

# LIGNIN INTERDIFFUSION – A MECHANISM BEHIND IMPROVED WET STRENGTH

*Tove Joelsson*\*<sup>1,2</sup>, *Amanda Mattsson*<sup>1</sup>, *Jukka A. Ketoja*<sup>1,3</sup>,  
*Gunilla Pettersson*<sup>1</sup>, and *Per Engstrand*<sup>1</sup>

<sup>1</sup> Mid Sweden University, SE-85170 Sundsvall, Sweden

<sup>2</sup> MoRe Research Örnköldsvik AB, Box 70, SE-89122 Örnköldsvik, Sweden

<sup>3</sup> VTT Technical Research Centre of Finland Ltd, P. O. Box 1000,  
Espoo, FI-02044 VTT, Finland

## ABSTRACT

We have studied ways of improving strength properties of paper made from high yield pulps and lignin-rich chemical pulps by utilizing the thermoplastic properties of the lignin present in the fibre walls. Both dry and wet strength can be improved by hot pressing of sheets made from lignin-rich pulps. In this paper, we focus on aspects of the wet-strength development as a function of lignin content and temperature. Here we apply an activation energy evaluation approach to study lignin intermixing or interdiffusion. By means of hot pressing, it is possible to reach wet strength levels up to 50% of the dry strength level, provided that we use pulps with high enough lignin content. Our study included hot pressing of high yield pulps such as thermomechanical pulp (TMP), chemithermomechanical pulp (CTMP), high-temperature chemithermomechanical pulps (HTCTMP), unbleached northern softwood kraft (NSK) and northern bleached softwood kraft (NBSK). The sheet pressing trials were performed for varied temperatures from room temperature up to 270°C. As the activation energy for the high yield pulps and the lignin-rich NSK were all in the range of 20-32 kJ/mol, we

\* Corresponding Author

suggest that the wet strength development as function of temperature has a similar mechanism as long as the pulp fibres contain enough lignin. We also suggest that the phenomenon involves intermixing and/or interdiffusion of wood polymers between adjacent fibres when they are in a close contact. Most probably both the amorphous wood polymers, i.e. the linear hemicelluloses and the cross-linked lignin, mix with each other across the fibre-fibre or even more probable over the fibril-fibril contact surface. While the hemicellulose can intermix already at room temperature under moist conditions, the lignin intermixes more easily at the higher temperature we use. We do not know how far the hemicellulose or lignin could move within the fibre walls, but it seems that the amount of lignin present on the fibre surfaces plays an important role.

## **INTRODUCTION**

Improved wet bonding properties by means of heat treatments has been used for the paperboard and in the wood industry. It has been concluded that the thermo-plastic properties of lignin are the most important factors for establishing inter-fibre bonds that are strong also under wet conditions when applying hot pressing to material containing lignocellulose [1-5]. However, the role and the redistribution of lignin in the fibre network as well as the effect of chemical changes of the wood components is still an open issue.

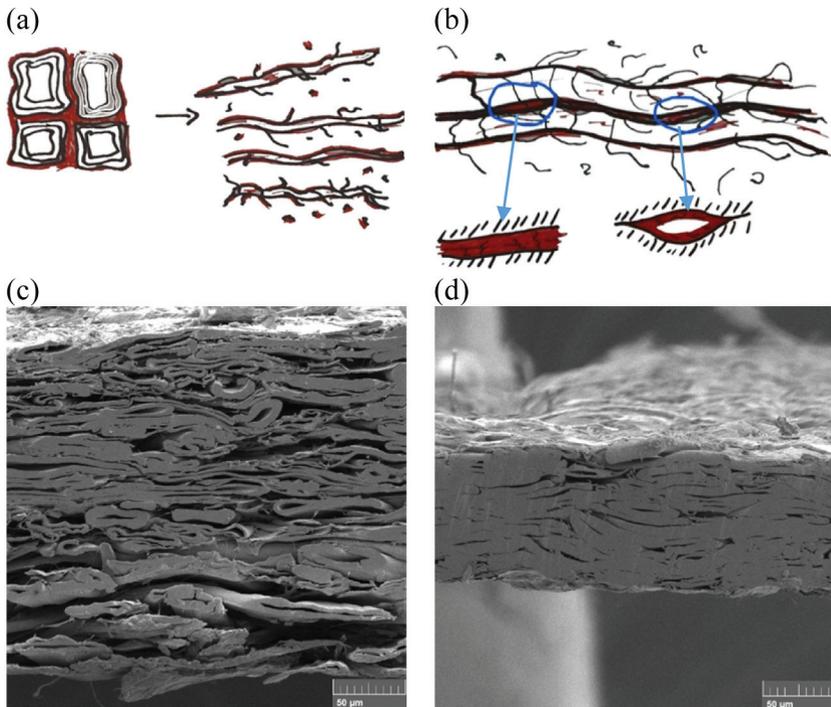
Some mechanisms that could explain the improved wet strength when consolidating paper webs at elevated temperature have been suggested earlier, such as a flow type of behaviour of lignin and increased number of crosslinks [6-8]. Goring and Byrd proposed that the hemicellulose is responsible for improved adhesion and dry strength development, while lignin acts as a protecting agent for already existing bonds whose wet strength improves when the lignin starts to flow [9,10]. However, these studies were performed at temperatures below 200°C. The higher softening temperature of lignin is restricting the flow of lignin compared to the corresponding flow of hemicellulose. The softening temperature of the wood polymers was studied by Goring [7] and Salmén and Olsson [11] among many other researchers. Chemical changes like reduced crosslinking during pulping or increased crosslinking during pulping or papermaking occur depending on process conditions such as; temperature, pressing/drying time and moisture content. Hemicelluloses undergo hydrolysis upon heating under moist conditions, and the extent of reaction depends on temperature, moisture content, and time. Formation of furfural and furan moieties from the decomposition of hemicelluloses has been suggested. Further reactions such as furfural self-polymerization, lignin-furfural cross-linking, and condensation reactions in lignin have also been proposed. [12].

Increased physical contact between fibres enabling entanglement or mixing between lignin from two fibres at high enough temperature which could contribute to improved wet strength (Figure 1). At high temperature it is possible that softened wood polymers move to such an extent that they start to mix or diffuse. Mechanisms as interdiffusion or intermixing require mobility and miscibility of the surface layers, and they have been described by McKenzie [13]. We would therefore suggest that at high enough temperature and pressure, lignin polymers at surfaces of contacting fibres start to intermix or interdiffuse so that a homogeneous mix of lignin from two adjacent surfaces is created. In this way, either new fibre-fibre bonds are created or existing inter-fibre bonds consisting of intermixed hemicelluloses become protected by lignin.

Norberg and Back [8] showed by the end of the 1960s how increased wet strength was obtained in the temperature range of 150-290°C for hot pressing of Asplund hardboard, based on coarse TMP (CSF 785 ml) pulp. It was also pointed out that dry lignocellulose materials softened in the range of 160-240°C, and that cross-linking and degradation reactions occurred above 170°C in the cellulosic material within the time-span studied. Swenson's patent from 1987 [14] describes the advantage of the heat treatment of linerboard based on kraft pulp. Also, Back and Andersson, and Saidan studied the benefits of the thermal treatment of unbleached kraft pulp papers [15, 16].

Increased degree of sulphonation of low-energy high-temperature chemimechanical pulp (HTCTMP) has been shown to significantly improve the densification of moist paper sheets and thereby the strength development, but the wet strength is not influenced [17]. The type and shape of fibres and fines have a quite large influence on both dry and wet strength. Increased fines content has been shown to contribute clearly to the paper strength [18-20]. Fines from high yield pulps normally contain higher amounts of lignin than the rest of the pulp, which is an advantage for wet strength development. Coarse and thick-walled fibres with high shives content have shown poorer bonding capacity that was significantly improved after hot pressing [21].

Increasing lignin content in kraft pulp increases wet strength linearly [22] up to a level that is well above the wet strength of a high yield (mechanical) pulp where the lignin content is much higher. It is possible that this is related to the type of lignin and also to the way how the lignin is distributed within the fibres. Most probably kraft pulp fibres have much higher content of lignin on their surfaces than high yield pulp fibres. A surface lignin coverage in the range of 66-70% for a kraft pulp with a total lignin content of 12% is reported when calculated from ESCA data published by Carlsson et al. [23]. The surface coverage of lignin for a variety of high yield pulps has been reported to be in the range of 35-40% [24]. It is possible that the preferable location of remaining lignin of kraft fibres at their surfaces, together with their higher flexibility and characteristic strength, explains



**Figure 1.** Schematic illustration of the postulated mechanisms of physical entanglement/mixing and interdiffusion/intermixing and comparison with SEM imaging: (a) Separation of fibres from a wood chip. (b) Two adjacent fibres partly coated with lignin and with fibrills and fines, where lignin intermixing may take place if the contact is close enough. (c) SEM image of pressed CTMP sheet without heat treatment. (d) SEM image of hot pressed (at 270°C) CTMP where both type of inter-fibre regions, as visualized in the schematic of figure b, can be observed. The sheet density of 1000 kg/m<sup>3</sup> as obtained using X-ray tomography [26] was higher than the one obtained with standard thickness measurement (ISO 534 2005) because of the contribution of paper roughness to the latter.

the difference in wet-strength development between lignin-rich kraft fibres and high yield pulp fibres.

In the present study, we compare different pulps based on calculations of activation energy, which describes the assumed lignin movement through interdiffusion as an explanation for improved bonding between wet fibres. Interestingly, wet strength developed in the same temperature range for many different pulp grades and independently of chemical modification of lignin.

## **EXPERIMENTAL PROCEDURE**

### **Materials**

A wide range of pulps with different lignin contents were used for sheet making including TMP, CTMP, HTCTMP, NSK, and NBSK [21]. The pulp properties were characterised with standard methods such as Canadian standard freeness measurement (CSF, ISO 5267-1,-2) and the Klason method (T222-88) for the lignin content.

Never-dried TMP was produced from Norway spruce (*Picea abies*) by preheating the chips at 115–150°C in a pressurized system and refining the unbleached pulp to a freeness of 115 ml. In addition to this normal pulp, we also studied accept and reject fibre fractions, obtained with a 50-mesh wire (where the fine fractions were removed) from TMP with freeness of 80 ml. The lignin content of these pulps varied in the range of 24–28%.

In producing the CTMP pulp, the spruce chips were impregnated with 20 kg/bdt solution of sodium sulphite ( $\text{Na}_2\text{SO}_3$ ), followed up peroxide bleaching and flash drying. This led to lower fines and shive contents than for the TMP. The study included two different freeness levels, 400 ml and 420 ml. The lignin content of 26–27% was similar as for the TMP.

The HTCTMP pulps were produced in laboratory scale from spruce, and they had three different degrees of sulphonation: low sulphite (LS, 25 kg/bdt, CSF 661 ml), medium sulphite (MS, 50 kg/bdt, CSF 724 ml), and high sulphite (HS, 120 kg/bdt, CSF 738 ml). These pulps were refined with very low energy consumption, which led to high fibre coarseness. The pulps were mixed with 20% of a bleached kraft pulp to achieve sufficient sheet strength.

The NSK was industrially produced kraft liner pulp. The chips were cooked according to the chemical sulphate process so that a kappa level of 80 (lignin content of 12%) was achieved. This pulp was compared with the never-dried NBSK obtained as a standard mix of pine and spruce pulps with almost vanishing lignin content.

### **Hot Pressing**

We performed hot pressing trials for standard laboratory sheets produced in the Rapid Köthen (RK) sheet former (Paper Testing Instruments, Pettenbach, Austria). Here the ISO 5269-2 2004 standard was followed, except that the target solids content was in the range of 50–65%.

The wet sheets were hot pressed at a series of conditions using two different hot-pressing machines [21]. The first one was an oil-heated cylinder press, which was used for temperatures  $\leq 200^\circ\text{C}$  [25]. Sheets were fed into the press on a felted

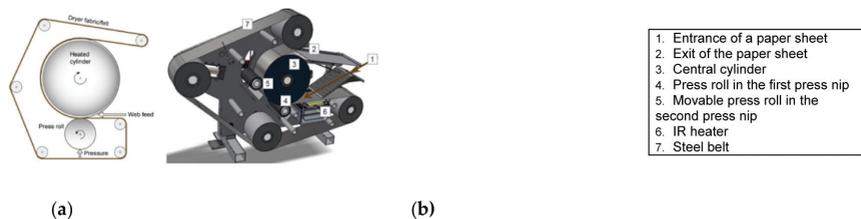
fabric with a rate of 1 m/min, and a nip pressure of 6 MPa was applied. The pressing time in the nip was 1500 ms (at a nip length of about 25 mm), and the after-hold was 70 s. The temperature of the cylinder surface was recorded using a surface contact sensor that was built in the machine. The pressing time was long enough so that the sheets dried completely. Moreover, their temperature was expected to be close to the cylinder temperature immediately after the nip.

Hot pressing at temperatures higher than 200°C was performed using a test pilot press with an infrared-heated steel belt carrying the paper samples through a nip [25]. The speed in this case was 3 m/min, corresponding to a pressing time of 40 ms (nip length was about 2 mm), and the after-hold was 23.5 s. The nip load was estimated to be 8 MPa, and the press load of the steel belt was 0.15 MPa. The temperature was measured with a surface temperature sensor (SWT316 Swema) from the cylinder surface after pressing (a distance 1.73 m from the nip).

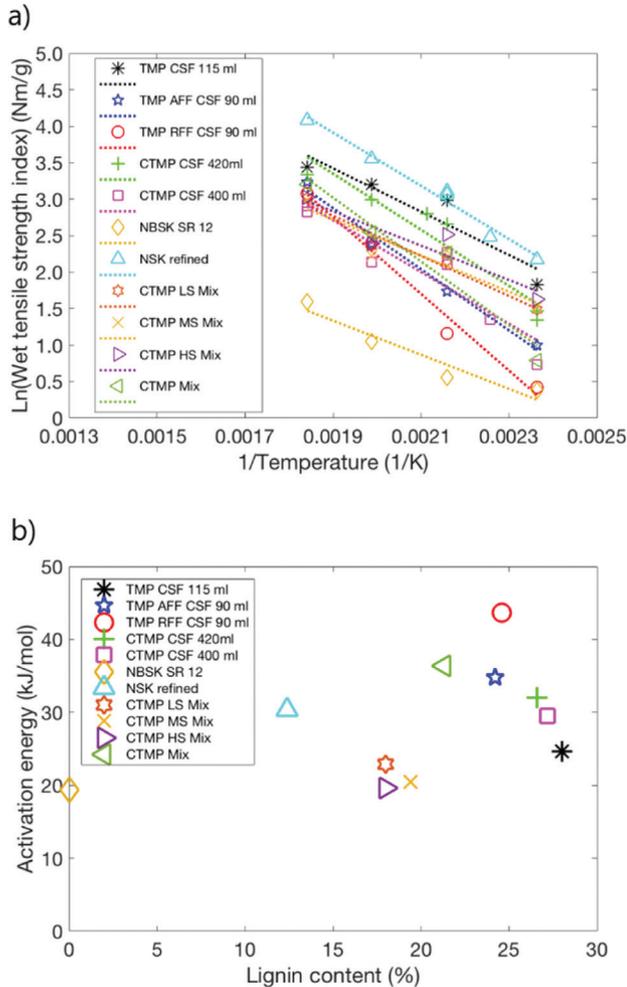
For both types of machines, nip lengths were measured with sensor films (Fuji, Prescale LW) suitable for pressures of 2.5-10 MPa. Thin blotter papers were used on both sides of an actual paper sample in all tests to prevent sticking. The solids content of paper sheets was 50-65% before pressing.

## RESULTS AND DISCUSSION

The interdiffusion is expected to be proportional to  $\exp(-Ea / RT)$ , where  $Ea$  is the activation energy,  $T$  is temperature, and  $R$  is the gas constant. We obtained  $Ea/R$  by plotting  $\ln(\text{Wet tensile strength index})$  vs.  $1/T$  and taking the slope of the linear fitting line in the temperature interval of 150-270°C (Figure 3a). One should notice that this temperature range includes data from both the cylinder press (Figure 2a) and the press with heated steel belt (Figure 2b) with different nip dwell times. Still, a single line is able to describe both cases in the operated interval for any pulp used in the data set of the present study. This means that the



**Figure 2.** (a) Felted, oil-heated cylinder press operated at MoRe Research (Örnsköldsvik, Sweden). (b) Infrared-heated press based on a steel belt produced by Ipco AB (Sandviken, Sweden). The picture is reproduced from ref. [26] under the terms of the CC BY licence.



**Figure 3. (a)** The logarithm of wet tensile strength index plotted against  $1/\text{Temperature}$  when pressed with either cylinder press ( $T=150^{\circ}\text{C}$ ,  $190^{\circ}\text{C}$ ; 6 MPa) or steel-belt press ( $T=230^{\circ}\text{C}$ ,  $270^{\circ}\text{C}$ ; 8 MPa): Varied pulps and pressing conditions for standard laboratory sheets with uniform fibre orientation. Solids content varies in the range of 50–65%. The overall trend is described by a similar activation energy of  $26 \text{ kJ mol}^{-1}$ . **(b)** Activation energies obtained from (a) as a function of their lignin content.

exponential temperature dependence of lignin diffusion rate dominates over other factors, such as pressing time or compression force (in our trials, even a rather low pressure was sufficient for increasing the wet strength).

We also found that a minimum of lignin content of 7% was required to raise the wet strength to a similar level as for samples containing more lignin [26]. On the other hand, lignin content above 12% did not seem to improve wet strength further (Figure 2b). In summary, the relative changes in wet strength with temperature were similar for very different pulps with the minimum of lignin content of 7-12%. Possibly, the amount of surface lignin in fibres is relatively high, and the lignin inside the fibre walls do not significantly move towards the fibre surface. Thus, the lignin at the interface of fibres would give the prominent contribution to wet strength.

The average activation energy of all pulps was  $E_a/R \approx 3080$  K, i.e.,  $E_a \approx 26$  kJ mol<sup>-1</sup>. However, there was some variation among the values obtained for different pulps as shown in Table 1. The activation energy values for TMP, CTMP and NSK range from 25 to 32 kJ/mol, while the lignin-free bleached kraft pulp ends up at 19 kJ/mol. This could be interpreted so that intermixing or interdiffusion works in the same way regardless of lignin content as long as there is enough lignin on the fibre surfaces. It is more difficult to understand what happens when we use fines-free reject and accept hydrocyclone fractions made from a TMP that had 90 ml CSF before the fractionation. The reject fraction, RFF, contains a higher proportion of stiff thick-walled summerwood fibres, and the accept frac-

**Table 1.** Activation energies calculated based on the development of wet-strength  $\rho_{wet}$  as function of temperature T during hot pressing of paper sheets made from different pulp types. The last column shows the logarithm of wet strength extrapolated to  $1/T=0$  using the linear fits of Figure 3a.

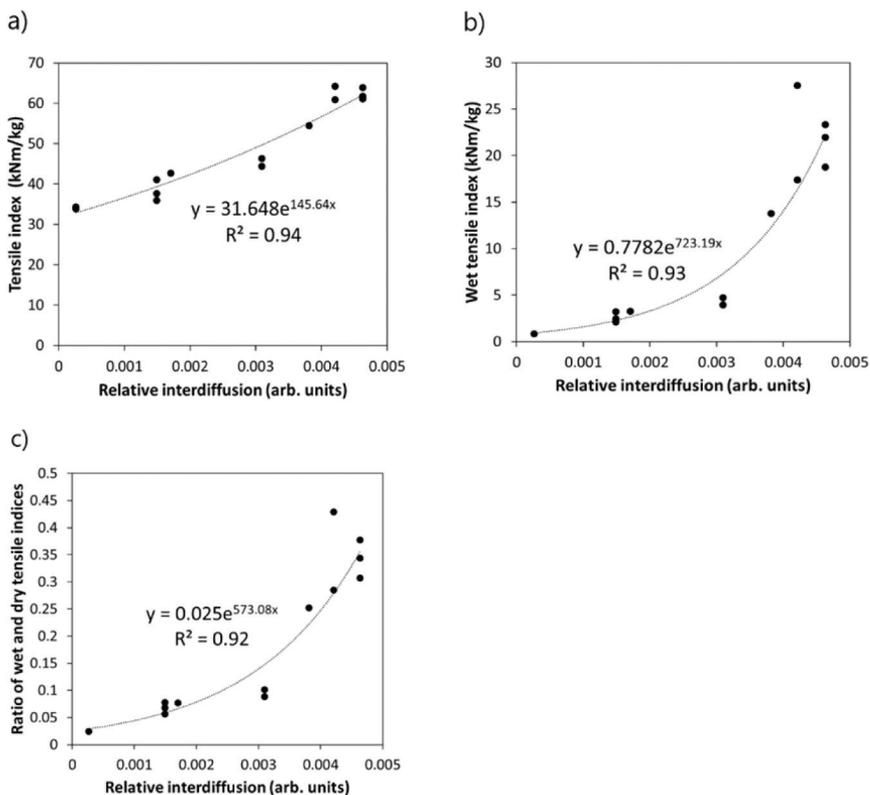
|                    | Lignin content (%) | Activation energy (kJ/mol) | 1/T = 0 limit of log $\rho_{wet}$ |
|--------------------|--------------------|----------------------------|-----------------------------------|
| TMP (CSF 115ml)    | 28.0               | 24.7                       | 9.1                               |
| TMP AFF (CSF 90ml) | 24.2               | 34.8                       | 10.8                              |
| TMP RFF (CSF 90ml) | 24.6               | 43.7                       | 12.8                              |
| CTMP (CSF 420ml)   | 26.6               | 32.0                       | 10.7                              |
| CTMP (CSF 400ml)   | 27.2               | 29.5                       | 9.5                               |
| NBSK (SR 12)       | 0.0                | 19.4                       | 5.8                               |
| NSK refined        | 12.4               | 30.4                       | 10.9                              |
| * HTCTMP LS Mix    | 18.0               | 22.9                       | 8.0                               |
| * HTCTMP MS Mix    | 19.4               | 20.5                       | 7.4                               |
| * HTCTMP HS Mix    | 18.0               | 19.6                       | 7.3                               |
| * CTMP Mix         | 21.3               | 36.4                       | 11.3                              |

\* HTCTMP LS, MS, and HS and CTMP (CSF 400 ml) all mixed with 20% NBSK SR 12.

tion, AFF, is dominated by thin-walled more compressible springwood fibres. This would influence the densification as well so that wet strength does not only depend on how lignin polymers intermix or interdiffuse but also on the physical structure of a paper sheet. This structure is to a great extent influenced by the shape and conformability of the fibres, which becomes obvious when studying hydrocylcone fractions as in this case. The fact that both fractions have higher activation energies than a TMP containing all fractions including fines could be caused by the fact that the lignin-rich fines fraction of a typical TMP becomes easily conformable when hot pressed.

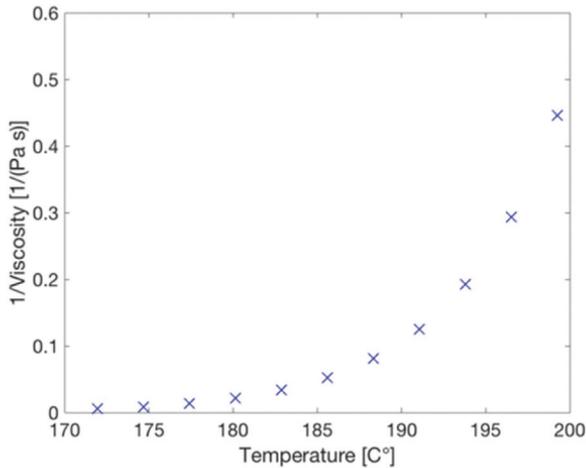
With increased sulphite charge, the refining energy to equal shives content of HTCTMP was reduced, the degree of sulphonation was increased, and the dry strength was improved, whereas wet strength was not affected [17]. Here we studied how the activation energy developed for these HTCTMP samples when mixed with 20% NBSK, and we called these samples HTCTMP mix (Figure 3b). The sample with the highest sulphite charge showed the lowest activation energy, 19 kJ/mol, and the sample with lowest sulphite charge showed a slightly higher activation energy of 23 kJ/mol, whereas the conventional CTMP mix had a value of 36 kJ/mol. The conventional CTMP had the highest wet strength, 28.2 Nm/g at 270°, while this CTMP mixed with NBSK had the strength value of 24.6 Nm/g at equal pressing temperature. The wet strength for the three mixed HTCTMP-LS, MS and HS furnishes was almost equal, 20.5, 19.8, and 20.7 Nm/g. The NBSK shows a similar activation energy, 19.4 kJ/mol, but as there is no lignin in this pulp, the corresponding activation energy is most probably connected to changes in the hemicelluloses. This also leads to a discussion related to the combined functionalities of lignin and the hemicelluloses on the fibre surfaces. When we try to estimate the activation energy related to wet strength development, we have to consider that we are probably evaluating the influence of the mix of both the amorphous wood polymers, i.e. the linear hemicelluloses as well as the cross-linked lignin. An interesting observation is that even when the wet strength level of sheet made from unrefined NBSK is only 4.9 Nm/g at 270°C, it has actually increased from the level of 2.9 Nm/g at 230°C. At the same time, the dry strength decreased from 44 Nm/g at 230°C to 31 Nm/g at 270°C.

The *interdiffusion or intermixing of lignin* macromolecules for increasing pressing temperature was studied more closely for the HTCTMP pulp with varied pressing conditions. In Figure 4 we plot both dry and wet tensile index and their ratio as a function of the factor  $e^{-E_a/(RT)}$ , which describes changes in the relative diffusion rate for varied temperature. Interestingly, both dry and wet tensile index improve exponentially with the interdiffusion but the dependence is stronger for the wet tensile index. The same holds for their ratio as seen from the last plot. Here we have used the average activation energy as determined for all other pulps also for HTCTMP (Figure 4).



**Figure 4.** Tensile index (a), wet tensile index (b) and their ratio (c) for HTCTMP as a function of relative interdiffusion rate as determined by  $e^{-E_a/(RT)}$  for varied temperature ( $E_a = 26 \text{ kJ mol}^{-1}$ ).

The higher diffusion rate with increased temperature enables the redistribution of lignin between fibres surfaces that are in close contact with each other within a fibre network without deteriorating other fibre polymers such as hemicelluloses. This behaviour is somewhat similar to earlier findings on viscosity of different types of extracted lignins with varied temperature. The decay in lignin viscosity is illustrated in Figure 5 using the values obtained by Sun et al. (2016) for extracted poplar lignin [27]. A drastic decrease in viscosity (plotted as  $1/\text{Viscosity}$  in the figure) takes place around the same temperature interval where we observe a rapid increase in wet strength. However, the activation energy underlying the viscosity decay of extracted poplar lignin was 240-280 kJ/mol [27], which is clearly higher than the values obtained for different pulps in Table 1. The latter values are in fact



**Figure 5.** 1/Viscosity as a function of temperature for extracted lignin according to data given in ref. [27].

quite close to the value 29 kJ/mol of dissolved lignin from the interior of the chip to the bulk liquor during kraft pulping of eucalyptus [28]. However, this feature may be just accidental because the properties of extracted organosolv lignin could be very far away from the properties of in-situ lignin that is still in direct contact (intermixed) with hemicelluloses and cellulose within the fibrils the surfaces of high yield pulp or lignin rich kraft pulp fibres.

In the kraft process, we have a very high concentration of sulfide and hydroxyl ions that reduce degree of crosslinking, so lignin can be transported from the fibre wall into the lumen and then through the pores successively out from the wood chip to the surrounding liquid at a liquor to wood ratio of 4:1. The transport phenomena in kraft pulping were recently described by Brännvall and Rönnols [29]. In hot pressing, we have a system with pH 5-7, i.e. no sulfide or hydroxyl ions, and a liquor to wood fibre ratio of between 1:1 and 0.5:1. Most probably, it is not possible for the lignin situated in a fibre wall to move out to the fibre surface even if the temperature is above 200°C. However, such a movement is not necessary as we already have enough lignin at the surface left as the last lignin by the kraft pulping process. In the case of high-yield pulp, the surface lignin content is connected to the conditions we use in the mechanical pulping process as well as to the type of wood we use.

The initial part of sulfonation in a CTMP process has an activation energy of 20 kJ/mol, while the bulk part has an activation energy of 60 kJ/mol [30]. This does not mean that the activation is directly connected to the yield loss in such a

process, because the yield is still >95% even at a very high degree of sulfonation when the lignin has reduced the degree of crosslinking and high charge. The lignin is softer but it does not move out of the fibres. If the lignin would move easily from inside the fibre wall to the surface, then the CTMP fibres containing 26% lignin would result in higher wet strength than the NSK fibres containing 12% lignin when compared at equal sheet density.

In summary, the definition of the optimal hot-pressing interval for maximized wet strength without reducing dry strength could be found by taking into account the temperature – diffusion rate relationship for softened lignin within the fibre wall.

## CONCLUSIONS

While studying ways to improve strength properties of paper made from high yield pulps and lignin-rich chemical pulps, we have tested an approach where we use activation energy calculations to evaluate aspects of lignin intermixing or interdiffusion. We have shown that hot pressing enables wet strength levels up to 50% of the dry strength level if the pulp fibres contain a high enough amount of lignin. The following pulps were studied by means of activation energy analysis; thermomechanical pulp (TMP), chemithermomechanical pulp (CTMP) and high temperature chemithermomechanical pulp (HTCTMP), unbleached northern kraft (NSK) and northern bleached softwood kraft (NBSK).

The interdiffusion or intermixing mechanisms related to lignin seem surprisingly similar for lignin-rich kraft and high-yield pulps, with similar level of activation energies. For that reason, temperature is the most dominant parameter for the development of wet strength in hot pressing. There are some differences how dry and wet strength develop, both related to differences in fibre wall stiffness between kraft and high-yield pulps and because of the relative importance of the hemicelluloses.

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# LIGNIN INTERDIFFUSION – A MECHANISM BEHIND IMPROVED WET STRENGTH

*Tove Joelsson*<sup>1,2</sup>, *Amanda Mattsson*<sup>1</sup>, *Jukka A. Ketoja*<sup>1,3</sup>,  
*Gunilla Pettersson*<sup>1</sup>, and *Per Engstrand*<sup>1</sup>

<sup>1</sup> Mid Sweden University, SE-85170 Sundsvall, Sweden

<sup>2</sup> MoRe Research Örnköldsvik AB, Box 70, SE-89122 Örnköldsvik, Sweden

<sup>3</sup> VTT Technical Research Centre of Finland Ltd, P.O. Box 1000, Espoo,  
FI-02044 VTT, Finland

*Thomas Harter*      Graz University of Technology

I have a question regarding your slide 19 about the wet strength test at different times. First about the methodology: Have you kept the samples in a wet condition during this time or have you tested it after one minute, one hour and 24 hours, according to standard procedure? Do you have any idea what's going on here? Why is there a decline in wet strength over the time?

*Tove Joelsson*

It is likely that we have an increased number of covalent bonds and cross-links, which resist water. On the other hand, there are certainly still bonds in the structure that are susceptible to water, which then break when water penetrates.

*Thomas Harter*

After 24 hours, was this the last measurement you did? Or did you also check it after a longer time period?

## *Discussion*

*Tove Joelsson*

No, but I have had samples in a glass in my kitchen for months. And I could still feel the strength.

*Hak Lae Lee*      Seoul National University

I am very interested in your research. When a moist paper is pressed at high temperatures, the moisture turns into water vapour. When you release the pressure at the exit of the press, the moisture turns into steam. This may cause sheet delamination. We experienced sheet delamination from time to time when drying wet sheets in Condebelt drying, which is very similar to a hot pressing system. Did you have any experience with such sheet delamination?

*Tove Joelsson*

The samples I have tested have mainly been high yield pulps of different types that have a fairly open structure. I think that is one reason I have not seen any delamination in these samples. I also want to show you this picture, slide 7. Here the sample is first subjected to a short and high nip pressure and then to a lower pressure (from the steel band or fabric) which continues for a longer time until the sample is dry and exits the pressing machine. I believe that during this time in strain at high temperature it is possible that the vapour pressure equalises and the curing (lignin structures and cross-linkages) and drying of the paper structure takes place. These can be reasons for not getting steam explosion, as has been the case in impulse technology where there were many problems with delamination.

*Babak Mirzaei*      West Rock – Richmond VA

The concept of using high temperature for bonding wood is not new, as you mentioned some patents were published by Mason in 1930s, which led to the development of the Masonite process for the wood products industry. The component creating these bonds is mostly lignin. So no surprise, it works better for paper products with higher lignin contents. But the process is mostly withdrawn by wood products industries, because it is not cost effective. At the end, the wood had to stay on the press for prolonged time at higher temperature and it ends up being more expensive than the conventional products using synthetic adhesives. So, the same question arises here. Is this process cost effective compared to the conventional process?

*Tove Joelsson*

This process does not exist commercially yet. This is only at the beginning of research and development. But I believe in this technology. When considering costs, one must consider the total process from pulping to final paper, including the costs of manufacturing the additional strengthening chemicals and the sources from which they are made. In addition, you have to consider the environmental impact of a paper package and compare it with the environmental impact of plastic (plastic breaks down in about 500 years!)

*Babak Mirzaei*

You don't use any wet strength adhesives here.

*Tove Joelsson*

No.

*Babak Mirzaei*

So, I would say that's the conventional process. If you don't use adhesive, you use high temperature for bonding instead. Is it more cost effective at the end compared to using some synthetic adhesives for better strength?

*Tove Joelsson*

I have not made a calculation on that. But of course, using high temperature will increase costs, on the other hand it will replace the additives and you get a stable wet strength. Additionally, we utilise almost the whole tree when using mechanical pulp where the yield is above 95%. It is an argument for this technology.

*Markus Biesalski*      Technical University of Darmstadt

I'd like to have your opinion about comparing repulpability of paper made with regular wet strength agents like PAE etc. in comparison to your materials. Can you compare this and look a little bit more into sustainability issues rather than economics?

## *Discussion*

*Tove Joelsson*

I have not yet done a comparison of commercial wet strength paper and these samples regarding repulpability. So, I can't tell the difference right now.

*Markus Biesalski*

You can talk about repulpability, and how easy can your material be repulped? Can you maybe find out this again?

*Tove Joelsson*

The repulpability of this material needs to be studied more. But I have done one first test to dissolve the hot-pressed paper and chose the one with the highest wet strength. The test was carried out in water at room temperature in a kitchen mixer for two minutes. I could see that the paper was repulped, but the quality of the fibres has not yet been analysed (slide 20).

*Markus Biesalski*

So, basically, you can use a high yield pulp, which can save a lot of trees and also you can produce the wet strength papers, but it is easier to repulp compared to the conventional wet strength agent used once.

*Tove Joelsson*

Additionally, depending on the application in mind, the very highest wet strength may not always be needed.

*Jukka Ketoja*      VTT Technical Research Centre of Finland Ltd

A really crucial question I would like to come back to is that of the cost efficiency. I believe the answer very much depends on the thickness of the material. And that might be one big difference between wood and paper. So, paper thickness is a part of the calculation as well as how long is the heat contact you need and what's the thermal diffusivity of paper and factors like that. So, there might be some paper thickness for which hot pressing works.

*Tove Joelsson*

Yes.

*Elias Retulainen*     Fiber and Fibril

You are talking about interdiffusion. I recall that Back – that you also referred to – in his studies explained the improved wet strength after heat treatment as a result of crosslinking. So, can you comment on that? Can there be other mechanisms other than the interdiffusion?

*Tove Joelsson*

As heard in today's discussions about interdiffusion, I think it is about molecular segments moving into each other, so that mixtures are created in the interface between the fibres. This can increase the possibilities for multiple crosslinks and reactions at high temperatures. One does not exclude the other – both can happen.