Adsorption of Xylan onto Cellulose Fibers Pretreated with Cationic Polyelectrolyte and Its Effect on Paper Properties

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Xylan is known as a strength-enhancing agent for paper. However, it is difficult to adsorb xylan onto cellulose fibers because it carries the same negative charge as fibers. Therefore, either cationization of xylan or addition of cationic polyelectrolyte is required to maximize the effect of xylan use. In this study, cationic polyelectrolyte was used to promote xylan adsorption onto cellulose fiber. The effect of the polyelectrolyte type on the successive adsorption of xylan and its influence on paper properties was examined. The mechanism for improving paper strength by xylan adsorption on polyelectrolyte pre-formed layers was investigated through the viscoelastic properties of the preformed layers on model cellulose films using Quartz Crystal Microbalance with Dissipation (QCM-D). Both tensile and tear indices of paper were improved with the adsorption of xylan onto pulp fibers. It was suggested that the adsorbed xylan onto the polyelectrolyte preformed layer formed a complex layer that gave rise to a large contact area between xylan and fiber. The increase in the physical strength of paper depended on the structure of the polyelectrolyte-xylan layers. Highly charged cationic polyelectrolytes that form a flat adsorption layer gave a relatively lower increase in physical properties. On the other hand, thicker and more viscous adsorbed layers improved paper strength significantly.

Keywords: Xylan; polyelectrolytes; Adsorption; QCM-D; Paper properties

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INTRODUCTION

Commonly used dry-strength additives for paper and paperboard include derivatives of biopolymers. Some examples of these are starch, guar gum, and carboxymethyl cellulose (Moeller 1966; Dugal and Swanson 1972; Lindström *et al.* 1985; Alince *et al.* 1990; Ekevåg *et al.* 2004; Watanabe *et al.* 2004; Hubbe 2005; Ghasemian *et al.* 2012). Polyacrylamides are also widely used as dry strength agents (Spence 1999). Additives that afford effective strength enhancement for paper products have some common features. They are water-soluble materials, and they have a mechanism by which the material is retained efficiently on fiber surfaces. They also have sufficiently high molecular mass so that the additives remain on the outer surfaces of the fibers, and an ability to form hydrogen bond with fiber (Robinson 1980). The hemicellulose components of fibers share many of the abovementioned attributes. Thus the use of hemicelluloses in papermaking tends to increase the paper strength (Molin and Teder 2002).

Hemicelluloses, one of the main constituents of lignocellulosic materials, are heteropolysaccharides and contain small amounts of certain uronic acids. Some hemicelluloses are associated with the cellulosic portion of the plant, whereas others are more closely associated with lignin (Salmén and Olsson 1998; Åkerholm and Salmén 2001). Because the chemical and thermal stability of hemicelluloses is generally lower than that of cellulose, presumably due to their lack of crystallinity and the lower degree of polymerization, a large amount of hemicelluloses change dramatically and are more easily degraded and extracted with lignin during kraft pulping (Henriksson and Gatenholm 2001). Therefore, the percentage of hemicelluloses in the pulp is always less than in the original wood, and this is closely associated with the yield of chemical pulp, which is generally less than 50%. Unbleached pulp is generally stronger than bleached pulp, and this is attributed to the presence of a larger amount of hemicelluloses in the unbleached pulp. The presence of hemicellulose in pulp has been shown to give enhanced strength of the fiber due to improved adhesion properties between cellulose fibers (Meller 1965). This is because hemicelluloses have adhesive properties. Also, the role of hemicelluloses as papermaking additives has been investigated by Mobarak *et al.* (1992) and Bhaduri *et al.* (1995). They have shown that hemicellulose obtained from ramie fiber acts to considerably reduce energy consumption during the beating operation, and to improve the strength of paper sheets. Hemicelluloses extracted from seed (Lima *et al.* 2003), bagasse, and rice straws (Mobarak *et al.* 1992) were examined as papermaking additives as well, and it was shown that they improved the mechanical properties of paper sheets without affecting its optical properties.

Simonson (1963) showed that a considerable part of the hemicellulose was dissolved from the wood into the pulping liquor in the early part of the cooking process. It was suggested that a part of this dissolved hemicellulose would be re-adsorbed on the pulp fibers, and this affects not only the total yield but also the tensile strength and beatability of the manufactured pulp (Dahlman *et al.* 2003; Danielsson and Lindström 2005). It has been known that the adsorption of xylan on cellulose fibers is a relatively slow process, probably due to the fact that the xylan and the cellulose surfaces have the same electrical charge (Neuman *et al.* 1993; Claesson *et al.* 1995; Österberg *et al.* 2001). To overcome the adsorption interference caused by the electrostatic repulsive force, Linder *et al.* (2003) introduced a mechanism for xylan adsorption on cellulose surfaces based on the self-association of xylan at high temperatures under alkaline conditions. According to the cited investigation, the adsorption of xylan was strongly influenced by temperature under alkaline conditions, and consequently, an increase in temperature affects the alkaline degradation of the xylan side groups, *i.e.*, uronic acid. The degradation results in decreasing xylan solubility and causes the formation of self-associated xylan aggregations. Thus, it was suggested that the degradation of xylan side groups increases xylan self-associated aggregation so that the driving force for xylan assembly adsorption on a cellulose surface increases. However, the adsorption of xylan is strongly influenced by temperature and time under alkaline conditions; thus this adsorption treatment will consume considerable amounts of time and thermal energy.

The adsorption of xylan on cellulose fiber can be increased if xylan is cationically modified. The charge density of cationic xylan influences both the amount of adsorption and the structure of cationic xylan layers adsorbed on the model cellulose film (Galván *et al*. 2012; Lee *et al.* 2014). This suggests that electrostatic attraction plays an important role in xylan adsorption onto cellulose fiber. Xylan adsorption may be increased if one pre-adsorbs cationic polyelectrolyte on the fiber prior to xylan adsorption.

This study describes an attempt to improve the adsorption of xylan on cellulose fibers under alkaline or neutral conditions when xylan is present in a soluble form. We examined the effect of several cationic polyelectrolytes used in papermaking on xylan adsorption when they were used as adsorption promoters for xylan adsorption on cellulose fiber. The effect of xylan adsorption on paper properties has been examined as well.

EXPERIMENTAL

Materials

Commercial bleached mixed hardwood kraft pulp (Hw-BKP) supplied by Moorim P&P Co., Korea was used to prepare the handsheets. The pulp consisted of 80.7% cellulose, 18.2% hemicellulose, 0.5% lignin, and 0.3% ash. The uronic acid content was measured using the carbazole-sulfuric acid method (Li *et al.* 2007), which gave a value of 289±9 μmole/g. Commercial hemicellulose, 4-O-methylglucuronoxylan (Sigma X0502, hereafter noted as "XB") separated from birch wood was used as xylan in this study. As cationic polyelectrolytes, branched polyethyleneimine (PEI, M_w ~25,000, Sigma Aldrich), poly-diallyldimethylammonium chlorides (poly-DADMAC), one with a low molecular weight (M_w< 100,000, Sigma Aldrich), denoted as PDAD_L hereafter, and the other with a high molecular weight (M_w< 450,000, Sigma Aldrich), denoted as PDAD_H hereafter, two cationic polyacrylamides, one used to aid retention, denoted as PAM_R hereafter, (C-PAM, M_w 500,000 ~ 1,000,000, Ciba), and the other used as a dry-strength additive, denoted as PAM_S hereafter (C-PAM, M_w ~ 50,000, Eyang Chemical Co., Korea), and cationic starch (CS, M_w ~100,000, Samyang Genex Co., Korea), were used. The properties and structures of cationic polyelectrolytes are presented in Table 1.

Table 1. Properties of the Cationic Polyelectrolytes

Polyelectrolyte	ID	Molecular weight (g/mol)	Charge density (meq/g)
PEI	PEI	~25,000	+ 6.01
Poly-DADMAC High MW	PDAD_H	<450,000	+ 6.17
Poly-DADMAC Low MW	PDAD_L	<100,000	+ 6.17
C-PAM Retention Aid	PAM_R	<1,000,000	+ 0.47
C-PAM Strength Agent	PAM_S	~ 50,000	+ 2.24
Cationic Starch	CS	~ 100,000	+ 1.50

Methods

Determination of polymer addition levels

To determine the addition levels of polyelectrolytes, the change of zeta potential of Hw-BKP fibers was evaluated by using SZP-04 (Mütek, Germany). With the addition of polyelectrolytes, the zeta potential of fiber was reversed to cationic, and increased in value until it reached a plateau level. The zeta potential of the fiber at the plateau level differed depending upon the charge density, structure, and molecular weight of polyelectrolytes. A linear and high molecular weight PAM_R and two linear and highly charged PAM_S and PDAD_S show plateau level of zeta potential at the addition level of 0.6%. The addition level of branched PEI and CS required to reach the plateau level of zeta potential was 0.4%, and that for PDAD_H was 0.2%.

Preparation and properties of handsheets

Hw-BKP was disintegrated for 30,000 revolutions and screened using a vibration screen with 200-mesh wire to remove fines. The stock was diluted to 0.5% consistency (pH 7.5-8.0). The conductivity of the stock was 125 μS/cm. Prior to sheet preparation a cationic polyelectrolyte solution was added to the pulp slurry. Solutions of cationic polyelectrolyte at 0.5% were prepared and used for adsorption experiments. The dosage that gives the plateau level in zeta potential was used. After the addition of the cationic polyelectrolyte the mixture was stirred for 20 min, and then the pulp was washed with deionized water. Deionized water was used in rinsing stage and then the conductivity and pH were adjusted again to the condition of the pulp stock (pH 7.5-8.0, conductivity 125 μS/cm). Birchwood xylan was then added at addition rates of 1, 2, 4, and 6 wt % based on the oven-dried weight of pulp. After stirring for 20 min., the zeta potential of the pulp fiber was measured. Handsheets with a target basis weight of 80 g/m^2 were prepared according to the TAPPI method (T205 sp-95). Before measuring the physical properties of the handsheet, preconditioning was performed at least for 24 h. Physical properties of the handsheet were evaluated according to the ISO test methods.

Adsorption experiments using QCM-D

Quartz Crystal Microbalance with Dissipation (QCM-D) crystals were coated with trimethylsilyl cellulose (TMSC) using a spin coater. The TMSC films were hydrolyzed to cellulose by exposing the films to the vapor at 10% of HCl for 4 min (Kontturi *et al.* 2003).

The QCM-D sensor with model cellulose film was equilibrated in the electrolyte solution for at least 6 h before injecting the polyelectrolyte solution into the chamber to eliminate the effect of viscosity changes in the solvent and possible swelling or deswelling effects due to changes in ionic strength. After replacement of the electrolyte solution with a corresponding solution containing cationic polyelectrolytes, the frequency and dissipation changes were followed as a function of time. During QCM-D experiments, the solution was injected through the chamber at a constant flow rate of 100 μL/min. Then, the electrolyte solution was injected for washing followed by the addition of birch wood xylan. The washing process was performed to remove polyelectrolytes in the media so that xylan can adsorb onto the cellulose without any interfering hindrance caused by remaining polymers. The viscoelastic properties of the surface layer were determined quantitatively by the model presented by Voinova *et al.* (1999). This model was derived for a homogeneous film treated as a Voigt element, assuming that it conserves its shape and does not flow under shear deformation. The cited work resulted in an analytical expression for the responses in terms of frequency and dissipation shifts based on the properties of the bulk liquid, adsorbed film, and quartz crystal. These relationships were applied in reverse to calculate the properties of the adsorbed layer from the experimentally observed overtones of frequency and dissipation. The calculation routines, which are commercially available through the Q-tools software from the instrument supplier, were used to calculate the adsorbed layer thickness (Tammelin *et al.* 2004).

RESULTS AND DISCUSSION

Adsorption of Cationic Polyelectrolytes Studied with QCM-D

The adsorption of cationic polyelectrolytes on the cellulose surface was measured by QCM-D at the third overtone (15 MHz). Figure 1 shows the adsorption of cationic polymers on the cellulose surfaces. The data represent the frequency change monitored with time. In all cases, cationic polyelectrolyte adsorption followed a similar trend. When cationic polyelectrolytes were injected into the adsorption chamber, the polymers were adsorbed on the cellulose surface; this was driven by electrostatic interactions. There is no effect of molecular weight of PDADMAC on adsorption. However, the adsorption rates were somewhat different depending on the charge density of the polymer. In the early stage of adsorption, the polymer with higher charge density was adsorbed onto the cellulose surface more effectively. This means that adsorption equilibrium was reached relatively faster for highly charged polymers than for low-charged ones, *i.e.* PEI and poly-DADMAC reached saturation relatively faster than PAM and CS. However, the change in frequency was low for highly charged linear polymers, indicating that these polyelectrolytes adsorb in a flat conformation to cover the cellulose surface more effectively. Branched PEI adsorbed more on cellulose surface than highly charged linear polyelectrolytes such as poly-DADMAC, since it gave thicker adsorbed layer (Li *et al.* 2000). CS, which has a highly branched amylopectin as its main component, showed the highest frequency change among the polymers tested. This high adsorbed amount of the starch is likely to be due to its low charge density (Chin *et al.* 2012). In addition, it might be due to the water associated with the adsorbed starch.

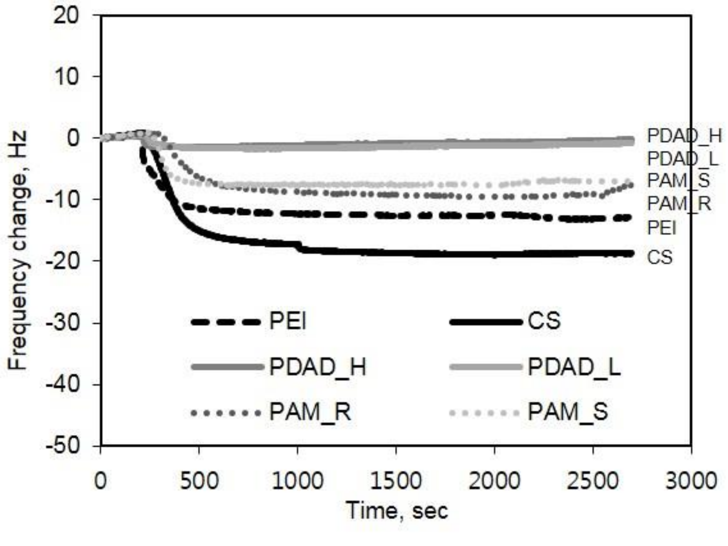

Fig. 1. Adsorption of cationic polymers on model cellulose film studied with QCM-D

Adsorption of Xylan onto Cellulose Surface Pretreated with Cationic Polyelectrolyte

A cellulose surface pretreated with cationic polyelectrolytes tends to adsorb more xylan because of the electrostatic interaction between cationic polymer and anionic xylan. Figure 2 shows the change in frequency that occurred when xylan was adsorbed on the cellulose surface pretreated with a cationic polyelectrolyte layer. Interestingly, even though CS was adsorbed more than PEI and PAM_R, relatively larger changes in frequency were observed for PEI and PAM_R layers by xylan adsorption. In general, low-charged polyelectrolytes (PAM, CS) seemed to have greater interactions with xylan than highly charged polymers (poly-DADMAC). The only exception was PEI. Polyelectrolytes that form large loops after adsorption, *i.e.* PAMs, or polyelectrolytes that have three-dimensional network structures like PEI and CS gave greater adsorption of xylan. When xylan adsorbed on a PEI or PDAD layer, the equilibrium was reached relatively quickly. In the case of low-charged PAMs, however, equilibrium was reached after relatively long adsorption time. This indicates that adsorption kinetics is influenced by the charge density of polyelectrolytes.

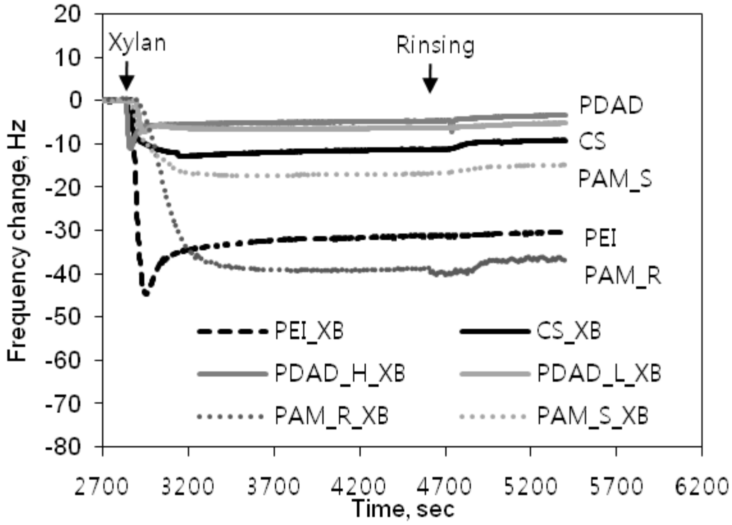

Fig. 2. Frequency change curves when xylan was adsorbed on polyelectrolytes-layers ('XB' means commercial birch xylan)

Properties of Adsorbed Xylan Layers

The dissipation value for all polyelectrolytes and xylan was quite substantial (Fig. 3). The Voigt model was used to calculate the thickness of the adsorbed polyelectrolyte-xylan layers, and results are depicted in Fig. 4. There was obvious difference in thickness between low- and high-charge-density polyelectrolytes. Thickness of adsorbed polymers was greater when low-charge-density polyelectrolytes, such as PAM and CS, were used than that for high-charge-density polymers, such as PDADMAC. Low-charged PAM_R, which forms extended loops and tails at the interface, gave the greatest adsorption of xylan, while high-charge PDADMACs, which adsorb in a flat conformation on the surface, gave very low adsorption of xylan. The thickness of CS layer was greater than that of PAM_R despite the fact that CS has a lower molecular weight. This is because CS is a highly branched polymer that gives a soft and bulky structure (Chin 2010). CS, however, did not increase xylan adsorption as much as PAM_R. On the other hand, branched PEI gave greater adsorption of xylan even though other high-charge-density polymers. This indicates that a highly charged and branched structure of the cationic polyelectrolyte is more favorable for subsequent xylan adsorption. This is probably because branched structure gives a more extended structure after adsorption. Kolasińska *et al.* (2007) and Trybała *et al.* (2009) showed that when PEI is used as a first layer it acts like a scaffold, making multilayer structures thicker and more stable. In particular, PEI forms a thicker precursor layer at alkaline pH than at neutral or acidic conditions, since only a small fraction of amino groups of PEI was charged under alkaline conditions. This is also supported by the fact that the adsorption of PEI increased monotonically with increasing pH at fixed ionic strength (Mészáros *et al.* 2002). The charge density of PEI at pH 8 was relatively high, however, to make xylan form a thick layer. This unique characteristic of PEI can be very useful to increase xylan adsorption on cellulose fiber, which will increase interfiber bonding. Since the molecular weight of the PEI used in this work was not large, not much flocculation of fibers will occur with the use of such PEI as a pretreatment polymer, and consequently the formation uniformity of the paper will not be hurt.

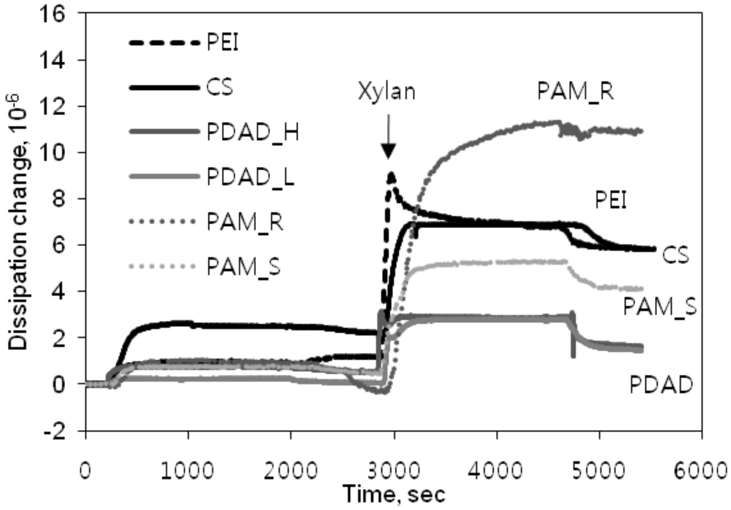

Fig. 3. Dissipation change curve when polyelectrolytes and xylan are adsorbed on the model cellulose surface

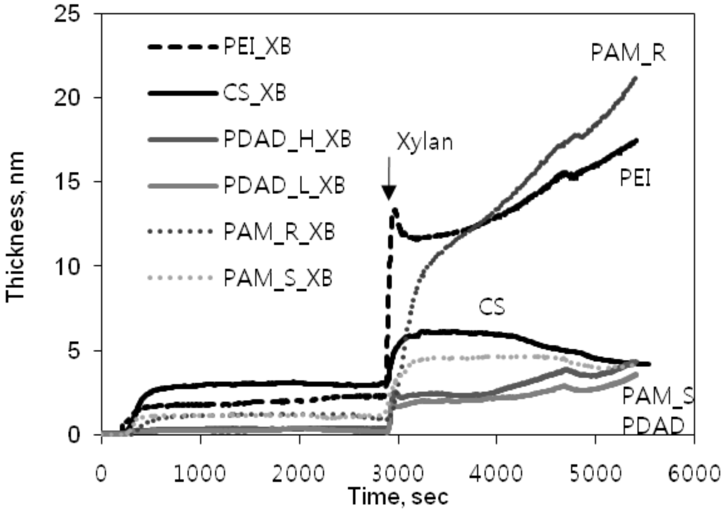

Fig. 4. The thickness calculated by Voight model for polyelectrolytes-xylan layers ('XB' means commercial birch xylan)

The thickness of the xylan layers adsorbed onto the CS layer was thinner than those adsorbed on the PEI and PAM_R layers. This indicates the interaction between CS and xylan was somewhat weak. On the other hand, the thickness of the xylan layer increased considerably when it adsorbed onto the PAM_R-layer. A xylan molecule appears to be adsorbed in long loop forms when its segments are anchored to the high molecular weight and low charged cationic PAM layer. On the other hand, in the case of CS, which is a highly branched and weakly charged polymer, the steric hindrance caused by starch branches and low electrostatic attraction appear to make xylan adsorption decrease.

Paper Properties and Adsorption Mechanism

The surface charge of the pulp fibers was affected by the type of adsorbed polyelectrolytes. Figure 5 shows the zeta potentials of pulp fibers that had been modified by polyelectrolyte-xylan layers. The initial zeta potential of the pulp fiber was −93.5 mV, and xylan addition decreased it to −102.6 mV, indicating that only a small amount of xylan was adsorbed onto fiber. When polyelectrolytes were added, the zeta potential of the fiber reversed to a positive charge, but it was different depending on the polyelectrolyte used. Generally, highly charged polyelectrolytes with greater molecular weight gave higher positive zeta potential. CS showed a very low zeta potential even though the amount of starch adsorption was largest among the polyelectrolytes tested, and this is attributed to its low charge density (Chin 2010). When negatively charged xylan (XB) was adsorbed on an oppositely charged polyelectrolyte layer, the zeta potential of the pulp fiber reversed again, and its equilibrium was reached at a xylan dosage level of 2 wt % (Fig. 5).

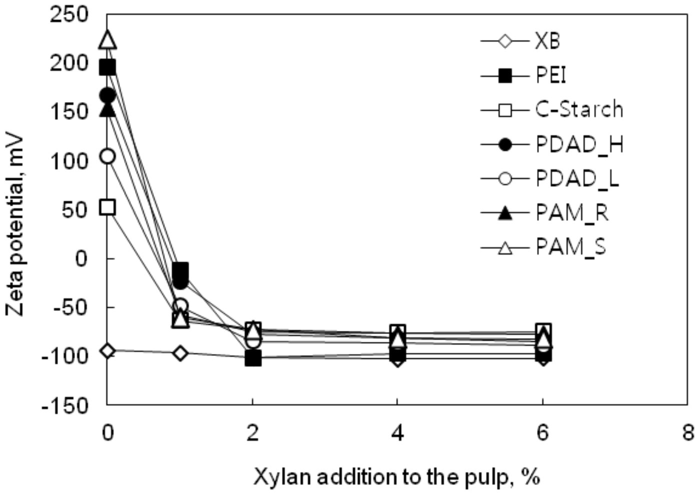

Fig. 5. Effect of xylan dosage in a presence of polyelectrolyte pre-formed layers on zeta potential of pulps (‘XB’ means the adsorption of commercial birch xylan on pulp fibers without polyelectrolyte layer)

Handsheets were prepared from Hw-BKP fibers treated with different amounts xylan. Polyelectrolyte adsorption on pulp fibers was followed by xylan adsorption. Preformed polyelectrolyte adsorption level corresponded to the maximum adsorbed amount according to the zeta potential results. Figures 6 and 7 show the physical strength of handsheets as a function of xylan addition. Addition of xylan alone without the pretreatment with cationic polyelectrolytes increased tensile and tear strength only slightly due to the lack of electrostatic attraction between the cellulose fibers and xylan. When CS was used, however, strength increased without any xylan adsorption, since it formed hydrogen bonds with fibers. Addition of other polyelectrolytes, however, did not give any significant difference in strength without xylan addition. PDADMAC decreased strength slightly probably due to a reduction in the conformability of the PDADMAC-covered fibers (Torgnysdotter *et al.* 2007).

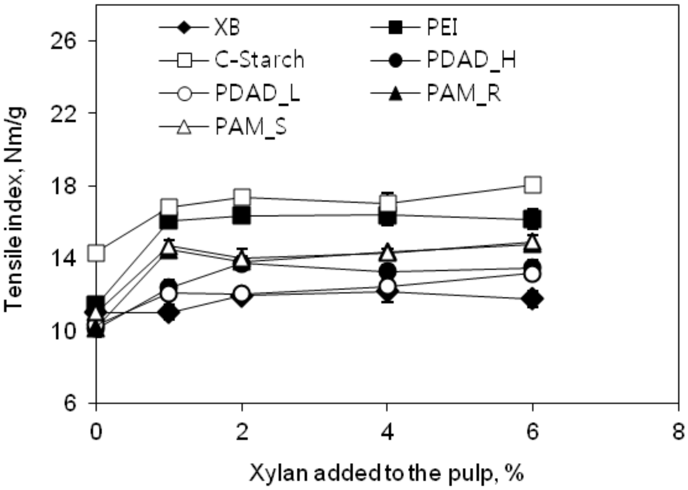

Fig. 6. The effect of xylan adsorption on polyelectrolytes pre-formed layers on tensile strength ('XB' means commercial xylan)

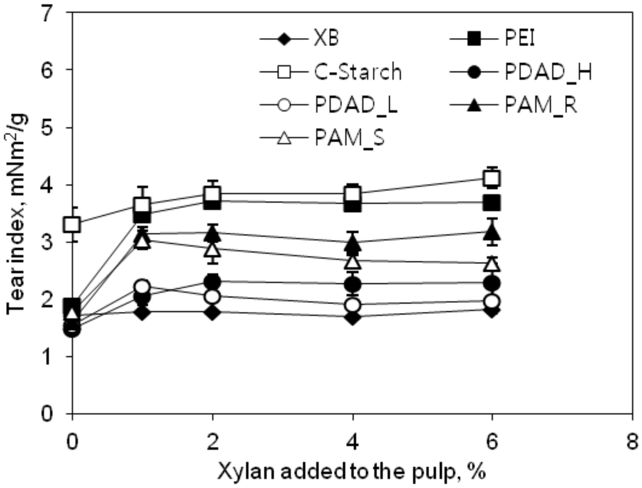

Fig. 7. The effect of xylan adsorption on polyelectrolytes pre-formed layers on tear strength ('XB' means commercial birch xylan)

Physical strength increased significantly with xylan adsorption. These results are in accordance with Maximova *et al.* (2001), who reported on the pre-adsorption of PDADMAC onto fiber surfaces with lignin. Both tensile and tear indices of paper increased, as shown in Figs. 6 and 7. These findings suggest that the adsorption xylan onto the polyelectrolyte preformed layer resulted in a complex layer, which gave rise to a large contact area between xylan covered fibers, resulting in improvement of tensile and tear strength. Since a tear strength measurement is obtained as a result of total energy consumption, the result may include energy spent in strain. A great increase of strain was also observed, as shown in Fig. 8. When the addition of xylan was higher than 2 wt%, the strength remained almost constant or decreased slightly.

When xylan was added to the pulp fibers treated with cationic polyelectrolytes, light-scattering coefficient decreased. This indicates a decrease in the free surface of paper and an increase of degree of interfiber bonding with xylan adsorption on polyelectrolyte-treated pulp fibers. Thus, the increase in physical strength by xylan addition is most likely attributable to the increase of the relative bonding area between fibers (Page 1969).

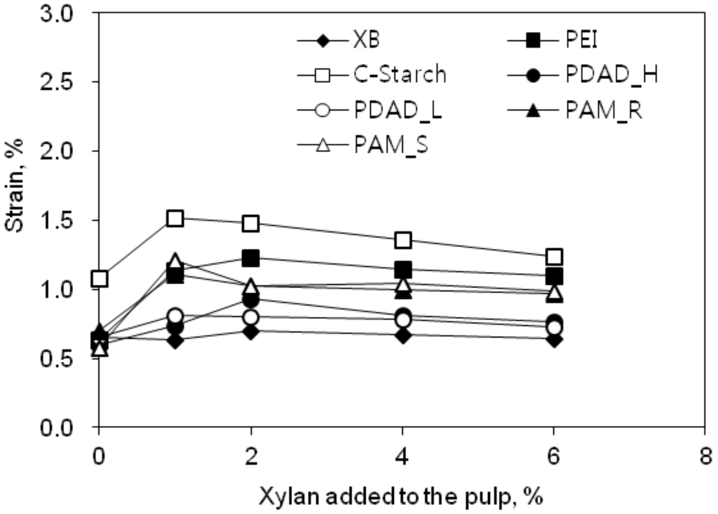

Fig. 8. The effect of xylan adsorption on polyelectrolytes pre-formed layers on strain ('XB' means commercial birch xylan)

The strength of polyelectrolyte-xylan treated papers at 2 wt. % of xylan addition was compared to that of untreated paper. Figure 9 shows the relative increase in paper strength for the cationic polyelectrolytes tested. PEI-treated fibers showed the most pronounced improvement, followed by PAM_R-treated fibers. It was shown that a thicker and more viscous xylan layer was formed on the fibers treated with PEI and PAM_R (Fig. 4). Thus, one may conclude that these layers improved paper strength through an increased degree of contact between fibers. On the other hand, high-charge-density polyelectrolytes, such as PDADMAC, adsorbs flat on fiber surface to give thin xylan layer (Fig. 10).

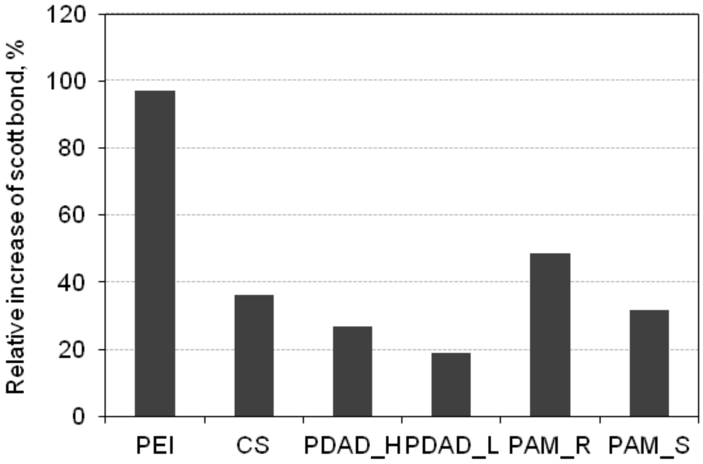

Fig. 9. Relative increase in tensile index of papers made of modified pulp fibers by xylan adsorption on different polyelectrolyte pre-formed layers

According to Notley *et al.* (2005), the more viscous the polyelectrolyte layer, the higher was the adhesion between multilayer-covered silica surfaces, as determined by AFM. In addition, Eriksson *et al.* (2005) showed that the multilayers increased the paper strength via an increased degree of contact between fibers and an increase in the number of fiber-fiber contacts. Since high molecular weight PAM_R causes strong flocculation of fibers and deteriorates paper formation, it may have well suited for practical use in papermaking. PEI appears to be the best polyelectrolyte for pre-adsorption.

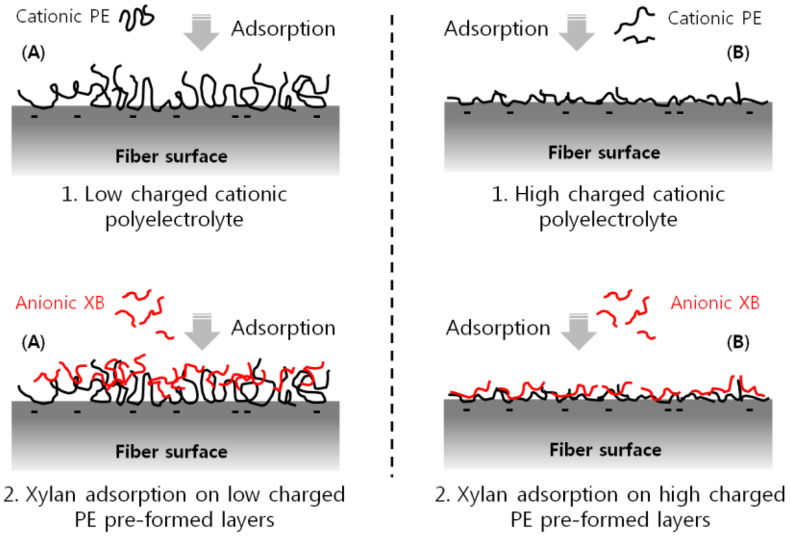

Fig. 10. Concepts of polyelectrolytes and xylan adsorption onto cellulose surface, (A) xylan adsorption onto low charged cationic polyelectrolyte pre-formed layer, (B) xylan adsorption onto high charged cationic polyelectrolyte pre-formed layer

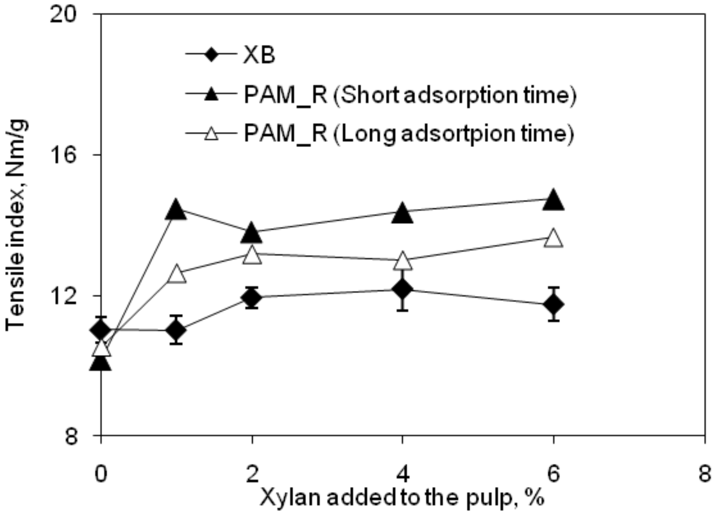

Fig. 11. Effect of adsorption reaction time of PAM on tensile strength after xylan adsorption on PAM pre-formed layer (‘XB’ means commercial birch xylan)

The effect of adsorption time of PAM_R on the tensile of paper was investigated. The cationic retention aid polyelectrolyte was adsorbed onto pulp fibers for a short (5 min) or long adsorption times (24 h). The zeta potential and paper strength for two different adsorption conditions were compared. Results showed that a long adsorption time of PAM_R decreased zeta potential by 77% compared to a short adsorption time. This is consistent with penetration of PAM molecules into the pores in fibers or a change in PAM conformation. Thus, the passage of time gave rise to a negative effect on tensile strength improvement by xylan (Fig. 11).

CONCLUSIONS

1. The adsorption of xylan was promoted by forming a pre-layer of cationic polyelectrolyte on pulp fibers. Also the charge density, structure, and molecular weight of cationic polyelectrolytes influenced their adsorption onto fiber. Polyelectrolytes with high charge density adsorbed more rapidly onto the surface in the beginning of adsorption, but the total adsorbed mass was less than that of the low charged polyelectrolytes.

2. The xylan adsorption on polyelectrolyte pre-formed layers was investigated using QCM-D. The thickness of the xylan layers adsorbed on CS preformed layers was thinner than those on PEI or PAM_R layers. The thickness of the xylan layer increased considerably when it adsorbed onto the PAM_R-layer, indicating that the xylan molecule adsorbs in long loop forms when its segments are anchored to the high molecular weight and low charged cationic PAM layer.

3. Tensile strength, tear strength, and internal bond strength were improved as the xylan was adsorbed onto pulp fibers.

4. The increase in the physical strength of paper depended on the structure of the polyelectrolyte-xylan layers. The thicker and more viscous layers improved the paper strength more significantly, presumably through an increased degree of contact area between the fibers. In contrast, high charge density polyelectrolytes showed a relatively lower increase in physical properties, and this was attributed to the lower interfiber bonding.

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