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Mechanical properties of woodfree paper sheets at different surface size starch amounts

Keywords
Woodfree, surface sizing, starch content, elastic modulus, bending stiffness

Abstract

This paper examines the effects of surface size starch addition on unsized and uncalendered paper sheets' mechanical properties. The more specific focus was to study such properties as elastic modulus, elongation at break, and bending stiffness as a function of sheet starch content. The study was based on vacuum-assisted impregnation of starch into the sheet structure and drying the sheets under an impingement hood. Differences in starch amounts were obtained by repeating this sequence, which resulted in a set of specimens with varied starch amounts. Applications with corresponding water amounts were also performed as references in order to remove the effect of water and the wetting-drying cycles from the results. The net effect of starch amounts on the studied sheet properties could then be investigated. Results suggest that the surface sizing starch promotes elastic modulus in cross direction, but not in machine direction. In increasing the elastic modulus – and further the bending stiffness of the sheet – starch may merely promote shrinkage potential, which then leads – when shrinkage is not allowed – to increased drying stress that, in turn, leads to an increase in elastic modulus and further to increased bending stiffness. However, when drying shrinkage is allowed, bending stiffness is increased despite a drop in elastic modulus. This is due to increased thickness, which compensates for the loss in elastic modulus. Therefore, the presence of starch affects bending stiffness in two ways: when drying is restricted, starch promotes increased elastic modulus through increased drying stress; with drying shrinkage allowed, bending stiffness increases as a result of increasing sheet thickness. Since a practical papermaking process involves a large variety of shrinkage behaviors in the cross direction, the mechanism behind bending stiffness can vary in the cross direction based on the shrinkage profile across the entire width of the web.

Tiivistelmä

Puuvapaan hienopaperin mekaaniset ominaisuudet pintaliimatärkkimäärän funktiona
Tässä työssä tutkittiin pintaliimatärkkelyksen vaikutusta pintaliimaamattomien ja kalanteroimattomien tuotantokonehienopaperiarkkien mekaanisiin ominaisuuksiin. Erityisesti tarkasteltiin tärkkipitoisuuden vaikutusta kimmomoduuliin, murtovenymään ja taivutusjäykkyyteen. Tutkimus suoritettiin alipaineavusteisena tärkkiliuoksen impregnointina hienopaperiarkkien läpi. Arkit kuivattiin kuumailmapuhalluksella alipaineistetun viiran tukemana kahdella eri kuivatuskutistuman rajoitustasolla. Eri tärkkimäärät tuotettiin arkkeihin toistamalla käsittely. Vertailunäytteitä tuotettiin käsittelemällä arkkeja vastaavilla vesimäärillä ja kostutus/kuivatussykleillä. Näin tärkin nettovaikutus voitiin saada selville ottamalla huomioon veden ja kuivatussyklien aikaansaamat vaikutukset paperiin.

Tuloksista voitiin päätellä, että tärkkelys parantaa paperin kimmomoduulia poikkisuunnassa, mutta ei konesuunnassa. Mekanismi selitettiin tärkkelyksen aikaansaamalla kutistumapotentiaalin kasvulla, joka on suurempi poikkisuunnassa. Kutistumaa rajoittava jälkikuivatus johtaa kasvaneisiin kuivatusjännityksiin; kuivatus korkeamman kuivatusjännityksen alaisena kasvattaa puolestaan paperin kimmomoduulia ja sitä kautta taivutusjäykkyyttä erityisesti poikkisuunnassa. Kuivatuskutistuman salliminen pienentää kimmomoduulia, mutta tämä kompensoituu kasvaneen paksuuden kautta taivutusjäykkyyden suhteen. Näin ollen tärkki vaikuttaisi taivutusjäykkyyteen kahdella tavalla riippuen jälkikuivatuksen kutistumaolosuhteista, jotka vaihtelevat paperinvalmistuksessa mm. kutistumaprofiilin kautta.

Introduction

The effect of starch on the properties of paper has been widely studied with respect to wet end starch /e.g. 1, 2, 3, 4, 5/. The emphasis with adding starch (either internal or surface sizing) is to increase such properties as water resistance, grease and solvent resistance, and surface strength /6/. The main role of surface size is to promote surface properties, e.g. strengthen the surface and bind particles, such as fibers and fillers, to the surface. Additionally, starch is expected to add internal strength to the sheet through the penetration of liquid in the z-direction /7/. The main advantage of surface sizing over internal sizing is the excellent retention of the starch solution /8/. At a flooded nip size press liquid pickup is mainly determined by the dryness of the paper and the level of internal sizing /9/. Küstermann /10/ and Felder /11/ studied surface sizing using a metered size press (MSP) with varied solids contents and aiming for a constant wet film amount, which resulted in differing starch amounts in the paper. They report effects on tensile strength and plybond strength, for example, as do Brogly and Harvey /12/ in their study in which starch pickup is varied at a constant (7%) solids content. Using surface sizing starch (and especially starch concentrating on the sheet surface through different surface sizing techniques or starch solids contents) is reported to increase the bending stiffness of paper /e.g. 13, 14 and 15/.

Information on the elastic modulus /e.g. 16/ of the sheet structure is used in the determination of the bending stiffness of a sheet. Generally, Lekhnistkii /17/ has expressed the bending stiffness theory for an anisotropic thin plate. However, we now present paper as a specific case of this theory as an orthotropic, isotropic and with the Poisson's ratio ν in all directions = 0. Then, a symmetrical layered structure where the elastic modulus of the surface plies is equal

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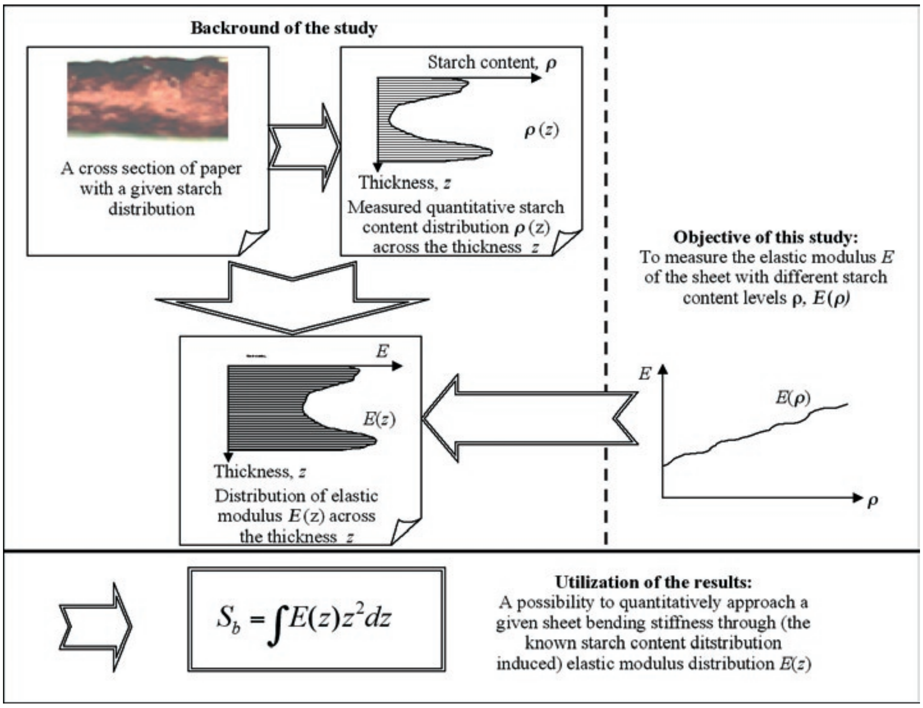

Fig. 1. Illustration of the background of determining the elastic modulus E as a function of starch content ρ, E(ρ).

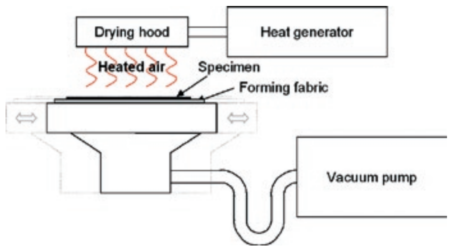

Fig. 2. Schematic picture of the surface sizing and drying arrangement used in the study.

to E_1 and that of the mid-ply to E_2, while the total sheet thickness equals d and the thickness of the mid-ply equals d_2 , bending stiffness S_b can be expressed as bending of a beam /18/:

$$S_b = \frac{E_2 d_2^{\,3}}{12} + \frac{E_1(d^3 + d_2^{\,3})}{12} \qquad (1)$$

Then, based on definition of bending stiffness mentioned in *Eq. 1*, elastic modulus should increase in the function of the starch content in z-direction, i.e. when concentrating starch on the surface (increasing the starch content of the surface layer). Generally, without assuming sheet symmetry or definitive thickness and elastic modulus properties for each ply, bending stiffness can be expressed in more general terms as /18/

$$S_b = \int E(z) z^2 dz \qquad (2)$$

A varied z-direction elastic modulus distribution (i.e. *E(z)*) can be used when determining the bending stiffness of a sheet. This would require studying the factors governing the elastic modulus of paper structure. Here, the effect of starch content on elastic modulus is of interest. A quantitative approach to looking into the behavior of bending stiffness S_b would then be possible when determining the z-directional starch distribution of a sheet. This approach is illustrated in detail in *Fig. 1*.

In our work the objective was to apply varied amounts of the surface sizing starch to sheets and to observe the resulting effect on the mechanical properties of the sheet. The aim was to apply starch evenly throughout the z-direction. Such parameters as the elastic modulus could then be determined at different starch contents. This information could be used to calculate the physical properties of mill-produced paper based on actual starch content distributions across the z-direction. Here, in determining the starch z-distribution in a surface sized sheet, a method presented by Lipponen et al /19/ can be used.

Experimental

Materials

Woodfree base paper sheets produced on a fine paper machine without surface sizing and with a basis weight of 89 g/m² were used for the study. The properties of the base paper are presented in *Table I.*

Table I. Properties of Base Paper.

Properties		Values
Grammage	g/m²	89
Bending stiffness, MD	mNm	0.64
Bending stiffness, CD	mNm	0.25
Elastic modulus, MD	N/mm²	7079
Elastic modulus, CD	N/mm²	3206

A modified potato-based starch was used in our study (Ciba Specialty Chemicals Inc., Raisio, Finland). A native potato starch was initially modified through oxidation with hypochlorite and the content of carboxylic groups in the oxidized starch was about 1% ($DS_{\text{-COOH}}$ = 0.031). The viscosity of this starch (Raisamyl 01020) was 20 mPas (Brookfield 100 rpm, 60 °C) at a concentration of 10%.

Methods

Surface Sizing of the Sheets

The surface sizing of the sheets was performed by applying a thin layer of 1% starch solution with a paint roller. Starch was applied to one side of the sheet and the sheet was then immediately placed on a forming fabric-supported vacuum table to impregnate the starch into the sheet. After 5 second impregnation the vacuum table was brought under an impingement hood for 30 seconds of drying, still maintaining the vacuum. The temperature, impingement air velocity, and moisture content of the impingement air were set to 150°C, 30 m/s and 50%, respectively. Two different vacuum levels were used in the study: First, a 3 kPa vacuum level was used allowing the sheet to shrink during the trial. Then, a higher vacuum (12 kPa) was used to restrict drying shrinkage. Here, the restriction of shrinkage was assisted by fastening the edges of the sheet to the fabric using aluminum tape.

Each starch application was performed on both sides of the sheet. The different total starch amounts were then obtained through sequences of multiple application and drying stages. As the application and

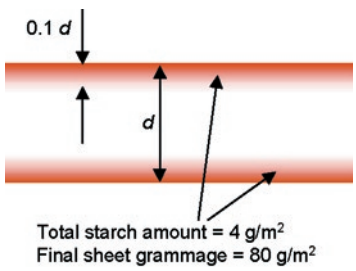

Fig. 3 .The effect of a schematic starch distribution in a commercial paper sheet on the starch content range studied.

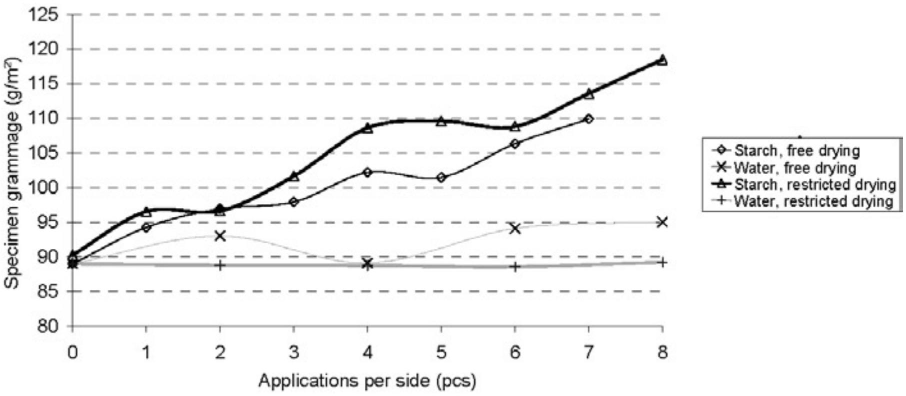

Fig. 4. Grammage of the sheet with different trial points of free and restricted drying with water and starch. The basis weight of the base sheet was 89 g/m².

drying sequences proceeded, the pores of the sheet were eventually filled with starch and no more starch could be impregnated into the paper. This level would then represent the maximum reachable starch content in the sheets and the sheets were considered saturated. *Fig. 2* illustrates the surface sizing and drying arrangement used.

The applied starch amount was measured from the starch impregnated samples. With paper samples prepared with water as a reference, paper samples were weighed right after the application of water to control the amount of water. Here, the same amount of water was targeted as in the 1% starch solution treatments. It was then possible to isolate the net effect of starch on the results.

A fairly broad range of different starch contents was selected for the study. The reason for this lies in the starch distributions achieved with different surface sizing techniques and starch solids contents /e.g. 13 and 14/, which sometimes result in starch concentrating intensively on the surface layer of the sheet. The applied starch amount of 2 g/m²/side will give an average starch content of 5% in the sheet with 80 g/m² standard copy paper. If we now assume that starch remains in the outermost 10% layer of the sheet on either side, starch content in the surface layer equals approximately 25%. This roughly determined the upper limit of the range of interest in terms of the starch content used in this work. This principle is illustrated in *Fig. 3*.

The starch amounts obtained in the study are presented in *Fig. 4*. One can observe that the starch content reached in the sheets varied from roughly 5% to more than 30% of starch. This gave a sufficient starch amount range for the study to consider the results representative of the highest starch contents in the top layer of a surface-sized sheet of paper.

In order to evaluate drying shrinkage during the procedure, parallel specimens were prepared with a millimeter grid photocopied on the sheets. The shrinkage of the sheets could then be measured through changes in the millimeter pattern.

Measurements

Paper properties were measured based on the standards reported in *Table II*. After performing the bending stiffness measurements the same individual strips were taken to elastic modulus measurement in order to reduce scatter and to improve the comparability of elastic modulus and bending stiffness results.

Table II. Measurement Methods and standards.

Methods	Standard
Grammage	SCAN-P 6:75
Starch amount	TAPPI 419 om-91
Bending stiffness	SCAN-P 64
Elastic Modulus, Young	SCAN-P 38:80

Results

The following figures are presented as a function of cumulative water addition, which was applied to the paper during the complete set of multiple application sequences in both 1% starch application and the water treatments performed for reference. The water reference results can thus be evaluated together and compared with the starch application results.

Shrinkage results are presented in *Fig. 5*. It can be noted that starch promotes strong shrinkage especially in the cross direction in unrestricted drying. However, some shrinkage can also be observed in the results based on "restricted" drying despite the attachment of the sheets to the fabric during drying. This is most probably due to the relaxation of drying stresses after sheets were released from tape attachment in the drying apparatus used, which resulted in some degree of shrinkage. Elastic modulus results both in the cross direction and machine direction are presented in *Figure 6*. Elastic modulus values with both starch and water treatments are presented in these figures for both restricted drying and free drying. One can observe in the figures that the modulus of elasticity decreases with less restrained drying, as reported by Setterholm and Kuenzi /16/ and Htun /20/, for example, who reported that the modulus of elasticity decreases significantly when drying shrinkage is allowed. One can suggest that starch-induced excessive shrinkage leads to a "crimped" sheet structure with very low

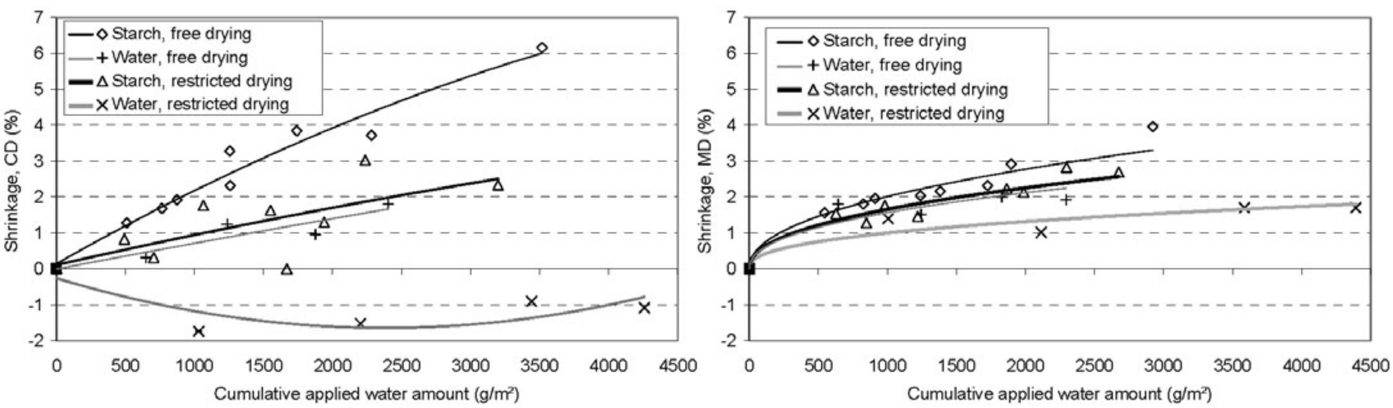

Fig. 5. Drying shrinkage in the cross direction (left) and in the machine direction (right). as a function of cumulative water addition. Starch added as 1% starch solution.

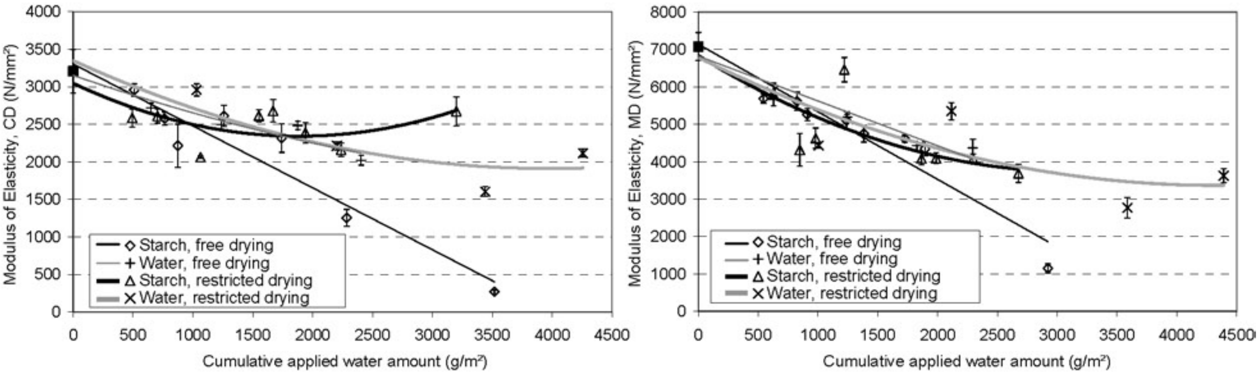

Fig. 6. Modulus of elasticity in the cross direction (left) and in the machine direction (right) as a function of cumulative water addition. Starch added as 1% starch solution.

resistance against tensile stress and therefore to low elastic modulus values in the specimen, as suggested by Silvy /21/. Water treatment decreased the elastic modulus of sheets as well, but not as drastically as starch. On the other hand, starch-treated samples developed a higher elastic modulus under restricted drying than water-treated samples under restricted drying: this resulted in a net increase in elastic modulus at higher starch amounts. This suggests that the prevention of starch-induced shrinkage resulted in a remarkable increase in drying tension – much as suggested by Silvy /21/ – and therefore a higher final elastic modulus. Drying tension has earlier been reported to increase the elastic modulus of fibers and that of the fiber network /e.g. 22, 20/. Drying stress has been shown to increase fiber stiffness through fiber segment activation /23/.

Different drying arrangements with starch and water treatments have an effect to the paper thickness, as seen in *Fig. 7*, where the results are prensented as a function of applications per side. Here, thickness seems to increase due to drying restriction. The phenomena is more evident with starch treated samples. The thickness increase may be explained through the known phenomena of thichness increase during wet straining: wet straining has explained to increase thickness of the sheet through fiber "undulations", when some fibers are pulled straight in the network /24/. Here, the effect of starch – through the suggested mechanism of increased drying stresses – may be inducing wet straining of the fibers and fiber bonds, resulting in promoting of increase of sheet thickness when starch is present with restricted drying conditions.

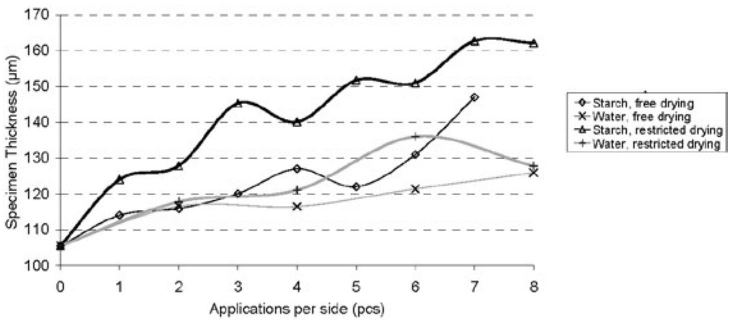

Fig. 7. Thickness of the sheet with different trial points of free and restricted drying with water and starch.

Breaking elongation (*Fig. 8*) in the machine and cross direction reflected the elastic modulus behavior as the highest breaking strain values were achieved with starch-treated free-dried samples in both directions. Starch application with restricted drying also produced decreasing breaking elongation as more starch was introduced to the sheet. Shrinkage during drying and elongation at rupture are well correlated as shrinkage resulted in the "crimped" sheet structure noted above, which allowed both a decrease in elastic modulus and an increase in breaking elongation as the crimped structure opened up during loading and elongation continued further before breaking. All of these results on the correlation of elastic properties and elongation at break agree well with previously published results, e.g. Gates and Kenworthy /25/, and Silvy /21/

The measured bending stiffness values of the specimens are presented in *Fig. 9*. Here, with added starch, the effect of the cubed sheet thickness ($S_b = E\,d^3 / 12$) can be seen to dominate the bending stiffness behavior of paper despite decreased elastic modulus with both drying arrangements.

Fig. 10 presents the net effect of starch

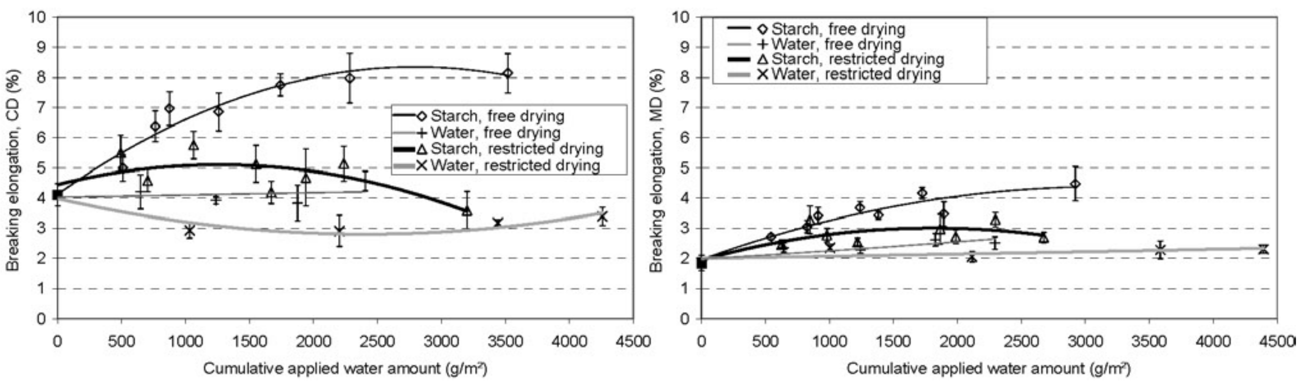

Fig. 8. Breaking elongation in the cross direction (left) and machine direction (right) as a function of cumulative water addition. Starch added as 1% starch solution.

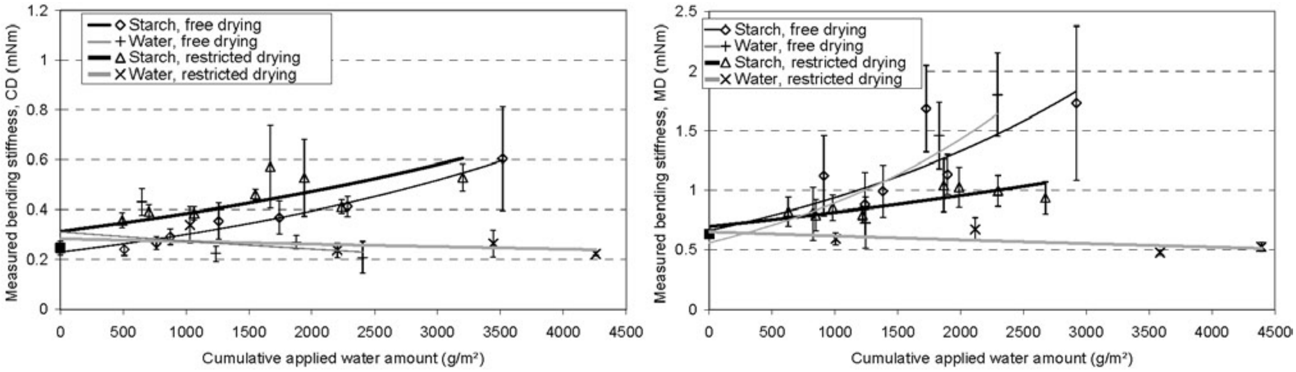

Fig. 9. Bending stiffness values in the cross direction (left) and machine direction (right). as a function of cumulative water addition. Starch added as 1% starch solution.

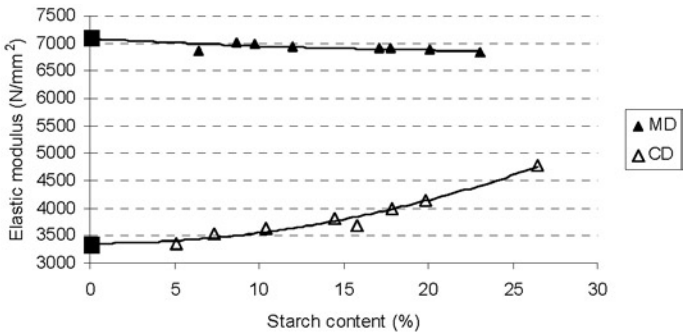

Fig. 10. Net elastic modulus effect of starch plotted as a function of starch content in the cross and machine direction.

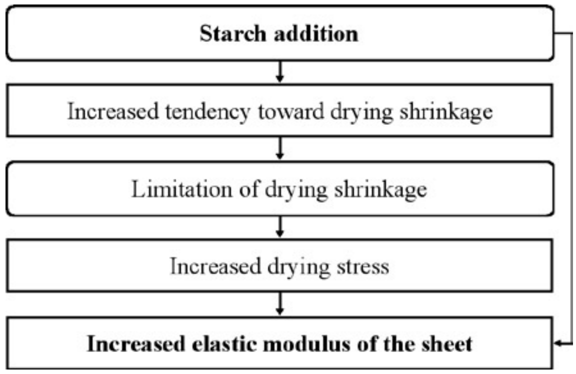

Fig. 11. Suggested mechanism for the development of the elastic modulus of a sheet with starch promoting shrinkage potential which – after drying with limited shrinkage – results in an increase in elastic modulus through increased drying stress.

as a function of starch content. The figure was produced by substituting corresponding water amounts in elastic modulus trend line equations for both starch and water-treated results (*Fig. 6*). The net effect of starch was then reduced to the difference between these values. The results were then plotted with starch content on the x-axis in *Fig. 10.* One can observe that starch increased the cross directional elastic modulus of sheets. However, machine direction elastic modulus was not positively affected by the presence of starch.

This observation can be supported by the findings of Lipponen *et al.* /15/ who reported a clear improvement in cross directional bending stiffness when they increased the starch solids content (and therefore concentrated the starch on the sheet surface). On the other hand, they noticed that the effect of starch remaining on the sheet surface is much less clear with respect to machine direction bending stiffness. This agrees well with the different CD and MD effects of starch on the elastic modulus of paper observed here.

Discussion

Measurement results were reported in this paper for different starch amounts in uncoated woodfree base paper sheets. These results showed two different starch application-induced behaviors regarding the elastic modulus of the sheet:

1) Starch application with low drying restriction (shrinkage allowed) resulted in a decrease in the elastic modulus of the sheet both in the cross and in machine direction of the sheet.
2) Starch application with highly restricted drying (shrinkage limited) resulted in an increase in the (water reference corrected) net elastic modulus of the sheet, especially in the cross direction where an increase in elastic modulus compared to parallel water additions was noticed. However, an increase in the machine direction elastic modulus of the sheet did not occur compared to water treatment.

The effect of restricted drying together with starch addition increasing the elastic modulus of the sheet suggested that the prevention of starch-induced shrinkage resulted in remarkably increased drying tension in the sheet – and therefore higher final elastic modulus. Drying tension and stress has earlier been reported /e.g. 20/ to increase the elastic modulus of fibers and the fiber network, as well as fiber stiffness, through fiber segment activation /23/. The explanation of starch inducing increased drying stresses was supported by thickening of the starch treated sheet with drying restriction compared to freely dried specimens. Therefore, starch may not – at least to the extent that has been believed – directly increase the elastic modulus of the sheet through fiber/fiber or starch/cellulose molecule bonding, i.e. as the general term "binder" suggests. Also, wet straining has explained to increase thickness of the sheet through fiber "undulations", when some fibers are pulled straight in the network /24/. In increasing the elastic modulus – and further the bending stiffness of the sheet – starch may merely promote shrinkage potential instead, which then leads – when shrinkage is not allowed – to increased drying stress. This, in turn, leads to an increase in elastic modulus, as described in related literature. This mechanism is illustrated in *Fig. 11.*

However, allowing shrinkage may lead to higher thickness of the sheet, which again increases the bending stiffness of the sheet by the third power of thickness. This will compensate the decrease in elastic modulus associated with the shrinkage of the sheet mentioned earlier. The suggested mechanism is illustrated in *Fig. 12*. The actual development of bending stiffness is most probably a combination of both mechanisms, which can also vary in an actual pa-

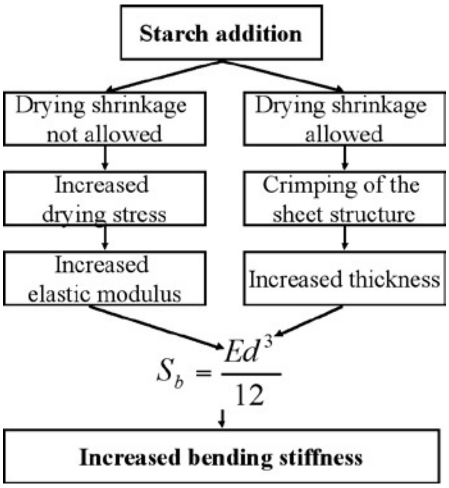

Fig. 12. Suggested mechanism for the role of starch in the development of the bending stiffness of a sheet.

per making process in the CD direction as the shrinkage profile of the sheet is seldom flat in the cross direction of the paper machine.

Results also suggested that the presence of starch increased the elastic modulus of the sheet only in the cross direction of paper but not in the machine direction. This behavior can also be explained through the shrinkage prevention and drying stress approach described earlier. If we remember that individual fibers experience greater shrinkage in the transverse direction than in the longitudinal direction /e.g. 26/, one can state that drying stress (due to prevented drying shrinkage) is potentially greater in the cross direction as fibers are more oriented in the machine direction. In the machine direction, however, where fibers tend to shrink less, the drying stress levels do not get as high. Starch-induced elastic modulus development due the presence of starch is then much less distinct in the machine direction than in the cross direction. It can also be suggested, that the elastic modulus of the fiber nework in the machine direction – the direction in which most of the fibers are oriented – can not be increased through starch addition to a great extent since the elastic modulus can be argued to be determined through the elasticity of the fibers itself.

Concluding remarks

Our paper presents a novel interpretation of the role of surface sizing starch in terms of the development of elastic modulus and bending stiffness. In increasing the elastic modulus – and further the bending stiffness of the sheet – starch may merely promote shrinkage potential, which then leads – when shrinkage is not allowed – to increased drying stress that, in turn, leads to an increase in elastic modulus and further to increased bending stiffness. Furthermore – when concentrating surface sizing starch on the sheet surface – one can explain this increase in bending stiffness through the buildup of a higher elastic modulus in the topmost surface layer of the sheet caused by the prevention of drying shrinkage and creation of high drying stresses in the sheet surface. However, when drying shrinkage is allowed, bending stiffness is increased despite a drop in elastic modulus. This is due to increased thickness, which compensates for the loss in elastic modulus. Therefore, the presence of starch affects bending stiffness in two ways: when drying is restricted, starch promotes increased elastic modulus as described; with drying shrinkage allowed, bending stiffness increases as a result of increasing sheet thickness. Since a practical papermaking process involves a large variety of shrinkage behaviors in the cross direction, the mechanism behind bending stiffness can vary in the cross direction based on the shrinkage profile across the entire width of the web.

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