III

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Observations on interfibre bonding and fibre segment activation based on the strength properties of laboratory sheets

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KEYWORDS: Bonds, Bonding, Activation, Drying stress, Beating, Paper strength properties

SUMMARY: The aim of this study was to gain better understanding of bonding and activation in paper and to clarify the relationships between these two phenomena and the strength properties of paper. Bonding and activation were studied through examining the mechanical properties of paper. Both in-plane strength properties (tensile strength, tensile stiffness, elastic breaking strain and in-plane tear strength) and z-directional strength properties (Scott bond strength) were examined. The properties of fibre network were varied by beating of chemical pulp fibres and by introducing different levels of drying stress to the network at different solids contents. Different mixtures of chemical and mechanical pulps were used as well. As a generalisation, it can be said that the in-plane properties of laboratory sheets were improved by increasing drying stress. This improvement arises from increasing activation, ie. the removal of slackness and curls from the unbonded fibre segments within the network. Activation straightens those segments into load-bearing units. On the other hand, z-directional strength properties, or bond strength, in most cases decreased by increasing drying stress. The strain introduced to the network affects the bonded areas of fibres negatively, but the extent of this effect depends quite much on the fibre composition, degree of drying stress and the solids content at which strain has been introduced. It seems that the structure of the bonded area influences sheet behaviour and the development of strength properties significantly. Bonding and activation are both essential properties of fibre network in relation to its strength properties.

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An *interfibre bond* can be defined as the zone where two fibres are so close to each other that hydrogen bonding, van der Waals' interaction or molecular entanglement can occur (Retulainen et al. 1998). Bonds hold fibres together and therefore contribute to the internal cohesion of paper. Mechanical interaction is made possible through interfibre connections. Formation of interfibre bonds begins as the web's solids content increases during papermaking. At first, bonding happens through surface tension forces pulling fibres together when water is removed (Campbell effect) (Campbell 1959). The Campbell effect changes gradually to hydrogen bonding. The solids content at which actual interfibre bonding starts is not known exactly. The bonding layer is formed when external fibrils and fines come close during dewatering and couching, and become pressed and packed between fibres. As the web dries, external fibrils and fines material form the bonding layer. Since microfibrils are more likely to form hydrogen bonds, they can contribute more to the strength of the bonding layer (Nanko and Ohsawa 1989). Formation of bonds is promoted by external fibrillation and fines, which link two fibre surfaces more closely together. Internal fibrillation contributes to the swelling and flexibility of fibres and therefore improves interfibre bonding: adjacent fibres are able to conform onto each other during pressing and drying. Fibre-water interactions also influence bond formation (Retulainen et al. 1998).

The structure of interfibre bonds is influenced by beating, pressing and drying. Other important factors affecting the bond structure include fibre morphology and the pulping procedure. Fines play a significant role in bonding. Flexible fibres can form wrap-around type of bonds and fibrils, and fines form bridges between fibres (Retulainen et al. 1993). On a microscopical scale, fibrils also form entanglements (Uesaka 1984). Nanko and Ohsawa (1989) studied the structure of fibre bonds. According to their findings, an amorphous bonding layer is formed between the S1 layer of two beaten fibres by external fibrils, and probably polymer chains as well. The more beaten the fibres are, the thicker the bonding layer and the better the contact between two fibres will be, partly because of the increased amount of fines. They also detected structures like skirts (elongated part of the S1 layer extending from bond edges) and covering layers consisting of external fibrils and fines covering smooth edges. The skirt and covering layer structures have an important role in strengthening the bond.

Fibre segment activation means the modification of originally kinky, curly or otherwise deformed fibre segments, unable to carry load, into active components of the network. Activation of fibre network occurs during drying, when lateral shrinkage of fibres is transformed into axial shrinkage of the neighbouring fibres in bonded areas. If this shrinkage is restrained, the free fibre segments dry under stress and the slackness is therefore removed (Lobben 1975, Giertz and Rødland 1979). Once the segments are activated the axial elastic modulus of fibres increases, which leads to further increase in drying stress. Thereafter, both the segments and bonded areas are capable of carrying load. Interfibre bonding and shrinkage of fibres are prerequisites for activation. The amount of drying stress needed to activate free segments depends on the morphology of the fibres (Retulainen 1997). Activation can be improved by maximising the bonded area and average fibre length. Beating of chemical pulp increases swelling ability (and therefore shrinkage potential) of fibres and also fibre flexibility. Beating also increases the number of bonds and the number of free segments and decreases the length of the free segments (Lobben 1975, 1976).

Drying stress acting on a fibre network has a significant effect on the final strength properties of paper. The mechanical properties of dried paper have been shown to correlate strongly with the final drying stress developed during restrained drying (Htun 1980). This relationship does not depend on the type of pulp, beating or wet pressing. During drying, also shrinkage takes place in the paper network and this will have an effect on the paper properties. The final impact of drying on paper strength is in fact a combination of drying stresses, the degree of shrinkage, the solids content at which shrinkage takes place and the drying strategy. The changes in mechanical properties caused by drying under load can be associated to two main mechanisms: first, the increase in crystallite orientation and second, more uniform distribution of stress among fibres (Jentzen 1964, Retulainen 1997).

The objective of this study was to gain better understanding of bonding and activation in paper and to clarify the relationships between these two phenomena and the strength properties of paper.

Materials and Methods

The sheets used in this study were made from bleached kraft pulp, (Finnish commercial softwood kraft pulp made from Scotch pine (*Pinus sylvestris*)) bleached TMP (Finnish commercial thermomechanical pulp made from Norway spruce (*Picea abies*)) and a 50/50 mixture of the kraft (SR° 14) and TMP. The kraft pulp was disintegrated in a Valley hollander for 30 minutes and then beaten to three different levels, for 8, 15 and 40 minutes (corresponding SR numbers 14, 17 and 34). The thermomechanical pulp (CSF 44) was hot disintegrated. Laboratory handsheets were made with a sheet mould producing 240 x 290 mm^2 test sheets. The TMP and mixture sheets were made using circulation water. The sheets were wet-pressed by 400 kPa for 5 minutes.

Drying of the sheets was carried out with the PDR (Paper Drying Rheometer) device enabling biaxial straining, with 6 x 500 W halogen lamps for drying (*Fig 1*). First, the sheets were attached to the drying device and a preliminary straining was done to obtain a 'zero' stress level. Drying was then commenced immediately after this. In order to reach different drying stresses, the sheets were strained or let relax either at the solids content straight after wet pressing (30 – 35 %) or during drying at three different solids contents: 49, 52 or 55 %. Strain levels between -1 % and +1 %, simultaneously on each direction of the test sheet, were used.

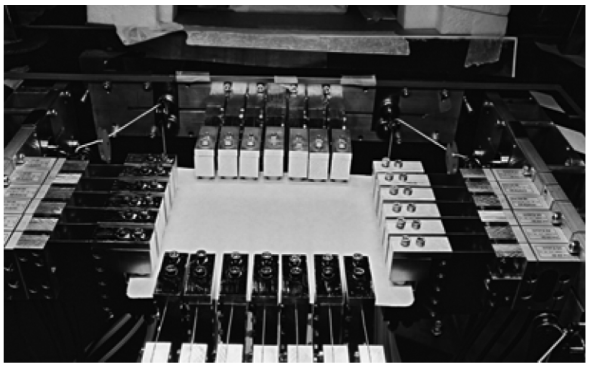

Fig 1. The Paper Drying Rheometer (PDR) with an attached test sheet.

The dried sheets were conditioned (23°C, 50 % RH) and tested for tensile properties, bond strength (Scott bond strength and elastic breaking strain) and in-plane tear strength, which was measured with MTS 400 tensile tester according to the procedure described by Kettunen and Niskanen (2000a). Also damage analysis was carried out. It produces two parameters, damage width and pull-out width: damage width measures the extent of damage or fibre de-bonding from the actual crack line, and pull-out width describes the extending of fibre ends from the crack line (Kettunen and Niskanen 2000b).

Certain assumptions were made when assessing the results of this study. Tensile stiffness or elastic modulus is an indicator of the level of activation in the sheets. Scott bond strength represents the z-directional bond strength, whereas elastic breaking strain, calculated from tensile index divided by tensile stiffness index, gives an indication of bonding in the in-plane direction. A calculated variable (in-plane tear index divided by damage width) is assumed to combine these two aspects of bonding at least to a certain extent. Damage width indicates how far a fracture will progress perpendicular to the fracture line in a paper network, and its extent depends of bond strength and fibre strength in the fibre network.

Results and Discussion

Bonding

Bonding decreases linearly with increasing drying stress in most of the test sheets (*Figs 2-7*). Increased stress seems to reduce the bonded area and even break up bonds in the sheet. Similar results have been published by Wahlström et al. (2000). The decrease is most drastic in sheets made from highly beaten pulp (*Fig 2* and *3*), probably because the initial 'capacity' for bonding is higher than in the sheets made from less beaten kraft pulp. Beating creates fines material, external fibrillation and increases fibre flexibility, which all promote bond

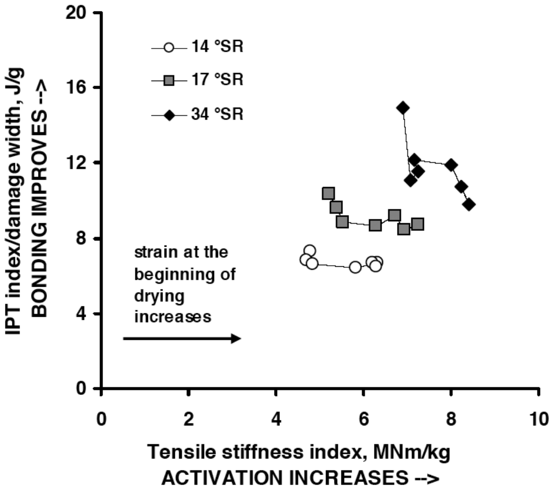

Fig 2. The relationship between bonding – activation in test sheets made of differently beaten kraft pulp, strained at a solids content of 35 %.

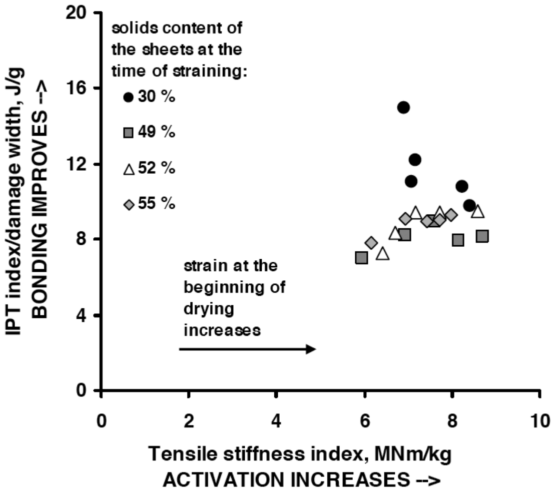

Fig 3. The relationship between bonding and activation in test sheets made of kraft pulp, 34 °SR., strained at four different solids content levels.

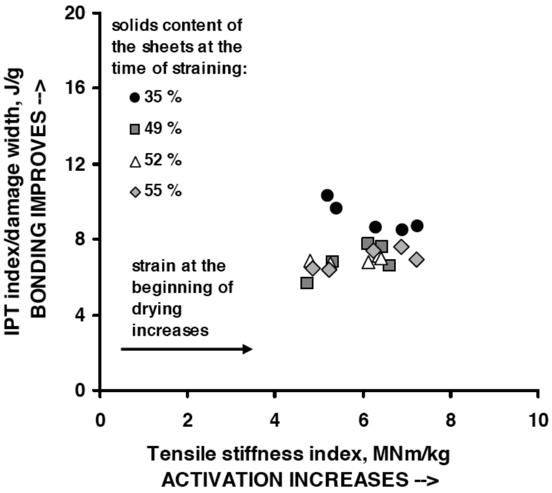

Fig 4. The relationship between bonding and activation in test sheets made of kraft pulp, 17 °SR, strained at four different solids content levels.

strength and increase the bonded area (Page 1989, Robinson 1980).

Sheets made from highly or moderately beaten kraft pulp (*Fig 3-4*) behave somewhat differently in straining, compared to the sheets made of very mildly beaten kraft pulp (*Fig 5*): there is a significant difference in bonding between the sheets strained prior to drying at a solids content of 35 % and those strained at higher solids contents during drying. Sheets that were strained prior to drying exhibit a linear decrease in bonding, whereas in the sheets strained at higher solids contents, bonding remains at a comparatively low level. In the sheets containing more water, some of the water is situated in the bond areas, which are thus probably more flexible and therefore able to yield to the strain. At the higher solids contents, the initial 'zero' strain introduced to the sheets at the beginning of the drying procedure seems to inhibit proper bond formation. Although the sheets are allowed to relax at different points during drying, the structure of the bonded areas can no longer be repaired at this later stage. This suggests that at a certain solids content level, bonds have reached their final structure. The phenomena that are most important for bond formation seem to take place at a critical moisture content level at which the Campbell effect starts to change to hydrogen bonding. According to Retulainen et al. (1998), this level is around the solids content of 50 % in kraft pulp, reflected as a sudden increase in elastic modulus. In the TMP and mixture sheets, similar boundary-level solids content cannot be detected from these results (*Fig 6-7*).

The straining itself does not cause so much damage to the bonds, once they have reached a more or less finished, solid structure (that is, in the test sheets strained at solids contents higher than 49 %). As can be seen in *Fig 3-7*, bonding remains quite stable, independent of the amount of strain introduced into the sheets, although the bonding is considerably poorer. At the strains used in these experiments, the bonds mostly seem to be able to withstand quite high drying stress, without being damaged or broken.

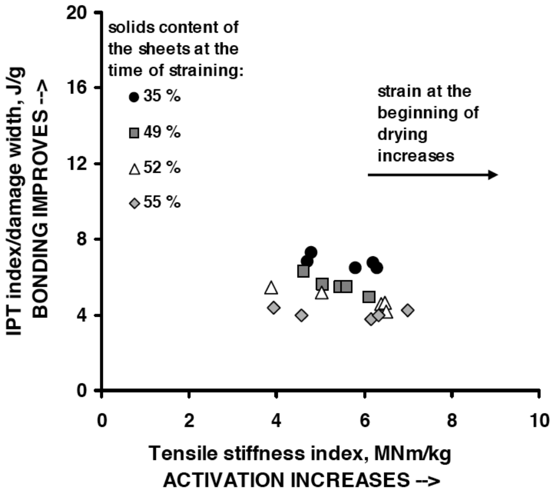

Fig 5. The relationship between bonding and activation in test sheets made of kraft pulp, 14 °SR, strained at four different solids content levels.

Sheets made from TMP and a mixture of kraft pulp and TMP and strained at different solids contents display similar differences in bond strength. However, in the TMP sheets, bond strength apparently has not developed properly until the highest solids content (55 %) (*Fig 6*). In terms of bonding and bond behaviour in drying, these two types of pulps (TMP and kraft) appear to behave somewhat differently. For example, the mechanical fines present in the TMP and mixture sheets influence the way in which moisture is removed from the fibre network, affect the structure of bonded area, and even out local stress concentrations in the network (Luukko 1998). In the mixture sheets, there is no significant bond deterioration at any of the different solids contents. Bond strength is probably affected favourably by the bond-contributing properties of both of the fibre types (i.e. by the flexibility and external fibrillation of kraft fibres and by the fines contained in TMP), so bonding is not impaired by increasing drying stress.

Activation

Activation is increased by increasing drying stress independent of the solids content at which the straining is performed (*Fig 2-7*). The straightening of initially slack fibre segments leads to an increase in the elastic modulus

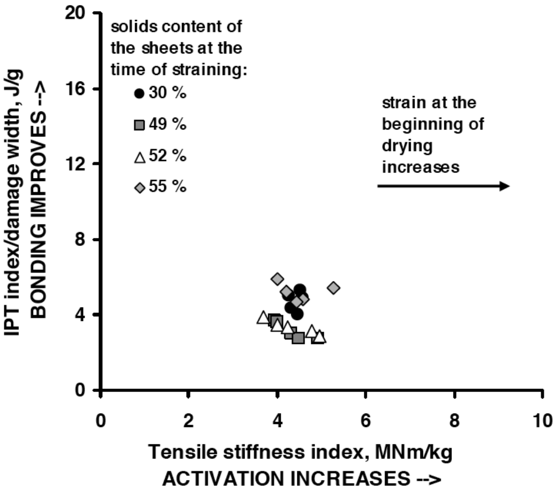

Fig 6. The relationship between bonding and activation in test sheets made of TMP, strained at four different solids content levels.

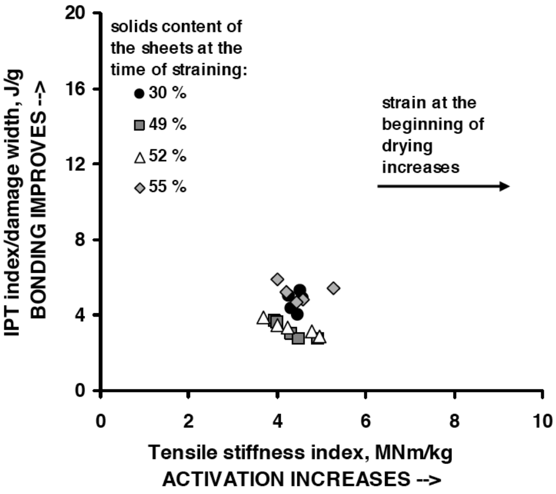

Fig 7. The relationship between bonding and activation in test sheets made of a 50-50% mixture of kraft pulp and TMP, strained at four different solids content levels.

of the paper web, as has been reported for example by Htun (1980). Tensile stiffness values five times the original can be reached with increasing strain (Wahlström 1999). The increase in activation is highest in the sheets made of kraft pulp. The beating level does not seem to have any major effect on how much the tensile stiffness increases, although sheets made of highly beaten kraft pulp display a higher level of tensile stiffness (*Fig 3*) than moderately or lightly beaten kraft sheets (*Fig 4-5*). Beating contributes favourably to the fibre properties significant for activation: fibre swelling, flexibility and bonding capacity (Hiltunen 2003). Straining of the sheets at higher solids contents (>50 %) seems to cause somewhat more activation in the kraft pulp sheets. In the kraft pulp and TMP mixture sheets (*Fig 7*), the increase in activation also behaves linearly, probably because the properties of kraft fibres contribute more to the network than those of the TMP fibres. One possible explanation for activation behaviour of highly beaten fibres is that the flexible fibres are more conformable and hence able to wrap around each others in bonding. This way the free segments of flexible fibres may also be slacker and less straight after wet pressing, and thus have greater potential for activation as straining of the sheets prior to drying then reduce their slackness. In TMP fibres the slackness of free segments is probably much less pronounced, since they are generally stiff and have a poorer swelling ability.

An exception from the expected activation behaviour can be seen in TMP sheets strained prior to drying at solids content of ~ 30 % (*Fig 6*): the test sheets still contain a lot of water, which may be located especially in the fines material concentrated around bonded areas. During straining, neighbouring fibres are able to slide over each other, because bonds have not yet reached a sufficiently dry, solid structure and therefore yield to the strain. This is also suggested by the bonding behaviour. At higher solids contents, the response of activation to straining seems to follow the normal linear trend, although the extent of activation is much lower than in the sheets made of kraft pulp or a mixture of kraft pulp and TMP. This is explained by the lower swelling ability of the TMP sheets and also by the lower bond strength (there are less bonds and the bonds are weaker between stiff TMP fibres). Interestingly, other results obtained during these test series also suggest that the fines present in TMP sheets may contribute to activation more than what has been assumed previously.

Conclusions

Straining of test sheets before or during drying has a clear effect on both the in-plane and the z-directional strength properties of paper. Based on the results of these test series, certain observations on bonding can be made. First, the essential phenomena for bond formation seem to take place at a solids content somewhere under 50 %, independent of the sheet composition. After some kind of boundary solids content, the bonds reach their final structure and cannot be repaired if they are damaged, even if the drying stress is removed from the network. Second, factors such as internal and external fibrillation, fines, fibre swelling, increase in flexibility and strength-increasing polymers contribute highly to the initial bonding potential of fibres. The benefits brought by beating, fines or other variables affecting directly the structure and strength of bonds seem to be extensively affected by drying stress.

Fibre segment activation is also affected by fibre properties, but the drying strategy plays a significant role for its development. In most sheets, activation increases linearly with increasing drying stress. Beating of kraft pulp does not seem to have much influence on the increment of activation, even though the fibres should have different swelling properties due to their different beating levels. In TMP sheets, the overall extent of activation is rather small, and in the kraft pulp and TMP mixture sheets, the properties of kraft fibres seem to govern activation.

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Continues from page 339

The influence of pigment size distribution and morphology on coating binder migration

This reduction in fine material seems to explain the reduction of gloss when the coating is applied to the absorbent substrate.

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