well recognized that some parameters, including fibre length, fibre strength, fibre-fibre bonded area, bond strength, the dimensions of fibre cross-section and fibre orientation, are important for the tensile strength of paper. A general understanding of the relationship between the tensile strength of paper and these fundamental parameters

has been established based on previous studies. For example, it is well known that longer and stronger fibres usually make stronger sheets. However, the mechanism through which these fundamental parameters contribute to the tensile strength of paper is still inadequately understood. The mechanism, by which the fracture of paper is triggered, is still the subject of research.

The other aspect of modelling the tensile strength of paper is verification of the models developed. The work is only completed when the model has been fully verified by experimental data. A complete set of data is essential for the model verification. However, some of the parameters mentioned above are very difficult to measure. New techniques need to be developed for measuring parameters such as dimensions of fibre cross-section in paper, fibre-fibre contacts in paper and relative bonded area.

The complexity and importance of paper structure makes it always a challenging topic in paper physics. Although enormous work has been done in this area, most of our knowledge in paper structure still stays in qualitative level. No techniques are available for measuring some important quantities of paper structure such as the number of fibrefibre contacts and the dimensions of fibres in paper. Data of these quantities are critically important for understanding the mechanism by which fibres develop strength of paper. Such data is also desired for testing the previous models of the tensile strength of paper. Clearly, further study in modelling the tensile strength of paper requires quantitative study of paper structure at the fibre level.

The objective of this thesis is to quantitatively study the paper structure at the fibre level to provide valuable data and knowledge for development of a new model for the tensile strength of paper. The project starts with the development of a new technique to quantitatively analyse the paper structure at the fibre level. By using this technique, fibre cross-sectional dimensions, fibre collapse and fibre orientation will be measured directly in paper. This technique will also be used to study the mechanisms of densification of the paper structure in wet pressing. Based on this technique, a technique will be developed to measure the properties associated with fibre-fibre contacts, including the nature of the contacts, number of fibre-fibre contacts, free fibre length and out-of-plane angle. A new model for the number of fibre-fibre contacts will be presented and fully verified with experimental data. Model structures of a fibre of

interest connecting to the fibre network will be constructed based on the experimental data and the load distribution along the length of a fibre will be simulated for no bond breakage, as well as with bond breakage. All of the experimental data and data from simulations will be used for testing the final model for the tensile strength of paper.

Chapter 2 reviews the literature in both studies in paper structure and modelling the tensile strength of paper. Chapter 3 presents a new simple fibre fracture model for the tensile strength of paper and a new model for the number of fibre-fibre contacts. The new techniques for quantitative study of the paper structure are discussed in Chapter 4. Chapter 5 illustrates the important experimental methods used in this thesis. Chapter 6 discusses the mechanisms of densification of the paper structure in wet pressing. In Chapter 7, the new model for number of fibre-fibre contacts is verified with experimental data and the distribution of the free fibre length is also discussed. Tests of the simple fibre fracture model for the tensile strength of paper is discussed in Chapter 8. Firstly, the Page equation is tested with experimental data obtained in this thesis, and then the application of the shear lag analysis for calculation of load distribution along the length of a fibre is discussed. Finally, a new direct load transfer theory is presented and simulations of load distribution along the fibre based on the new load transfer theory are discussed. Chapter 9 summarises the conclusions drawn from the work in this thesis and gives the recommendations for future work.

2 Literature review

2.1 Introduction

Paper is a complex network of fibres that are bonded together through hydrogen bonds at points where fibres cross each other. Every paper grade needs certain strength for both converting operation and end use. Tensile strength is the most commonly used parameter for describing the mechanical properties of a sheet of paper. In other words, the tensile strength of paper is the most important strength property of paper. The tensile strength of a sheet is determined by the properties of the fibre network and the properties of the fibres comprising the network. A better understanding of the relationship between the tensile strength of paper and the fundamental properties of its component fibres and paper structure is important for both paper research and production.

After years of theoretical and experimental research, it has been found that the important parameters for tensile strength of paper include fibre length, fibre strength, bond strength, relative bonded area (RBA), the number of fibre-fibre contacts, dimensions of fibre cross-section and apparent density.

Numerous researchers have attempted to develop models, either using analytical or simulation methods, to relate the tensile strength of paper to the properties of fibres and the structure of paper. Models of this relationship are critical, because they allow for the prediction of the behaviour of paper with structures outside the range of previous experimental data.

Modelling tensile strength always has to answer two questions. The first question is what is the mechanism by which load is transferred from one fibre to its adjacent fibres? This controls the load distribution along the axis of the fibre. The second question is what is the trigger of the fracture of paper (macroscopic fracture)? The trigger could be the failure of fibre-fibre bonds or the failure of fibre segments in the sheet. There are still controversies over both these questions.

Another important aspect for the modelling tensile strength of paper is model verification. However, some of the parameters are very difficult to measure. This makes the studies incomplete and to some extent hinders the advance of these studies.

In this chapter, the important properties of fibre and paper structure for tensile strength of paper are briefly outlined. This chapter reviews previous theories for stress transfer between fibres and the initiation of the fracture of paper. The important parameters in stress transfer are also discussed. This chapter also reviews the important analytical models for tensile strength developed in previous studies. These models are compared with each other and the assumptions they make are discussed. Verification of these models by experimental work is also discussed. Measurement of those parameters that are difficult to measure is discussed extensively.

2.2 Fibres and fibre network

In this section the important parameters of the fibres and the fibre network are discussed.

2.2.1 Fibres

Fibres are the basic component for paper. A sheet of paper consists of millions of fibres. These fibres should have a certain length and strength in order to form a fibre network with certain strength. A fibre also has transverse dimensions including fibre wall thickness, fibre width, wall area and lumen area.

The importance of transverse dimensions of wood pulp fibres has been long recognized. In early studies, fibre coarseness, defined as the mass of per unit fibre length, was commonly used for characterization of fibre cross-section. The effects of fibre coarseness of softwood pulp fibres on paper physical properties have been investigated intensively by Seth (Seth 1990) and Paavilainen (Paavilainen 1993). Both authors emphasized the importance of the effect that fibre coarseness has on paper physical properties. For a given sheet grammage, finer fibres being more numerous, make more fibre-fibre contacts per unit area, and therefore make stronger sheets. Paavilainen (Paavilainen 1993) showed that the effect of fibre coarseness on the tensile strength of paper is more significant than that of fibre length. She believed that, to obtain high paper strength, the abilities of the pulp fibres to form strong bonds and high intrinsic fibre strength are more important than the number of load-bearing fibres.

A later study found that fibre coarseness alone is insufficient to define and predict pulp quality, as two fibres of similar coarseness can have quite different wall thicknesses if their perimeters are different (Seth 1997). The dimensions of fibre cross-sections should be characterised directly.

The shape of the fibre is also critical because it is not constant. Never-dried fibres tend to collapse under wet pressing. The more collapsed the fibres are, the better the bonds (larger bonding area) they will form. Fibres will flatten out as they collapse to be wider and therefore have greater potential bonding areas.

The transverse dimensions of fibres affect the degree of fibre collapse therefore the conformability of fibres. Several studies have been conducted to quantify the degree of fibre collapse (Kibblewhite 1991; Jang 1995; Jang 1998). Jang (Jang 1998) found a geometrical factor, $LP/2\pi t$, that controls the degree of fibre collapse, where LP is the total lumen perimeter (uncollapsed and collapsed) and t is fibre wall thickness. Jang (Jang 2001) has also presented a new theory on fibre collapse. This will be discussed in detail in Chapter 6.

2.2.2 Fibre network

We normally think of paper as a heterogeneous network of fibres bonded together by hydrogen bonds. The heterogeneity and the fact that there are always distributions for the fibre properties make the paper structure difficult to characterize. Scientists attempt to do this, however, in terms of network parameters. Network parameters attempt to define the structure in terms of the dimensions, physical properties, geometrical orientations of the fibres and the nature and extent of the bonding between fibres. The often used network parameters can be roughly divided into macroscopic and microscopic parameters. The former includes sheet density and formation, while the latter includes bond nature, bond area, bond strength, number of fibre-fibre bonds per unit length of fibre, relative bonded area (*RBA*) and fibre orientation. The features of these parameters are briefly discussed as follows.

2.2.2.1 Sheet density

The sheet density is simply defined as the mass per unit volume, or basis weight divided by calliper. The sheet density is the simplest measure of paper structure. There is generally a strong linear relationship between sheet density and the bonding degree of the network (El-Hosseiny 1979).

Wet pressing significantly affects sheet density. An understanding of the mechanisms of densification of paper in wet pressing is an important topic of paper structure analysis. Szikla believe that increasing fibre bonding is the dominant mechanism in the densifying effect of wet pressing (Szikla 1989). Gorres et al (Gorres 1993) conclude that there are two mechanisms controlling the relationship between the density of paper and the wet pressing pressure. Fibre collapse and deflection of free fibre segments are the predominant mechanisms at low pressing pressure, while at high pressure all spaces within the sheet which can be filled by such deflections have been filled, therefore completion of fibre collapse and perhaps other effects then predominate. They claimed that a second effect is needed to explain the continuing increase in density at high pressure where fibres are totally collapsed. The understanding of the mechanisms of the densification of paper structure in wet pressing is still incomplete. Sophisticated techniques for analysis of paper structure at the fibre level are essential for a better understanding of the densification of paper structure.

It was first noted many years ago that tensile strength is often a linear function of the apparent density (Clark 1943). A linear relationship is also very often seen in later studies, no matter whether the density of paper was changed by refining or by wet pressing (Luner 1961; Seth 1990; Kibblewhite 1993). No generally accepted explanation is known for this.

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Even though the apparent density of paper is not a well-defined quantity, it would be useful for practical applications to understand how density affects paper strength (Niskanen 1993). Thus, a model for tensile strength that includes apparent density is called for.

Niskanen (Niskanen 1993) proposed that the tensile index of paper is a linear function of sheet density at reasonably high densities. De Ruvo and his co-workers (Ruvo 1986) proposed that the tensile index depends on how efficiently the structure of sheet utilises the component fibres. The efficiency normally increases with beating of the fibres and with increasing intensity of wet pressing of the wet web. An increase in density can improve stress distribution in the sheet allowing the fibres to participate more homogeneously in the bearing of load. However, no analytical expression for this has been determined and so the concept has only qualitative predictive capability.

2.2.2.2 Bond nature and bond area

Page et al, (Page 1962) in a piece of classic research, studied fibre-fibre bonds by direct observation using polarized illumination. They found that fibre-fibre bonds in paper could be divided into two main types in terms of bond nature, i.e. simple bonds and obstructed bonds, as schematically shown in Figure 2-1.



Figure 2-1 Models illustrating, on the left, simple and, on the right, obstructed fibre-fibre bonds (Page 1962).

Fibre-fibre bonds form gradually as the solids content of paper increases. Page et al believed that there are two possible mechanisms by which fibre-fibre bonds may form

(Page 1962). Collapse of one fibre on to another may occur because of the large forces of wet pressing and surface tension. Alternatively, the contact areas may be developed by the accumulation of dissolved and suspended material in the residual water in the regions of a crossing. Campbell's (Campbell 1959) description for bond formation is that surface tension forces, such as colloidal interactions and mechanical interlocking of fibrils, pull fibres closer together when water is removed from the wet web. This is the Campbell effect. However, the solids content at which actual inter-fibre bonds form is not exactly known, since the Campbell effect changes gradually into hydrogen bonds. Lyne and Gallay (Lyne 1954) suggested that hydrogen bonds begin to form in the range of 10 to 25% solids content.

Fibres shrink during drying. The amount of shrinkage depends on the swelling degree of the wet fibre wall (Laivins 1993). The internal fibrillation and chemical composition of the fibre wall affect swelling. Lignin reduces and hemicellulose increases swelling.

Fibres shrink primarily in the lateral direction (Weise and Paulapuro 1996). The competition between the lateral shrinkage tendency and longitudinal stiffness of fibres creates shear stresses in the bond area, especially at its periphery, as shown in Figure 2-2 (Van Den Akker 1962). The shear stresses at inter-fibre bonds generate axial compressive forces on the crossing fibres and may even cause deformations in bonded fibre segments. These deformations are sometimes called microcompressions (Page 1962). They modify the mechanical properties of the bonded segments in comparison with the freely dried fibres. The mechanical properties of the fibres in a paper sheet are therefore related to the presence of inter-fibre bonds.



Figure 2-2 Schematic of the stress of a shrinking bond area: a free bond (a) and the same configuration under external load (b) (Van Den Akker 1962).

No simple definition for the bond area is available because the overlap area of two fibres is not necessarily completely bonded. It is believed that fibres in optical contact may not necessarily form bonds because the distance for optical contact is two or more orders of magnitude greater than the distance necessary for hydrogen bonding.

Nanko and Ohsawa (Nanko 1989) did an excellent study on the structure of fibre bonds of a bleached hardwood kraft pulp by using a transmission electron microscope. They identified four major features in the bond: the bonding layer, wrinkles, skirt and covering layer, as illustrated in Figure 2-3 (Nanko 1989).



Figure 2-3 Structural features of fibre bonds according to Nanko and Ohsawa (Nanko 1989).

The major variables that may affect the bonding size (or the bonding area) include furnish, beating, pressing and drying conditions. The fibre width is a controlling factor for the bond size. Furthermore, the surface topography and plasticity of fibres in different furnishes will have a major effect. Beating significantly increases the bond size of both simple and obstructed bonds. Wet pressing has similar effects to beating. Dry tension, has only small effect on the bond size and, if anything, tends to reduce it.

2.2.2.3 Fibre-fibre bond strength

The strength of a fibre-fibre bond usually refers to its shear strength. This is the maximum load that the bond can carry when the bonded fibres are displaced relative to one another parallel to the bonding area (Niskanen 1998). The bond strength is fibre dependent. A summerwood fibre bond has higher bond strength than a springwood

fibre bond although the former has smaller bond area. A typical springwood fibre bond can bear less than half a gram before fracture (Stratton 1990).

Another often used parameter is specific bond strength, which is defined as the strength per unit bonding area. However, Button (Button 1979) concluded that the specific bond strength could not characterize the bonding capacity of the fibres. He showed with a bond model that the stress distribution within the bond is nonuniform when the bond is loaded. He also showed experimentally that the bond strength is almost independent of bond length or area but increases as the square root of the thickness of the bonded fibres. This result can explain why the summerwood bonds are stronger than the springwood bonds.

2.2.2.4 Fibre-fibre contacts

The fibre-fibre contact is the most important fundamental parameter for a paper structure. The properties related to fibre-fibre contacts include number of fibre-fibre contacts per unit length of fibre, the nature of the contacts, the free fibre length and its distribution and the out-of-plane deflection angles of fibre segments in the paper. These variables as a whole determines the arrangement of fibres in a network, therefore they determine the mechanical behaviour of the fibre network under external stress.

Fibres are brought together to form fibre-fibre contacts during the densification of a sheet in wet pressing. For a random sheet, the number of fibre-fibre contacts is determined by the sheet density and the cross-sectional shape of fibres in the sheet. One has to know both to determine the number of fibre-fibre contacts in the sheet. There are still no techniques available for quantifying the shape of fibres in a sheet. Therefore it is crucial to develop new techniques to measure the cross-sectional dimensions of fibres in sheets in order to further study the number of fibre-fibre contacts in sheet.

Free fibre length is a critical parameter for paper structure. Different parameters have been used to represent a free fibre length. The free fibre length has been defined as the distance between the centres of fibre crossings (Kallmes and Corte 1960); the intercrossing distance represented by the distance between the centres of bonded crossings (Kallmes 1963); and the projected interbond distance, which is the distance between the projection of the bonds on the fibre axis (Page 1962). Kallmes and Bernier (Kallmes 1963) have shown that these three definitions of a free fibre length will have different absolute values but will produce similar distributions of free fibre length.

The distribution of free fibre length is also critical for modelling the structure of paper. Kallmes and Bernier (Kallmes and Corte 1960; Kallmes 1963) have shown that a 2-dimensional sheet has a negative exponential distribution of free fibre length. However, the measurements of Page *et al.* (Page 1962) clearly cannot be fitted to a negative exponential distribution. Dent (Dent 2001) recently showed that the negative exponential distribution apples only to perfect random of sheet made of infinitely long fibres. He proposed that a random sheet of fibres of finite length has a general gamma distribution of the free fibre length. This model is based on a 2-dimensional model and has not been verified by experimental data.

The arrangement of fibres in the z-direction can be layered or felted. Differences in the mechanical properties in the z-direction of the sheets are attributed to the differences in interwoven structure or z-directional fibre arrangement in the sheets. A felted sheet structure should give better out-of-plane or z-directional strength than a layered structure because fibres are stronger than bonds (Niskanen 1998). The out-of-plane-angle is then an important quantity for describing the fibre arrangement in the z-direction. A felted sheet will have a higher out-of-plane-angle than a layered structure. However, there are still no effective techniques available for measuring the out-of-plane-angle in a sheet. New microscopy and image analytical techniques can provide new information.

2.2.2.5 Relative bonded area

The relative bonded area (*RBA*) is the fraction of the total available fibre surface that is bonded and is a quantity that has found widespread application in theories of paper mechanical properties. The standard definition of relative bonded area is $RBA = (A_t - A)/A_t$ where A_t is the total area available for bonding and A is the unbonded area in the sheet after is has been formed. The *RBA* is a derived property. It is determined by a combination of the number of fibre-fibre bonds, bond area and area of fibre surfaces.

It is believed, in general, that refining and wet pressing can increase the *RBA* of a sheet therefore increase the tensile strength of the sheet. According to Page's theory (Page 1969), the relationship between *RBA* and tensile strength of paper can be illustrated as Figure 2-4. Such a relationship has also been shown by experimental work by many other researchers (Ingmanson and Thode 1959; El-Hosseiny and Abson 1983; Retulainen and Ebeling 1993).



Figure 2-4 The relationship between tensile strength and *RBA* according to the Page equation (Niskanen 1998).

2.2.2.6 Formation

Formation is the variability of the basis weight of paper. Good formation is important for the mechanical properties of the final sheet (Lyne 1954). We expect that uniform paper bear higher stresses than non-uniform paper. Variations in local basis weight have been explained by the tendency of the fibres in a suspension to flocculate forming relatively dense assemblies known as flocs (Radvan 1980). Longer fibres have a greater tendency to floc due to the greater tendency for mechanical entanglement (Roberts 1996). Curled fibres entangle more easily to flocs and settle more rapidly out of suspension, both of which affect the formation of the sheet (Page 1985). Niskanen (Niskanen 1993) has emphasised, in his review paper, the importance of the formation for tensile strength. However, analytical methods can do little to consider the effects of the formation on the tensile strength of paper. It is believed that computer simulations, in combination with new measurements and effective data analysis will lead fruitful results in this aspect. This project will not consider the effects of the formation on the tensile strength.

2.3 Mechanisms of stress transfer and stress distribution in fibre

As discussed above, paper is usually seen as a network of fibres with finite length. When the paper is loaded, the load has to be transferred somehow from one fibre to its neighbouring fibres. Whatever assumptions are made in network theories it is still necessary to consider how the loads may be transferred from one fibre to another. The mechanism by which load is transferred determines to a great extent the mechanical properties of the paper, and the load transfer mechanism itself is determined by the fundamental properties of fibre and paper structure.

The major variables that determine the mechanism of stress transfer include fibre length, bond stiffness and sheet density. Previous theories attempted to relate the mechanism of stress transfer to these fundamental parameters. These theories are reviewed in this subsection.

Cox (Cox 1952) contributed the first model for stress transfer. The Cox model is established based on an ideal structure in which each fibre is considered as embedded in a continuous solid medium of resin. The resin matrix as a whole is strained homogeneously, but locally this state of uniform stress and strain is disturbed by the transfer of load to the fibres. When the resin matrix is strained the stress is transferred to fibres via the shear stresses in the interface region between the fibre and matrix. The Cox model is often called the shear lag model and this model was the theoretical basis for later studies (Page, Seth et al. 1979; Aström, Saarinen et al. 1994; Räisänen 1996; Räisänen 1997). Ochiai and Hojo (Ochiai 1994), and more recently Johnston (Johnston 1995) reported modified shear lag models that account for the whole stress-strain curve for a fibre reinforced composite.

When the shear lag model is applied to a fibre network, it predicts that stress is transferred from each fibre to its adjacent fibres in the regions of the fibre ends. If we consider sheets of different degrees of bonding, it is expected that the higher the degree of bonding, the shorter the length over which transfer of stress would occur (Figure 2-5). The stress in a fibre is at its maximum at the centre and diminishes to zero at the ends. For constant grammage and fibre coarseness, reduction in fibre length increases the number of fibre ends in the sheet, and decreases the number of fibre-fibre contacts per fibre. Since stress is only transferred across a fibre end, the distribution of stress in the network is more uneven if there are more fibre ends. Thus, an increase in fibre length improves the stress distribution in the network, therefore enhances the tensile strength.



Figure 2-5 Load distribution in a fibre predicted by the Cox's shear lag model (Page, Seth et al. 1979).

The finite element method (FEM) has been successfully used for analysis of stress transfer in fibre network. Rigdahl et al. (Rigdahl 1984) simulated a simple fibre network (a completely square lattice) using FEM. They analysed the effects of the fibre ends, the sheet density and the bond stiffness on the stress transfer. They found that the axial stress is low close to the fibre ends. It increases rather sharply with increasing distance from the ends and tends to reach a plateau level. If, however, a neighbouring fibre has an end beside the fibre under consideration, this will give rise to a relatively

sharp peak in the stress distribution, as shown in Figure 2-6. Rigdahl et al stated that these stress peaks are of great significance for the mechanical properties of paper structures, especially for the **onset** of rupture. The cause of the appearance of the stress peaks is that the load carried by a fibre close to its end must be redistributed to the next fibres. They concluded that the fibre ends are very pronounced in stress transfer and therefore the strength of the paper.



Figure 2-6 Load distribution according to Rigdahl's simulation (Rigdahl 1984).

In the same study, Rigdahl et al also found that a decrease in sheet density reduces the efficiency of stress transfer. The reduction is mainly due to a decrease, with decreasing density, of the number of crossing fibres that transfer the stress between fibres in the network structure. Another important finding by Rigdahl et al is that the bond stiffness does not significantly affect the stress transfer between the fibres unless it is rather low.

The findings of Rigdahl et al (Rigdahl 1984) emphasise the importance of the fibre length (longer fibres have less fibre ends in the network) and the number of fibre-fibre contacts for stress transfer in a paper structure. These findings are significant for the understanding of the mechanisms of stress transfer in paper structure, although the model used is rather simple.

In a later computer simulation study, Aström et al (Aström, Saarinen et al. 1994) analysed the stress transfer mechanisms in fibres constructed within the effective medium approximation first developed by Cox. Their computer simulations showed that the shear lag model holds for random fibre networks at high densities.

Raisanen et al. (Räisänen 1997), in a more recent study, extended the shear lag model to flexible bonds in simulations of random fibre networks. They analysed the stress transfer mechanisms by using a force balance approximation. In the force balance approximation, fibre stress is allowed to be transferred from one longitudinal fibre directly to the next one. In other words, the stress of a given fibre may at any bond be 'taken up' by the crossing fibre, therefore there should be stress fluctuations along the fibres. They concluded that the shear lag model does not apply to random fibre networks.

The shear lag type models give an expression for the stress distribution along a fibre in a sheet as follows (Aström, Saarinen et al. 1994):

$$\sigma_{f}(x,k) = E\varepsilon_{x} \left(1 - \frac{\cosh\left[k\left(\frac{1}{2} - \left(x/l_{f}\right)\right)\right]}{\cosh(k/2)} \right)$$
2.1

where *E* is the elastic modulus of the fibre and ε_x is strain of the sheet in the *x* direction (the direction of the longitudinal axis of the fibre), *x* is the distance from one fibre end, $x \in (0, l_f)$, and *k* is a factor determined by the stress transfer mechanism.

The average load in the fibre is obtained by integrating Equation 2.1 and dividing by l_f . Therefore:

$$\overline{\sigma_f} = E\varepsilon_x \left(1 - \frac{2 \tanh(kl_f / 2)}{kl_f} \right)$$
 2.2

The value of k has been determined by different methods. It was determined analytically by Cox (Cox 1952) and by Page et al (Page and Seth 1980). It has also been determined by computer simulations in more recently studies by Aström et al (Aström, Saarinen et al. 1994) and by Raisanen et al (Räisänen 1997). Determination of the value of k will be discussed in detail in Chapter 9.

2.4 Initiation of the fracture of paper

Fracture here means the macroscopic fracture of paper in a tensile strength test. The tensile strength is the maximum load that occurs at the moment macroscopic rupture begins. Once the rupture has been initiated, it proceeds rapidly during the tensile testing. Therefore the mechanism by which the fracture of paper is initiated is of critical importance for the tensile strength of paper. One has to determine what initiates the fracture of paper before attempting to predict the tensile strength. The fracture of paper can be triggered either by fracture of fibre segments or by fracture of bonds in the paper. Correspondingly, two viewpoints exist - the fibre-fracture triggered viewpoint and the bond-fracture triggered viewpoint.

Page (Page 1969) and Kallmes et al (Kallmes 1977) believed that the fracture of paper is initiated by the failure of the fibres oriented in the direction of the applied load. Cox (Cox 1952) has shown theoretically that the tensile strength of a random twodimensional sheet does not exceed one third of the strength of its component fibres. On the other hand, other researchers (Van Den Akker 1958) (Niskanen 1993) believed that fibre bonds control the tensile strength of paper. They assumed that paper fails when the external stress equals the bond yielding threshold.

While Page (Page 1969) has provided some evidence supporting the fibre-fracture triggered viewpoint, Van Den Akker (Van Den Akker 1958) has shown that many bonds fail before macroscopic failure has commenced, and many fibres rupture only after the macroscopic failure has commenced. However, none of this evidence is strong enough to support one viewpoint and against the other.

The best way to answer the question is direct observation of the fracture process of a sheet. Working with very thin, two-dimensional sheets, Corte el al (Corte 1961) were able to detect the failure of individual bonds when a sheet was strained. However, such observation is only limited to very thin sheets. Page et al (Page 1962) observed individual fibre bonds of normal sheets under a light microscope. They found that in contrast to the thin sheets studied by Corte et al, the bonds ruptured only partially when paper was strained to failure. The partial rupture of the bonds can be explained not only

by the irregular shape of the bonded area (Page 1962) but also by the stress distribution induced by drying (Van Den Akker 1962).

Giertz and Roedland (Giertz 1979) measured the local strains as a function of the external elongation of a paper sheet. They also observed that the bonds did not rupture completely. The bonds that yielded at small strain often did not deform more at higher strain. When the external load was removed, permanent elongation was found to have taken place primarily in the bonded areas of fibres. In summary, the bonded fibre segments seemed capable of governing the load-elongation behaviour of paper. In addition to this, many researchers have shown that typical single fibres can elongate plastically, much like paper (Dumbleton 1972; Seth 1983). Seth and Page (Seth 1983) showed experimentally that the plasticity of paper is mainly caused by the plasticity of fibres and the effect of bond breakage is usually small. Page recently also showed that most of the work done in plastic deformation is due to the deformation of the fibres (Page 2002). The computer simulations of Raisanen et al. (Räisänen 1996) support Seth and Page's viewpoint.

In summary, there is still no sufficient evidence showing that the macroscopic fracture of paper is triggered by rupture of fibre segments or bonds in the paper. However, it is clear that, before macroscopic fracture, when paper is stretched, fibre segments become permanently elongated and fibre-fibre bonds rupture, both in small steps. The partial bond ruptures and the related yielding of fibre segments relax the local stresses, and this will increase the stresses in neighbouring elements. One can adopt the viewpoint that the plasticity of paper is mainly caused by the plasticity of the bonded fibre segments in the paper. The partial rupture of bonds will relax the stresses in those bonds and this can increase the stresses in nearby fibre segments. The stresses in fibre segments increase as the external stress increases and the number of partially ruptured bonds increase. When the stresses in the fibre exceed its strength, the fibre breaks and triggers the macroscopic fracture of the paper. Once the rupture has been initiated, it proceeds rapidly in the fibre network. Fibres break or pull-out depends on the bonding degree of the fibres.

2.5 Important models for tensile strength

Modelling tensile strength (or elastic properties) of paper has long been an interesting topic in paper physics. Over the past 50 years, a large number of attempts have been made using either analytical methods or computer simulations to develop models for tensile strength. In early years, models developed by analytical methods were confined to a simple and uniform network. However, they produced results of great interest and value. These models are referred to as analytical models (or closed-form models) and usually given by explicit expressions. During 1970s and 1980s, the arrival of computers with considerable computer simulations. The models developed by computer simulations are referred to as simulation models. The rapid developed by computer science in 1990s further enhanced the studies in paper network simulations, and resulted in more powerful simulation models, taking into account more and more variables, which were believed to be important

Numerous models for tensile strength have been developed in previous studies. Only the most often cited models are reviewed and discussed here.

2.5.1 Analytical models

The analytical models reviewed here can be roughly divided into two groups according to the assumptions they made. One group of models, including the Cox model, Kallmes-Bernier-Perez model, Page equation and Allan-Neogi model, are based on the assumption that the tensile strength of paper is dominated by the strength of the component fibres. We call this type of models fibre strength dominated models. The other group of models assume that a structure of paper can be treated as a fibre reinforced composite. We call this type of model a pull-out model. Examples of this type of model are the Kane and Shallhorn and Karnis models.

2.5.1.1 Cox Model

Cox (Cox 1952) contributed the first model for the elastic modulus of paper. The Cox model considers paper as a perfect homogenous plane consisting of long straight thin fibres oriented either at random or according to some statistical distribution. Each fibre is assumed to extend throughout the whole body of the material and to be loaded only at the ends. This ideal paper allows for no stress transfer between fibres and the strain in every fibre is equal to the local strain of the sheet in the fibre direction. This implies that the relation between the strain in a fibre and the strain in the paper is purely geometrical. From an analysis of strains and stresses in fibres, Cox related the stiffness of paper to the stiffness of fibres through a distribution function of fibre orientation. The distribution function of fibre orientation, f, used by Cox in his analysis is as follows:

$$\pi f(\theta) = 1 + a_1 \cos 2\theta + a_2 \cos 4\theta + b_2 \sin 4\theta$$
 2.3

where θ is the angle of fibre with respect to the horizontal strain, and a_1 , a_2 and b_2 are coefficients.

Cox found, for a general situation, the stiffness of paper was affected by fibre orientation in a rather complex manner. If the paper is random, the coefficients, a_1 , a_2 and b_2 , all become zero, and the relationship between the elastic modulus of paper and the elastic modulus of the fibres is given by the following equation:

$$E_p = 1/3E_f$$
 2.4

where E_p is the elastic modulus of the paper and E_f is the elastic modulus of fibre, where sheet and fibre moduli are normalised for grammage and coarseness respectively.

This model does not give an explicit expression for the tensile strength of sheet, however for a random sheet, assuming failure occurs when the paper failure strain reaches the fibre failure strain, the tensile strength can be predicted as:

$$T = E_p \varepsilon_f = 1/3E_f = 1/3\sigma_f$$
 2.5

Where T is the breaking length of the sheet (km), ε_f is the failure strain of fibre and σ_f is the breaking length of the fibre (km).

Cox verified this model with resin-bonded board in his study. Page and Seth (Page, Seth et al. 1979; Page and Seth 1980; Page and Seth 1980) tested the Cox model with paper and found the Cox model predicts modulus correctly only in the case of strongly bonded handsheets of long straight fibres.

2.5.1.2 Kallmes-Bernier-Perez Model

The Kallmes-Bernier-Perez model (referred to as the KBP model) was first developed by Kallmes and Perez (Kallmes and Perez 1965), and was later extended by Kallmes, Bernier and Perez (Kallmes 1977; Kallmes 1978). Kallmes et al. attempted to describe the whole load-elongation curve of paper. Kallmes et al. made some assumptions that are the same as those made in the Cox model. They assumed that the component fibres of a paper are purely elastic and the axial elastic modulus of each fibre is much greater than its lateral shear modulus. The strain of the sheet is assumed to be uniform. It is also assumed there are no variations of strain along the length of fibre, and that the strain in a fibre is equal to the local strain of the sheet in the fibre direction. The stress and strain in a fibre are related to the stress and strain in the sheet through geometrical analysis. To include fibre orientation, Kallmes et al used a distribution function of $1 + a_1 \cos 2\theta$ that is the first two terms of the four-term expansion function used in the Cox model (Cox 1952). However, the a_1 coefficient in the function was not determined. When Kallmes et al used the model in practice they set the a_1 coefficient to be zero by assuming the paper was random in fibre orientation.

Kallmes et al. suggested that fibre segments (the distance between two bonds) in paper can be divided into two types: 'inactive' or 'passive' type and 'active' type. The passive segments take no loads when the sheet is strained, and the active segments are those that take loads and are also referred to as load-bearing elements. Before the paper is strained, passive segments of fibres already exist, which were induced by the defects in fibres, such as curl and kinks, and they will not contribute to the tensile strength of paper. Kallmes et al proposed that during the process of paper straining, bond failure occurs, which causes the plasticity of paper. The difference between the load actually developed by a sheet under strain and that which would have been developed elastically is due to the complete removal of the passive fibre segments as load-bearing elements. In other words, a fraction of passive fibre segments is generated due to bond failure prior to sheet failure. This actually implies that the load in a sheet is taken by progressively fewer fibres during the process of straining, which is also one of the Page's assumptions (Page 1969). In the case of well-bonded sheet, Kallmes et al. assumed that the failure of paper is initiated by the failure of fibres oriented in the direction of maximum sheet strain. In the case of weakly bonded sheet, Kallmes et al. assumed that the failure of sheet is caused by failure of bonds at lower strain.

The fraction of passive fibre segments due to bond failure is given by:

$$f_b = c \frac{driving \ force}{resistance}$$
 2.6

The driving force setting up shear stresses in contact areas is the axial load developed in the fibres. The force that is resisting bond failure is equal to the shear strength of the contact areas. The constant c in Equation 2.6 was not determined by Kallmes et al.

Based on the above considerations, a well-bonded sheet ruptures when the sheet strain is equal to the fibre failure strain. Then the tensile strength of a sheet whose rupture is initiated by fibre failure is given by:

$$T_{f} = \frac{W}{w} t (1 - f_{f}) \left[\frac{1}{3} - \frac{\pi e}{4} - \frac{ct}{36 s \lambda \omega RBA_{f}} (5 - 4\pi e) \right]$$
 2.7

where T_f is the tensile strength of a strongly bonded sheet (kN/m), W is the basis weight of the sheet (g/m^2) , w is the coarseness of fibre (mg/m), t is the strength of fibre (kN), f_f is the proportion of passive or macro-curled fibres at sheet failure, s is the shear strength of the inter-fibre bonds (N/m^2) , λ is the length of a fibre (m), ω is the width of the fibre (m), RBA_f is the degree of bonding of sheets at its final failure and e is the eccentricity of the fibre network, which defines the degree of bias of the fibre-orientation distribution and c is the constant from Equation 2.6. For a weakly bonded sheet, the point of final rupture is defined as the maximum point on the load-elongation curve and the tensile strength is given by:

$$T_{b} = \frac{1}{16} \frac{W}{w} \frac{s\lambda \omega RBA_{f}}{c} (1 - f_{f}) \frac{(4 - 3\pi e)^{2}}{(5 - 4\pi e)}$$
2.8

Kallmes et al. believe that the failure of paper can be initiated by either fibre failure or bond failure, however, the mechanism by which the paper ruptures is determined by the degree of bonding (RBA_f) at the moment of the incipient sheet failure. A concept of 'critical degree of bonding' (RBA_{cr}) is defined explicitly by setting the strain of the sheet at the maximum point of the load-elongation curve equal to the fibre failure strain. When RBA_f is greater than RBA_{cr} sheet failure is initiated by fibre failure, otherwise, bond failure initiates sheet failure.

There are a number of criticisms that can be made of the KBP model. The basic idea of the Kallmes et al. is that the plasticity of paper is caused entirely by the failure of interfibre bonds. Seth and Page (Seth 1983), whose experiments show that the plasticity of paper is mainly caused by the plasticity of the fibres themselves and that the effect of breakage of bonds is usually small, argue against this viewpoint. Seth and Page's viewpoint is also supported by the recent simulation study of Räisänen et al (Räisänen 1996). Another criticism is that straining the sheet actually straightens fibres, reducing the number of passive segments. A final criticism is – how do you measure the proportion of passive fibres?

Kärenlampi (Kärenlampi 1995; Kärenlampi 1995) extended the KBP model by accounting for the distributions of fibre properties. The three nested integrals in the Kärenlampi model, however, are quite difficult to solve. This was overcome by Feldman et al. (Feldman, Jayaraman et al. 1996) by generating random fibres crossing the failure line of the paper and numerically evaluating the stresses.

2.5.1.3 Page Equation

The Page equation (Page 1969) is based on two important assumptions. The first assumption is that during the straining process the load is taken by progressively fewer

fibres crossing the rupture line, which is also one of Kallmes et al's (Kallmes 1977; Kallmes 1978) assumptions. The paper will break catastrophically when the fibres lying in the direction of the loading reach their rupture strain. According to Page, this implies that the fibres across the failure line can be divided into two fractions. One fraction (n_f) is composed of fibres that take the load at failure and then break, the other fraction (n_f) consists of fibres that pull out intact due to prior bond breakage and hence carry no load at paper failure. Using this assumption in combination with the relationship between finite-span tensile strength and zero span tensile strength (Z) and the Cox (Cox 1952) result of a Poisson's ratio of one third for random sheets, the breaking length of a sheet (T) (*km*) was expressed as the following equation:

$$T = 8n_f Z / 9(n_f + n_p)$$
 2.9

The second assumption assumes that the number of fibres pulled out to the number of fibres broken is only dependent on the ratio of fibre strength (ϕ) and bond strength (β) i.e. $n_f / n_p = f(\phi / \beta)$. The simplest form of the function, $n_f / n_p = \phi / \beta$, was chosen in the Page equation:

$$\frac{1}{T} = \frac{9}{8} \left[\frac{1}{Z} + \frac{1}{Z} \frac{\phi}{\beta} \right]$$
 2.10

After substitution of the bond strength and the fibre strength in Equation 2.10, Page gave the model for tensile strength as:

$$\frac{1}{T} = \frac{9}{8Z} + \frac{12A\rho g}{bPL(RBA)}$$
 2.11

where Z is the zero span breaking length (km), A is the average fibre cross section area (m^2) , ρ is the density of the fibre (g/m^3) , g is the acceleration due to gravity (m/s^2) , P is the perimeter of the fibre cross section (m), L is the length of the fibre (m) and RBA is the relative bonded area.

Verification and the validity of the Page equation and the KBP model have been discussed elsewhere (El-Hosseiny and Abson 1983; Williams 1983) and are also discussed in section 2.6.4. One point that should be noted is that both Page and Kallmes et al. believe that the failure of fibres oriented in the direction of sheet strain initiates the failure of the well-bonded sheet.

2.5.1.4 Allan-Neogi Model

Allan and Neogi (Allan and Neogi 1974) attempted to find an even more fundamental equation than the Page equation. The Allan-Neogi model makes a similar assumption as the first premise of Page's theory i.e. the fibres that have already debonded before the sheet reaches the peak stress do not contribute to the strength. Using this assumption in combination with the concept of zero span tensile strength and a Poisson ratio of 1/3 for a random sheet gives the following expression for sheet strength:

$$T = Z(1 - f_p) = Z(1 - \frac{14\sigma_f}{9b\sigma_b l})$$
2.12

where T is the breaking length of the sheet (km), Z is the zero-span tensile strength (km), f_p is the fraction of fibres pulled out cross the failure line, σ_f is the tensile strength of a fibre (N), σ_b is the bond strength per unit bond length (N/m) and l is the fibre length (m).

2.5.1.5 Kane Model

The Kane (Kane 1959a; Kane 1959b) model is a pull-out model assumes that paper is a thin, randomly orientated, fibre reinforced web whose tensile strength is gained either by the load to pull out fibres, and/or breaking fibres crossing the failure line. The behaviour of fibres at the failure of paper sheet depends on the length of fibres embedded into the fibrous matrix (referred to as the effective length by Kane). The critical event is described by the definition of the critical fibre length (λ_c). According to Kane's theory, fibres crossing the separation line pull out if the fibre's embedded length is less than half the critical length, and break if the fibre's embedded length is greater than half the critical length.

The theory also assumes that all fibres crossing the separation line pull out for the case of weakly bonded sheets while fibres break or pull out in the case of strongly bonded sheets. This results in two expressions for the tensile strength of paper. For weakly bonded sheets tensile failure load is expressed as:

$$T = PN_a \lambda_{ma} = P\left(\frac{2n_a \lambda_a l}{\pi}\right) \left(\frac{\lambda_a}{4}\right) = \frac{Pn_a \lambda_a^2 l}{2\pi}$$
 2.13

where *T* is the tensile failure load of weakly bonded sheet according to Kane model (N), *P* is the constant of proportionality which is a function of the static friction between unbonded fibre contacts and the bonding between fibres (N/m), N_a is the number of fibres that cross the line of separation, λ_{ma} is the mean embedded length of N_a fibres (m), n_a is the number of fibres per unit area in the plane of the sheet (m^{-2}) , λ_a is the length of a fibre (m), and *l* is the length of the fracture line (m).

For strongly bonded sheets $(\lambda_a > \lambda_c)$ the tensile failure load is given by:

$$T = PN_b \lambda_{mb} + CN_c = \frac{Pn_a \lambda^2 c l}{2\pi} + \frac{4cn_a (\lambda_a - \lambda_c)l}{2\pi}$$
2.14

where N_b is the number of fibres of embedded length less than half the critical length crossing the line of separation, λ_{mb} is the mean embedded length of the N_b fibres (m), N_c is the number of fibres of embedded length greater than half the critical length that cross the line of separation, C is the tensile strength of a fibre (N) and $\lambda_c (= 2C/P)$ is the critical fibre length (m).

The Kane model calculates the tensile failure load of a sheet by accumulating the forces required for pulling out fibres and breaking fibres that cross the failure line without accounting for the fibre orientation. The model actually assumes that fibres are perpendicular to the line of separation.

The Kane model gives a definition of the critical fibre length to describe the fibre behavior at sheet failure. However, the proportionality "*P*" used in the critical length calculation is not well defined. Moreover, the fibre behaviour at sheet failure, as described by the critical length, may only be true for thin paper.

2.5.1.6 Shallhorn and Karnis Model

Like the Kane model, the Shallhorn and Karnis Model (Shallhorn and Karnis 1979) is also a pull-out model. The Shallhorn and Karnis model considers paper as a continuum, in which all fibres have identical properties, and all are aligned in the direction of the applied load. The tensile strength of paper is calculated by summing the forces required either to pull out fibres totally or to break and pull out fibres across the failure line of the sheet. Whether the fibres will pull out or break depends on the length embedded in the matrix.

Johnston et al. (Johnston 1997) extended the Shallhorn-Karnis model to incorporate fibre length and orientation distributions. Johnston et al. showed that mean fibre length alone is a poor predictor of sheet strength. El-Hosseiny et al. (El-Hosseiny and Abson 1983) also found the ratio of fibre length to fibre strength is a more powerful predictor than length alone.

2.5.2 Simulation Models

Simulation methods have been widely used to study the strength properties in recent years. With the help of computer, a fibre network is first generated and its properties are then analyzed by means of the finite element method. Simulation studies attempt to describe the stress-strain behaviour of paper at the fibre level, something that is believed to be insoluble by conventional analytical techniques (Aström and Niskanen 1991; Heyden and Gustafsson 1998).

In the earliest studies, the network models are very simple in structure. For example, Rigdahl's (Rigdahl 1984) model is a two-dimensional mesh. As computer capacity has increased, more complicated network models have been simulated. Many models are random two-dimensional networks in which fibres are modelled as linear elastic beams of identical length. Heyden et al. (Heyden and Gustafsson 1998) introduced curled fibres and studied the effect of curl on the elastic stiffness of networks. Räisänen et al. (Räisänen 1996) introduced elastic-plastic fibres in the network, and found the shape of the stress-strain curve of the network is similar to that of a single fibre. Niskanen et al

(Niskanen 1997) made a novel approach to study the three-dimensional network structure of paper. The KCL-Pakka model combined different furnishes and filler into a random fibre network with a porous planar structure very similar to that of real paper. From the generated structure, all paper properties were simply related to the network geometry.

2.5.3 Comparison Between Models

Many researchers have compared these analytical models with one to the other. Williams (Williams 1983) showed that the Page equation is a limiting form of the KBP model. De Ruvo et al (Ruvo 1986) showed that the limiting form of the Page equation is of the shear lag analysis type and shows a direct resemblance to the KBP model. Niskanen (Niskanen 1993) showed that the Shallhorn-Karnis model could be derived from the two premises of the Page equation. More recently, Jayaraman and Kortschot (Jayaraman and Kortschot 1998) reviewed the analytical models for tensile strength of paper. They compared all of the six analytical models reviewed here and found that a striking similarity exists between the expressions for the weakly bonded paper provide by the Kane model, the KBP model and the Shallhorn-Karnis model, and that an impressive resemblance also exist between the expressions for the strongly bonded paper provided by the Kane model, the KBP model, the Allan-Neogi model and the Shallhorn-Karnis model, outside of the numerical constants. In summary, these analytical models for the tensile strength of paper are in general very similar to each other.

The complexity of the structure of paper and the wide distributions of fibre properties makes it difficult to model paper tensile strength. Thus, the modeling always starts from some assumptions. These assumptions simplify the structure of paper and idealize the properties of its component fibres, but are the major limitations of the network models.

The Page equation (Page 1969) and the KBP model (Kallmes 1977) have a common assumption that the fracture of paper is initiated by the failure of the fibres oriented in the direction of the applied load. On the other hand, some researchers (Van Den Akker 1958) believed that fibre bonds control the tensile strength of paper. Paper fails when

the external stress equals the bond yielding threshold. This viewpoint is encouraged by the experimental observation that many bonds fail before macroscopic failure has commenced, and many fibres rupture only after the macroscopic failure has commenced (Van Den Akker 1958). However, this observation gives no information about the 'trigger' of the rupture of the paper. There is still a controversy over whether bond or fibre failure initiates the rupture of paper.

All of the closed-form models, reviewed here except the KBP model, focus on calculation of forces required for breaking or pulling out of fibres only at the moment of sheet rupture. The KBP model (Kallmes 1977) tried to describe the stress-strain curve of sheet with the basic idea that the breakage of fibre-fibre bonds is solely responsible for the plastic behaviour of the sheet. Seth and Page (Seth 1983) argued against this point by showing experimentally that the plasticity of paper is mainly caused by the plasticity of fibre and the effect of breakage of bonds is usually small. The computer simulations of Räisänen et al. (Räisänen 1996) support Seth and Page's viewpoint.

2.6 Experimental verification of models

It is essential to verify the models in order to fully realize the potential of the models. Verification of the models with experimental data is also important for further refining the models. Therefore it is important to be able to measure the quantities involved in the models. Table 2-1 gives the variables that are required to calculate strength from a given model. Some of the quantities can be easily measured. However, most of the quantities are very difficult to measure. This subsection discusses the techniques for measurement of all of these quantities.

	Variable	Nature of	Method of Measurement
		Measurement	
Fibre	Fibre length	Easy to measure	Various commercial fibre analysers
properties	Fibre strength	Difficult to measure	Zero-span, single fibre measurements
		directly	
	Fibre shape	Difficult to measure	Coarseness (indirect measurement),
		directly	Microscopic methods (direct measurement)
Fibre	Apparent sheet	Easy to measure	Standard methods
network	density		
properties	Fibre-fibre	Difficult to measure	Microscopic methods
	contacts		
	Relative bonded	Difficult to measure	Optical method, Nitrogen adsorption,
	area (RBA)		Microscopic methods
	Bond strength	Very difficult to	No reliable method exists
		measure	

Table 2-1	Variables no	ed for cal	culating	sheet strengt	h from a	given	model

2.6.1 Easily measured quantities

2.6.1.1 Fibre length

Several methods have been employed for length measurement of wood fibres. In early studies, fibre length has been successfully measured by optical microscopy. This method was found to be too tedious. Fibre length has also been measured by using classifiers, such as the Bauer-McNett fractionator (Clark 1985). This is a series of cascading vertical screens, which separate stirred holding tanks of pulp. Most fibres pass through the coarser screens and as the screens get finer, the fibres that pass through them are generally shorter in length. The Bauer-McNett readings are traditionally translated into values of fibre length measurement by calculations using TAPPI method T-233.

The Kajaani Fibre Analyser is the most widely used device for measuring the distributions of fibre length (Bentley 1994). The Kajaani Fibre length Analyser measures single fibres which are drawn from a dilute 0.01% consistency through a narrow capillary, under suction. The fibres pass through a beam of polarised light.

Cellulose fibres, being birefringent to polarised light, create an interval of birefringence as they pass through. The interval is related to the fibre length of the fibres. The method is fast as it can measure thousands of fibres in a short space of time, and it can report statistical information on the fibre distributions. The significant difference between the Kajaani analyser and the Bauer-McNett classifier is that the former measures the number of fibres in each fraction whereas the latter measures the fraction weights. Bentley et al. (Bentley 1994) found that the precision of measurement of the two methods is comparable, generally to within $\pm 3 - 4\%$. In recent years, several other instruments to measure fibre length have also been developed, such as the FQA (Robertson, Olson et al. 1999), FibreMaster (Mohlin, Dahlbom et al. 1996) and CyberSizer (Lehto 2004).

2.6.1.2 Sheet density

The sheet density is usually calculated based on the measurements of the grammage and thickness of the sheet. The thickness is defined as the thickness of sheet under standard conditions of platen size and pressure. Some researchers have questioned the accuracy of the measurement of the thickness, and hence, the calculation of sheet density (Taylor 1964; Fellers 1986; Yamauchi 1987). They put forward several reasons for this. First, paper is compressible and the thickness, thus density, depends on the load applied during the measurement. Second, the surface roughness of paper affects the thickness measurement, which introduces an error in the density. The "density" and the compressibility of the surface regions are different from those in the bulk. Although these do affect the measurement of paper thickness, and therefore the calculation of density, the standard methods for measuring the sheet density are still commonly used. In this project, the APPITA standard method AS/NZS 1301.426s-94: 1992 was used for measuring sheet density.

2.6.2 More difficult to measure quantities

2.6.2.1 Fibre strength

Many attempts were made at measuring single fibre strength. Mark and Gillis (Mark 1994) presented an excellent summary of the techniques used to measure single fibre strength. The procedure for handling the samples is usually tedious and time consuming. More recently, Denis (Denis 1996) showed that there were difficulties in obtaining reliable and statistically valid results when measuring the single fibre strength of eucalypt fibres.

Due to the difficulties of measuring single fibre strength, the zero span tensile strength is the most commonly used method to determine the strength of fibres in the sheet (Van Den Akker 1958; Page 1969; Boucai 1971; Kallmes 1977). It has been shown that the zero-span tensile test provides a valid measure of fibre strength (Van Den Akker 1958) provided that the sheet grammage is kept at 60g/m² or under (Batchelor 2003). The zero-span tensile test indicates the strength of fibres while they are in the sheet. This is also important as the degree of drying restraint strongly influences fibre strength and it is difficult to replicate sheet drying conditions when preparing single fibres for testing. Another possible advantage of the zero-span test is that it is affected by distributions in fibre strength (El-Hosseiny and Bennett 1985). That is, the larger the distribution in fibre strength, the more the zero-span tensile strength will be reduced below the strength based on the expected average. A larger distribution of fibre strength should also have a similar effect on paper strength.

2.6.2.2 Fibre shape

In early studies, researchers used fibre coarseness to describe the cross-sections of fibres (Seth 1990; Paavilainen 1993). By measuring the total length of a known weight of sample, an average of coarseness of the fibres can be calculated. The coarseness can be adequately measured on most fibre length analysers but not the Kajaani F200 (Seth 1997), as the Kajaani F200 does not reliably count all particles.

In later studies, researchers tend to directly measure fibre wall area, fibre wall thickness, fibre lumen area, fibre width and thickness to characterize the cross-sections of the fibres. The first reliable measurement of fibre cross-sectional dimensions seems to be contributed by Kibblewhite and Bailey (Kibblewhite 1988). They used a technique of embedding fibres vertically in a resin block and sectioning the block to expose the cross-sections of the fibres. The fibres. The cross-sections of the fibres were then measured in either a SEM or light microscope. The fibre cross-section parameters they measured were width, thickness, cross-section wall area, fibre wall thickness and the fractions of wall and lumen areas.

In recent studies, the most commonly used technique is confocal laser scanning microscopy combined with image analysis. This technique has been used for measurement of fibre transverse dimensions and shapes of different pulp fibres (Jang 1992; Jang 1995; Seth 1997; Xu, Filonenko et al. 1997; Conn 1999). Nowadays, the measurement of fibre transverse dimensions and shapes using a confocal microscope combined with image analysis has become routine process.

The above studies measured the fibre transverse dimensions either on fibres freely dried on glass or embedded in resin. The measured dimensions were then used to predict the structure and physical properties of the paper made with the pulp. It is more important to measure the fibre transverse dimensions in a real paper structure since the shape of the fibres in a sheet dried under restraint may be different from that of freely dried fibres on glass slides. The shape of fibres in a sheet can also be changed significantly by the papermaking process, such as wet pressing, for example.

Several researchers have attempted to measured fibre dimensions in paper. Gorres, et al. (Gorres 1993) measured the transverse dimensions of fibres in very thin fibre networks pressed on glass slides. Hasuike et al (Hasuike 1992) have re-constructed the 3-D structure of paper, from a series of cross-sectional images of the paper, by using computer tomography. The sample size examined by this technique was quite small (0.2x0.2mm), therefore may not be fully representative. Recent studies also generated 3-D images of paper structure by using synchrotron x-ray source (Christine 2002). Although the authors did not measure the dimensions of individual fibres in the sheet, it seems that the quality of the images is not good enough for such measurements.

Dickson (Dickson 2000a) obtained quality images of paper cross-sections by employing a combined technique of resin embedding and laser scanning confocal microscopy. Dickson measured the dimensions of the fibres in the sheet cross-sections. However, the orientations of the fibres are still unknown, thus the shapes of the fibres are still unknown. It appears that new techniques that can measure fibre dimensions directly in paper sheet are called for.

2.6.2.3 Fibre-fibre contacts

As has been discussed before, fibre-fibre contacts in paper, including the number of fibre-fibre contacts per unit length of fibre, the nature of contacts, the free fibre length and its distribution and the out-of-plane deflection angle of a fibre segment, are the most important fundamental properties of a paper structure. Most previous studies of fibre-fibre contacts focused on the development of models for the number of fibre-fibre contacts using statistical methods either based on analysis of model structures of paper (Kallmes and Corte 1960; Kallmes 1961) or fibre assemblies (Komori and Makishima 1977; Pan 1993; Komori and Itoh 1994). These models have provided valuable knowledge for understanding of the fibre-fibre contacts in paper. However, verification of these models is always problematic because there is little experimental data on the properties of fibre-fibre contacts in paper. There is always a requirement for new techniques for measuring comprehensive data of fibre-fibre contacts directly in a sheet.

Some techniques have been used for measuring fibre-fibre contacts directly in a sheet, including examining the paper plane (x-y plane) of very thin fibre networks by their magnified projected images (Kallmes and Corte 1960), examining the x-y plane of a normal sheet under vertical illumination in polarized light (Kallmes 1961; Page 1962), and examining serial cross-sections of paper embedded in resin (Yang 1978). The technique of Page *et al.* (Page 1962) and the technique of Kallmes *et al.* (Kallmes 1961) can only measure fibres on the paper surface, and can only measure interbond distances between bonds on one side of the fibre. Page *et al.* (Page 1962) estimated the interbond distances within the sheet by superimposing two sets of measurements, one set representing the upper surface of the fibre and the other the lower surface. Yang *et al.* (Yang 1978) examined a series of images of paper cross-section acquired at different

depths from the paper cross-sectional surface, and measured the probability of fibrefibre contacts with a given fibre cross-section. This technique overcomes the limitations of only measuring the surface fibres, however, it cannot measure the interbond distances. In a later study, a new technique of evaluating the 3-dimensional geometric arrangement of fibres in a paper sheet was developed based on the measurement of the locations of fibre segments in the successive cross-sections of the sheet (Hasuike 1992). The relative coordinate positions of fibre peripheries in the sections were recorded by a digitising system attached to a computer. Using this technique, the authors measured the possibility of fibre-fibre contacts with respect to a given fibre cross-section and the number of fibre-fibre contacts per unit length of fibre. The sample size examined by this technique was quite small (0.2x0.2mm), and therefore may not be fully representative. It appears that a more powerful technique is required for measurement of fibre-fibre contacts directly in a paper sheet

2.6.2.4 Relative bonded area (RBA)

The measurement of *RBA* is always problematic. Traditionally, the Ingmansson and Thode pressing /beating/ extrapolation method (Ingmanson and Thode 1959) was often used for *RBA* measurement.

This method measures the Kubelka-Munk light scattering coefficient, $S(m^2/kg)$, of paper made from pulps with different beating degrees or of paper pressed under different wet pressing pressures. The results of light scattering coefficient are then plotted against the tensile strength of the paper. The light scattering coefficient, S_0 , of the unbonded sheet is obtained by extrapolating the plot to zero tensile strength of paper. *RBA* is then calculated from following equation:

$$RBA = \frac{(S_0 - S)}{S_0}$$
 2.15

The scattering coefficient S is the reflectance of a single sheet backed by a black cavity. The principle of the light scattering coefficient method is that a fibre surface element appears bonded if there is another fibre surface at a distance smaller than half the wavelength of the light used in the measurement. Two such fibre surfaces can form an optical contact. The Ingmansson and Thode pressing/beating/ extrapolation method

relies on at least two presumptions. Firstly, it is implicitly assumed that the light scattering properties of the sheet can be used to distinguish between bonded and unbonded areas even though the wavelength of light is at least one hundred times the bond length for hydrogen bonding. The second assumption is that the value, S_0 , determined by extrapolating the plot of tensile strength and light scattering coefficient is equal to the scattering coefficient of an unbonded sheet.

Ingmansson and Thode's study (Ingmanson and Thode 1959), showed that the relationship between S and tensile strength was independent of the degree of beating, and the data for various degrees of wet pressing at all refining intervals fell on one common curve. They concluded that the total dry fibre surface area available for fibre bonding, which is effective in developing tensile strength, remains constant with refining time and is unaffected by production of fines or the degree of fibrillation. In contrast, Swanson and Steber (Swanson 1959) found the plot of tensile strength and the scattering coefficient data obtained with various degrees of wet pressing and at different beating intervals did not fall on one common curve. Instead, the data points seemed to form separate curves for each refining interval rather than a single common curve. Nordman and Gustafsson (Nordman 1951) also noted separate curves for the scattering coefficient-tensile strength relationship for a variety of pulps beaten in the Valley beater. They suggested that this was due to the fibrillar structure of some pulps being retained during drying. Tensile strength can also change in beating in part for other reasons than an increase in *RBA*.

Niskanen (Niskanen 1998) has also criticised the method on the grounds that the tensile strength will fall to zero long before the bonding completely disappears in the network. The method is also completely unsuitable for measuring bonding in machine made papers, since a range of sheets with different strengths cannot be obtained.

Since the extrapolation of tensile strength usually does not give a reliable value for S_0 , some other studies worked on forming unbonded sheets and measuring S_0 directly (Parsons 1942; Ratliff 1949; Keeny 1952; Braaten 2000). Parsons (Parsons 1942) was the first to make an unbonded sheet. The sheet was formed in a non-polar liquid and suffered from poor formation. Ratliff (Ratliff 1949) improved the technique but still the sheets were formed in a non-polar liquid. Keeny (Keeny 1952) first formed sheets from water in an ordinary sheet former and then displaced the water by an alcohol and a non-polar liquid. These techniques are usually laborious and involve using hazardous chemicals. Drying the sheets from the non-polar liquid avoided forming inter and intra-fibre bonds. The non-polar liquid dried unbonded sheets avoid the formation of inter fibre bonds but the intrafibre bonds as well. Too high a scattering coefficient was yielded because of the intra-fibre scattering. A freeze drying technique has also been used to form unbonded sheets (Merchant 1957; Braaten 2000). Although the freeze-dried unbonded sheets were dried in a water base the inter-fibre bonds and intra-fibre bonds and intra-fibre bonds could not form. The additional intra-fibre scattering still results in an overestimate for the value of S_0 .

El-Hosseiny and Abson (El-Hosseiny 1979) have tried to relate scattering coefficient to density. They found that for a wider range of pulps, scattering coefficient was linearly related to sheet density. This in turn was used to estimate *RBA*. However, this method has not been further applied.

Nitrogen adsorption is another important technique that has been used to measure fibre surface area to calculate *RBA*. Haselton (Haselton 1954; Haselton 1955) is the first to propose the use of gas adsorption as a method of measurement of the surface area and bonded area of papers. After comparing bonded areas calculated by four methods he concluded that a value closest to the true bonded area could be obtained by applying a correction factor to the regular optical values. When nitrogen adsorption is employed for area studies, the surface area accessible to nitrogen molecules of approximately 4.3Å thickness (N₂ diameter is 16.3 Å²) is determine d. The bonded area in the external and internal portions of fibres is believed to be separated by distances of 4 to 5Å or less. Therefore, it is reasonable to believe that nitrogen is not absorbed on areas involved in bonding and that adsorption methods may provide an excellent tool for the measurement of the unbonded internal and external area of cellulose materials. The Brunauer, Emmett and Teller (B.E.T.) theory (Brunauer 1938) was used to calculate the volume of gas required to form a monolayer on a given adsorbent. The BET equation is given as:

$$\frac{p/p_0}{v(1-p/p_0)} = \frac{1}{v_m c} + \frac{c-1}{v_m c} (p/p_0)$$
 2.16

where p/p_0 is relative pressure, v is the volume of adsorbed gas (cm³/g STP) per gram of adsorbent at a certain relative pressure, v_m (cm³ STP) is the volume of gas per gram of sample required to form a monolayer, and c is a constant.

The plot of $\frac{p/p_0}{v(1-p/p_0)}$ against p/p_0 should therefore be a straight line with slope $s = (c-1)/v_m c$ and intercept $i = 1/v_m c$. Solution of the two simultaneous equations gives v_m and c. Multiplying the number of gas molecules corresponding to v_m by the cross-sectional area of each molecule gives the area of the solid accessible to the gas.

$$A = \frac{N v_m L}{22,400} x 10^{-20}$$
 2.17

where $A (m^2/g)$ is the specific area of the sample, N is Avogadro's number and $L(m^2)$ is the molecular cross-sectional area of the gas absorbed.

Another group of researchers attempt to measure *RBA* by analysis of sheet crosssections (Yang 1978; Paavilainen 1994; Niskanen 2002). These measurements are time consuming, involving a great amount of microscopy work. Niskanen and Rajatora (Niskanen 2002) compared their measured values of *RBA* with those reported by Yang et al (Yang 1978) and Paavilainen (Paavilainen 1994). The comparison showed that their values of *RBA* were smaller than the values measured by the other two studies. Niskanen and Rajatora (Niskanen 2002) emphasised the importance of the threshold separation for any measurement of *RBA* by analysis of cross-sections of paper. The values of *RBA* reported by these studies are, in general, smaller than those measured by indirect methods, for example in (Retulainen and Ebeling 1993; Braaten 2000).

Although numerous studies have been conducted on *RBA* measurement, the requirement still exists for new and reasonable simple techniques that can determine *RBA* with reasonably accuracy.

2.6.3 Very difficult to measure quantity

2.6.3.1 Bond strength

The bond strength, defined as the strength per unit bonded area, is a straightforward concept, but it is a parameter that is extremely difficult to measure. Numerous studies have focused on measurement of the strength of fibre-fibre bonds.

Earlier researchers (Nordman 1957; Nordman 1965; Skowronski 1991) focused on sheet Nordman (Nordman 1957) defined a so-called "Nordman Bond measurements. Strength" (NBS) as the ratio of the irreversible work of straining the sheet to the change in the scattering coefficient. Subsequent work by Nordman et al (Nordman 1965) revealed that the bond strength appears to be a characteristic of pulp. They found that the bonding strength of fibres was essentially constant in pulps of low hemicellulose content, while for high hemicellulose content pulps it appeared to increase with beating and decreased with wet pressing. Over the years many researchers have used the NBS in their studies. While some evidence supports it, other evidence contradicts it. Page (Page 2002) has shown that the Nordman procedure for determination of bond strength is fundamentally flawed as the work of straining a sheet is not consumed by bond breakage. It is consumed by the fibres as they deform plastically. Fibre-fibre bonds break because, when a sheet is strained, shear stresses are induced at the perimeter of each bond site. The bond breakage is seen as a strain-induced phenomenon rather than an energy-induced phenomenon.

Several investigators have directly measured the shear strength of individually prepared single fibre-fibre, fibre-shive, or fibre-cellulose-film crossings (McIntosh 1962; Page 1962; Schniewind 1964; Mohlin 1974). Such measurements are tedious, and the results suffer from large variability because of damage during testing, failure by peeling rather than shear, and errors in measuring the bonded area in the crossings. Moreover, these methods measure the strength of the bond structure rather than the shear strength of the interface, as pointed out in Uesaka's review (Uesaka 1984) of the measurement of fibre-fibre bond properties. Baum (Baum 1993) made a similar point. Since the S1 and S2 layers of the fibre cell wall have very different fibril angles, the two layers may respond differently to bonding stresses. This suggests that the S1-S2 interface may be a weak

zone. If the failure between two bonded fibres originates within one of the cell walls, rather than area between the fibres, as indicated in Figure 2-7, the meaning of bond strength becomes obscure. Nanko's (Nanko 1989) work suggests that such events are not rare. More recently, Stratton and Colson (Stratton 1993) also have reported similar fibre wall damage during bond failure. In addition to the above concerns, a single fibre-fibre bond test may not be able to represent the bonds in a sheet because fibres in the sheet will likely have properties markedly different from fibres dried outside the network (Baum 1993).



Figure 2-7 A schematic diagram illustrating a weak S1-S2 interface (Baum 1993).

In summary, as pointed out by Niskanen (Niskanen 1993) in his review paper, the microscopic interpretation of bond strength is still unclear.

2.6.4 Attempts at partial verification of models

Many attempts have been made to verify the models developed in previous studies. It is not necessary to review the verifications of every model. We will only discuss the verifications of the Page equation and the KBP model. The major point we will try to make is that it is usually very difficult to fully verify a model for tensile strength.

Page (Page 1969) tested his model by using data from the literature that indeed showed linear relationships between 1/T and 1/L and between 1/T and 1/RBA (refer to Equation 2.11). He further verified the model by calculating the zero-span tensile strength of the paper from the intercepts on the ordinates of these graphs. This was done by finding the values of 1/T for the cases 1/L = 0 or 1/RBA = 0 from these

graphs and calculating zero-span strength, Z, from the expression Z = 9T/8. The calculated values of Z have shown quite reasonable agreement with the measured values. Obviously, such verification of the model is quite limited.

Kallmes *et al* tested the KBP model by using data mainly from Seth and Page's work (Seth and Page 1975). They compared the calculated values of zero-span strength with the predicted values. The zero-span strength was calculated by arbitrarily setting the ratio of the proportion of active fibres at sheet failure to that at initiation of sheet strain equal to 1. The comparisons showed good agreements. Interestingly, they used the same data of Ingmanson and Thode (Ingmanson and Thode 1959) (which has also been used by Page) to show that the relationship between T/Z and RBA is linear when RBA is less than RBA_{cr} and non-linear when RBA is greater than RBA_{cr} thereby partially verifing the theory.

In fact, all of the models developed in previous studies have never been completely verified because some quantities, as discussed above, are very difficult to measure. It is of critical importance to develop new techniques to measure those parameters that were not able to be measured in previous studies.

2.7 Summary

Paper obtains its tensile strength from the strength of the fibres bonded together through hydrogen bonds in the network. How efficiently the fibre network uses the strength of the fibres depends on the properties of the network and the fibres. These properties include fibre length, bond area, fibre-fibre contacts, relative bonded area, sheet density and formation. One should note that these properties are not necessarily independent. Some of the relationships between them will be discussed in detail in later chapters.

Modelling the tensile strength of paper has to answer two questions. What are the mechanisms by which the stress is transferred from one fibre to other fibres bonded with it? What initiates the macroscopic fracture of paper? Many researchers have attempted to answer these two questions. Cox developed the shear-lag model based on an effective medium approximation for a fibre in equilibrium with average surroundings.

The shear lag model was later applied to fibre networks by Page and Seth. This model predicts that stresses transferred from one fibre to its adjacent fibres via shearing the bonds in the regions of the fibre ends. Therefore, the stress along the length of the fibre is at its maximum at the centre and diminishes to zero at the ends. The shear lag model has been shown to be valid for random fibre networks at high densities by computer simulations. There is still a controversy over the initiation of the macroscopic fracture of the paper. Two viewpoints exist – the bond failure triggered and the fibre fracture triggered. Some researchers tried to answer the question by providing some indirect evidence. Others tried to directly observe the fracture process of the paper. However, there is still no satisfactory evidence to support one viewpoint against the other. Kallmes *et al* described the initiation of the macroscopic fracture in their analysis of the zero-span tensile strength of paper. They concluded that it is the rupture of comparable fibres that triggers the final catastrophic failure of a sheet in a conventional tensile test. When the first fibres of a sheet fail in a conventional tensile test, a large amount of energy is released in an impact-type manner to their immediate neighbours. One can readily visualise that these fibres, or the bonds between them, fail in turn, causing further failures. In other words, the rupture of the first fibres triggers the catastrophic final failure of the sheet. This rupture once initiated at a point in the sheet, is propagated rapidly in two directions. The above mechanism for the initiation of the fracture of paper is quite reasonable although the authors could not provide direct proof for it. One question that needs to be answered is how we get fibre fracture if bond failure is the trigger.

Studies of modelling tensile strength of paper would only be completed when the final model has been fully verified with experimental data. In other word, verification of the models is a critical part of such studies. However, verification of the previous models is always problematic because some of the quantities involved in these models are very difficult to measure, such as bond strength, fibre dimensions in paper, relative bonded area and fibre-fibre contacts etc. As has been discussed, although numerous attempts have been made to develop new techniques for measuring these quantities, there are still, to the best of our knowledge, no techniques available for measuring these quantities with reasonable accuracy. It appears that development of new techniques, especially microscopic techniques, for measuring these quantities is of first importance for any further studies of modelling tensile strength of paper. Such techniques are not

only important for providing experimental verifications of the final model, but also important for better understanding the paper structure.