Addition of Polyvinylamine in Chemi-thermomechanical Pulp and Kraft Pulp and the Effects on Dewatering, Strength, and Air Permeance

Björn Sjöstrand [a] and Annelie Brolinson [b]

The addition of polyvinylamine (PVAm) in the wet-end of the papermaking process was investigated. The changes in pulp drainage, vacuum dewatering, tensile strength, and air permeance were measured with and without PVAm. Both chemi-thermomechanical pulp (CTMP) and bleached kraft pulp were used. The dewatering and drainage abilities of the different pulps was examined by measuring the dewatering resistance (°SR), the water retention value (g/g), and the vacuum dewatering. The tensile strength and air permeance values were tested on paper sheets. The results showed that the initial dewatering was faster for pulps with PVAm, and this effect was greater for the CTMP and at higher basis weights. The strength increased when PVAm was added but only if the pulp was washed before sheet forming. The unwashed pulp with PVAm had worse formation, which resulted in lower tensile strength values. The air permeance of the sheets was higher with the addition of PVAm, primarily as a result of higher flocculation. Adding PVAm to the stock suspension in the wet-end of the paper machine has great potential for end-products that require high air permeance and tensile strength properties. PVAm could also act as a dewatering enhancing agent, but caution must be taken regarding the potential of formation problems.

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Keywords: Air permeance; CTMP; Dewatering; Polyvinylamine; PVAm; Wet-end chemicals

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INTRODUCTION

The main part of the raw materials for paper is wood fibers. Wood fibers are prone to adhere to each other to form flocks, a phenomenon known as flocculation. Wood fibers are also efficient in retaining water. Prior to paper production, the fibers must be separated by dilution in water to avoid flocculation, with solids contents as low as 0.2% oven-dry consistency before the headbox. Specifics for fiber flocculation can be determined by the fiber crowding factor (Kerekes and Schell 1992). The huge amounts of water must then be removed by dewatering in several different steps. Dewatering during paper production is therefore a well-known bottleneck that affects both the productivity and energy efficiency of the process. Some strategies to increase dewatering rates include optimal machine design, forming fabric design, reduction of rewetting, and applying wet-end chemicals (Kuhasalo *et al.* 2000; Norman 2000). Heavy dilution in water, followed by huge efforts in water removal, is often referred to as the water paradox.

The removal of water from the sheet becomes more and more difficult as it progresses in the paper process. The energy required for dewatering increases in the direction of the paper machine with each progressing unit operation (Ramaswamy 2003). One of the reasons for this is the location of the water in the sheet structure. Water located around the fibers is most easily removed, followed by water in the fiber lumen, while the water within the fibers walls is the most difficult to remove (Stenström and Nilsson 2015; Paulapuro 2000).

Increasing dewatering efficiency and lowering the impact of dewatering-related issues in the papermaking process could have a huge effect both commercially and in the utilization of resources in a sustainable fashion.

One strategy to increase dewatering efficiency is the addition of chemical additives to the stock suspension in the wet-end before the headbox. Chemicals added at this point in the papermaking process are known as wet-end chemicals. If these wet-end chemicals affect the sheet formation by opening the sheet structure, then the dewatering rate can increase. This has been shown in several articles (Räisänen *et al*. 1995; Hubbe *et al*. 2008; Svedberg and Lindström 2012). The fibers' swelling can also be affected by cationic polyelectrolytes, where polyelectrolytes with low molecular mass are more effective (Swerin *et al.* 1990).

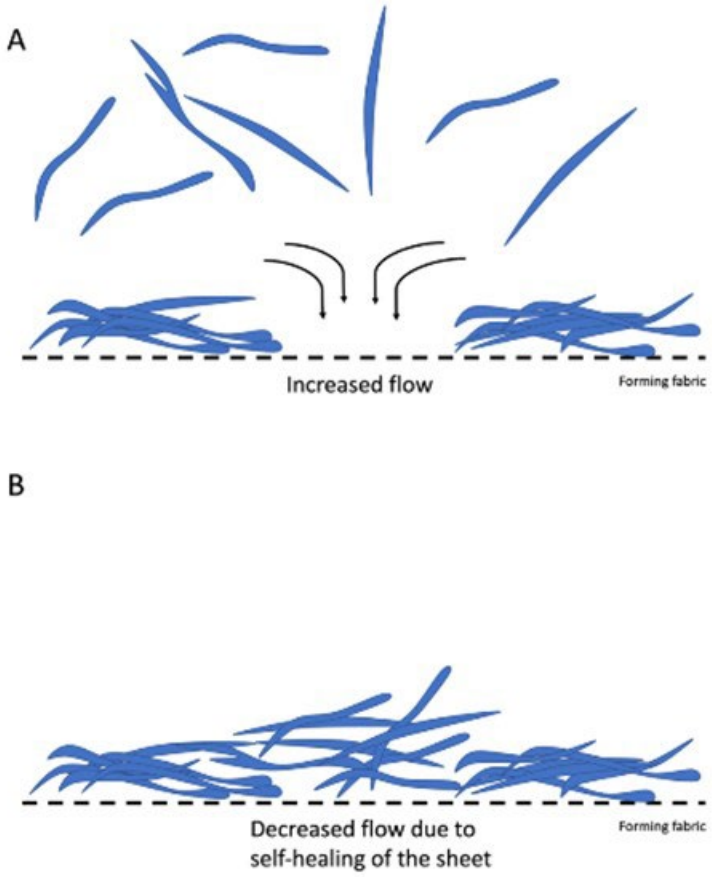

Fig. 1. Diagrams illustrating the self-healing mechanism where a) uncovered patches on the forming fabric increase the outflow of water in those locations, which in turn b) brings more fibers to cover the holes. Redrawn from Hubbe *et al.* (2020).

The fine material in a sheet of paper, and its relative position to the sheet structure, can affect mechanical dewatering such as vacuum dewatering and wet pressing. The dewatering process can also affect the fiber structure within the sheet. Several different mechanisms for slower dewatering by fine material transport have been reported by Hubbe *et al.* (2020), such as plugging of channels in the fiber network, sealing of fabric openings, inefficient dewatering by extreme flocculation, and membrane forming by fine materials. Self-healing of the fiber network is a phenomenon in which uncovered patches on the forming fabric increase the outflow of water in those locations, which in turn brings more fibers to cover the holes (Fig. 1) (Norman 1989).

When the dewatering is impeded by the plugging of flow channels in the fiber structure, retention aids can increase dewatering by attaching particles to fibers and preventing them from blocking channels, as shown in Fig. 2 (Balea *et al.* 2019).

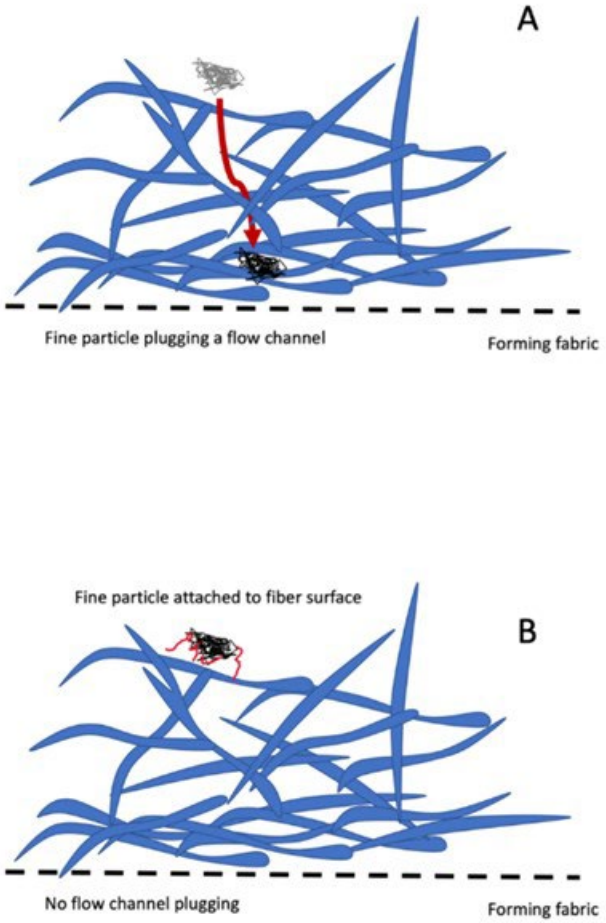

Fig. 2. Diagrams illustrating that dewatering is impeded by the a) plugging of flow channels in the fiber structure, but b) retention aids such as polyvinylamine (PVAm) can increase dewatering by attaching particles to fibers and keeping them from blocking channels. Redrawn from Hubbe *et al.* (2020).

Retention aids also keep chemicals and fine materials from blocking the pores of forming fabrics. The particles are held to the fibers by neutralization of repulsion with electrostatic forces by chemicals or salts in combination with bridging flocculation indicated by adding long polymers (Lindström 1989; Swerin and Ödberg 1997; Hubbe *et*

al. 2008; Sjöstrand *et al.* 2019). Papermaking with the addition of cationic polymers with branched structures can introduce patches of different charges on the fiber network, causing particles and fibers to adhere to the structure, which in turn can be positive for the early dewatering (Akari *et al.* 1996; Pfau *et al.* 1999). Bridging flocculation with the addition of cationic polymers can, in some cases, be reversible with sufficient shear force to break the network apart (Hedborg and Lindstrom 1996; Hubbe 2001; Tripattharanan *et al.* 2004).

Polyvinylamine (PVAm) has been used as a retention aid in papermaking (Shulga *et al.* 2003a; Merayo *et al.* 2017; Hollertz *et al.* 2017), as a dry- and wet-strength agent (Lindström *et al.* 2005; Yang *et al.* 2019), and for increasing the efficiency of sizing (Wu *et al.* 1997; Kumar *et al.* 2018, 2020). Polyvinylamine should not be confused with other varieties of polyamines, which are organic materials with a wide range of different structures, all of them with more than two amino groups attached. Wu *et al.* (1997) showed that for rosin emulsion sizing, the linear structure and small side chains of PVAm are beneficial for rosin particle retention. The adsorption of PVAm on cellulosic fibers under various conditions was investigated by Shulga *et al.* (2003a,b). They concluded that the adsorption was complex and that the strategy of addition in a paper machine would be critical to the successful addition of the PVAm. Westman *et al.* (2009) discussed the antimicrobial properties of PVAm, which inhibit the growth of microorganisms.

Because PVAm works as a retention aid by increasing adhesion between fibers and fine materials, the sheet porosity, formation, and air permeance might be affected by its addition, as well as the strength and dewatering performance. Previously published results by Ström and Kunnas (1991) state that the water retention value increase with additions of high mass cationic polymers.

This study focused on changes in the initial dewatering, vacuum dewatering performance, tensile strength, and air permeance with the presence of PVAm on chemi-thermomechanical pulp (CTMP) fibers and kraft pulp fibers. The experimental study investigated if PVAm can have a positive effect on dewatering in the forming section of a paper machine, dry strength, and air permeance when added to the stock in the wet-end of the paper machine.

EXPERIMENTAL

Fibers

Chemi-thermomechanical fibers from Norway spruce (*Picea abies*) were supplied by Rottneros AB (Sunne, Sweden) with a dryness of 95%. The CTMP fibers had a yield of 93% to 95% and a lignin content of 20% to 25%. The fines content was measured by an on-line PulpEye device (PulpEye AB, Örnskoldsvik, Sweden). The fines content was 35% with a standard deviation of 4.5%.

Bleached chemical kraft softwood pulp fibers mixed from Norway spruce and Scots pine (*Pinus sylvestris*) were supplied by StoraEnso AB (Skoghall, Sweden) with a dryness of 4%. The kraft fibers were refined in the mill to a drainage resistance of 25 °SR.

Chemicals

The PVAm was supplied by BASF SE (Ludwigshafen, Germany). The average molecular weight of the PVAm was 340,000 g/mol, and the polymer surface charge density was 15 ± 0.9 meq/g.

Methods

The CTMP fibers and kraft pulps were prepared into 0.2% mixture w/w with and without PVAm. The solutions with the PVAm were prepared as follows: 5.7 mL of 2 M sodium chloride (NaCl) was added per gram of dry pulp. The pH was adjusted to slightly above 9.5. One mL/g fiber of PVAm (10 g/L) was then added, amounting to 0.01 g PVAm/g fiber.

The zeta potential was measured by mixing 6 g of reference and treated CTMP fibers with 1 L of tap water each. A fiber potential analyzer (FPA Touch! – Fiber Potential Analyzer B0971, Emtec GMBH, Leipzig, Germany) was used, and standard measurements of the zeta potential and pH were performed. The polymer surface charge density of the PVAm was measured with a particle charge detector (Mütek PCD-03; BTG, Stockholm, Sweden), sometimes referred to as a "streaming current detector". The pH values of the untreated kraft and CTMP fibers before the addition of PVAm were 8.4 and 7.0, respectively.

One of the CTMP solutions with PVAm was semi-washed, where approximately 38% of the total volume (6 of 16 L) of the liquid phase was exchanged for fresh water without added chemicals. The PVAm that was already attached to fiber surfaces was assumed to be unaffected by the washing, as it most likely only affected the ratio of chemicals in the water. After sedimentation of the CTMP, colloidal matter and fines are expected to be present in the liquid phase; therefore, the concentration of these will be lower in the washed samples. The washed pulp was only used for strength and air penetration measurements.

The dewatering ability of the various pulps was examined by measuring the dewatering resistance (°SR) according to ISO 23714 (2014). The water retention value (WRV) (g water/g fiber) was determined according to ISO 5267-1 (1999). The vacuum dewatering was measured in a custom-built laboratory vacuum suction box with a commercial forming fabric with a triple layered structure (SSB) and an air permeability of 325 cubic feet per minute (cfm) delivered by Albany International (Rochester, NH, USA). Prior to the vacuum dewatering, isotropic sheets were formed in a custom laboratory sheet former with thorough agitation of the stock to ensure a consistent formation. These sheets were only for vacuum dewatering and were ruined by the dryness measurements. The apparatus used in this study is as described by Granevald *et al.* (2004), as redrawn in Fig. 3, with the exception that a plate with a single 5-mm opening was used. The vacuum level was set at -40 kPa; the basis weights of the sheets were 20 g/m^2 and 100 g/m^2; and the dwell times in vacuum were 0, 1, 2.5, 5, 10, and 20 ms. After each test, the dryness was measured according to ISO 638 (2008).

Sheets were also formed in another laboratory sheet former at 60 and 200 g/m^2, plane pressed, and restrained dried. The sheet former included agitation of the stock to achieve a consistent formation. The sheets were dried in a standardized climate according to ISO 187 (1990), under which the tensile and air permeance tests were also performed. Sheets that weighed as much as 200 g/m^2 were formed because they were intended to be subjected to air permeance testing according to ISO 5636-3 (2013). However, the air permeance testing was adjusted to a lower pressure than the standard because the sheets with PVAm were too open to work with the original settings. Single and double sheets were tested in the air permeance measurements in this study, and they were investigated at 0.7 kPa instead of 1.47 kPa because of the high flow. The CTMP and kraft sheets that weighed 200 g/m^2 were subjected to tensile testing according to ISO 1924-3 (2008). The 60 g/m^2 sheets were removed from the tensile tests because of their poor formation.

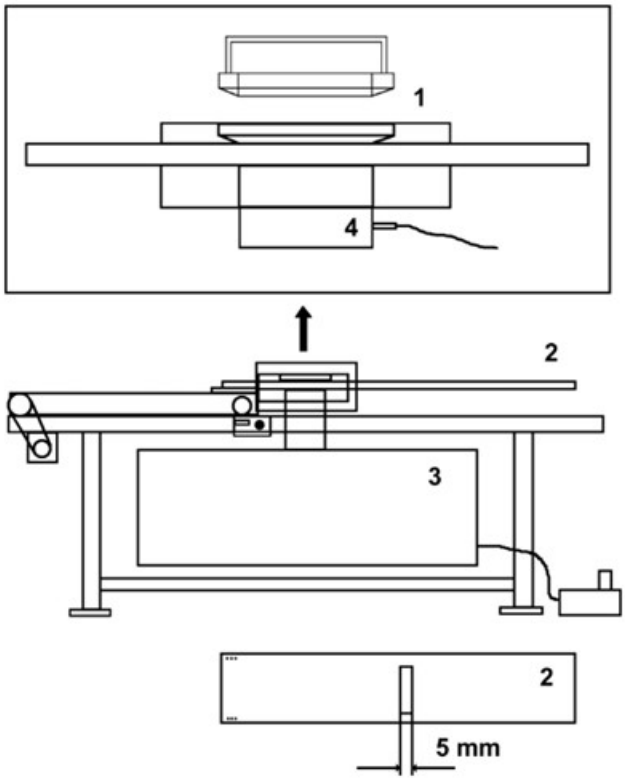

Fig. 3. Schematic of the laboratory suction box. The machine includes a 1) sample frame, 2) moveable plate with a 5-mm rectangular opening, 3) 300-L vacuum tank, and 4) transducer that logs the pressure directly underneath the sample. Redrawn from Granevald *et al.* (2004).

RESULTS AND DISCUSSION

The results from the zeta-potential measurements of the stocks are shown in Table 1.

Table 1. Zeta Potentials for the CTMP and Kraft Pulps with and without PVAm

Sample	Zeta Potential (mV)	95% Confidence Interval (mV)
CTMP ref	-87	± 1.5
CTMP + PVAm	-5.7	± 0.3
Kraft ref	-107	± 11
Kraft + PVAm	95	± 6.9

The drainage resistance and WRV results for the pulps are shown in Figs. 4 and 5. Examining the various pulps in the study, the drainage according to the °SR values resembles the early dewatering when water flows between fibers in the early stages of forming, and the WRV captures the later dewatering in high-vacuum suction boxes or in wet pressing with substantially higher dryness.

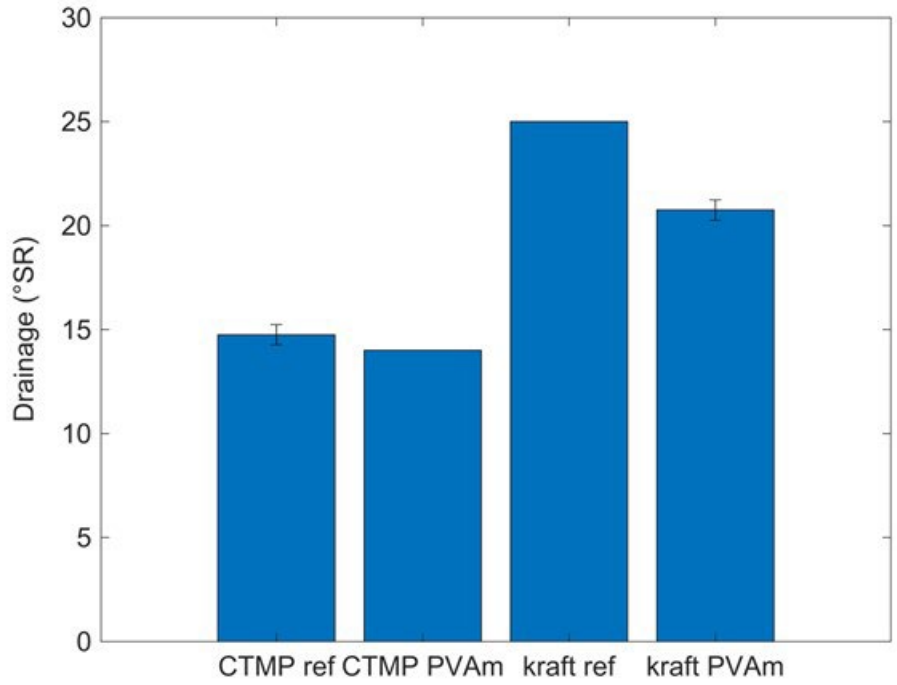

Fig. 4. Drainage results for the CTMP and kraft pulps with and without the addition of 0.01 g PVAm/g fiber. Error bars indicate 95% confidence interval.

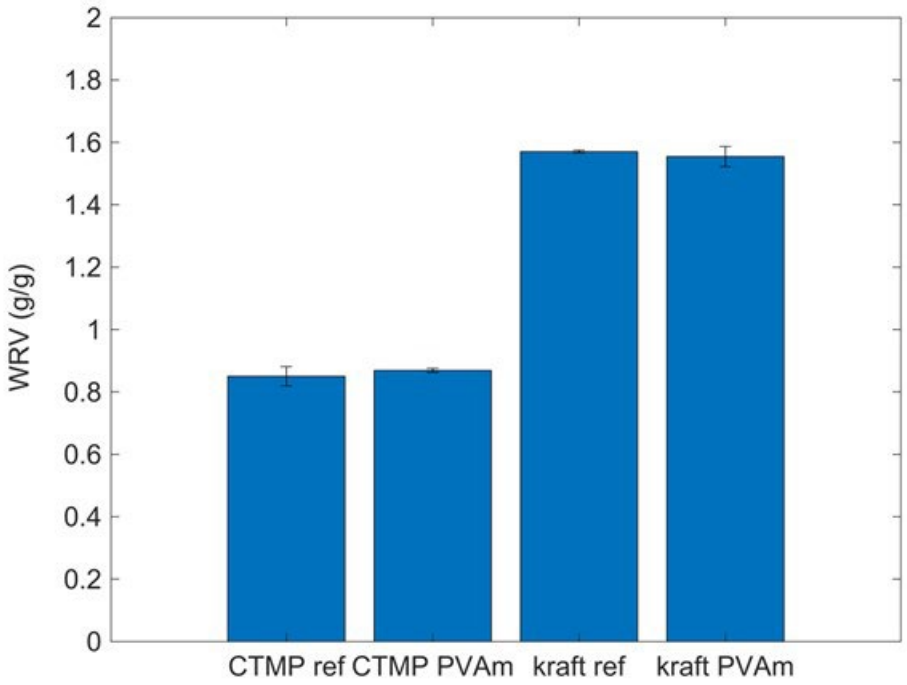

Fig. 5. WRV results for the CTMP and kraft pulps with and without the addition of 0.01 g PVAm/g fiber. Error bars indicate 95% confidence interval.

The drainage and water retention results indicate that addition of PVAm primarily affects the early dewatering, presumably by preventing the healing mechanisms, preventing the blockage of flow channels (Hubbe *et al.* 2020), and by increasing flocculation (Hubbe *et al.* 2008; Balea *et al.* 2019; Sjöstrand *et al.* 2019). According to the WRV results, the later dewatering was barely affected by the addition of PVAm under these conditions. This contradicts the literature (Ström and Kunnas 1991) and could be explained by the fact that the addition of PVAm does not affect the swelling of the fibers, as much as other high mass cationic polymers did (Ström and Kunnas 1991), but rather the flow behavior in lower-consistency suspensions. No effect on the swelling of fibers was observed. This may have occurred because the molecular weight of the PVAm was too high to affect bulk charges and to deswell the whole fiber, so surface charges might still have been affected.

The dewatering results are expressed as development in moisture ratio (g water/g fiber) with vacuum dwell time (ms). These are shown in Figs. 6 and 7. The dewatering behavior measured by the laboratory-scale vacuum dewatering equipment indicates that the early stages of dewatering are most influenced by the addition of PVAm. This can be explained by the same mechanisms of healing and flow channel blocking as for drainage (°SR). Schematic renderings of these are shown in Figs. 1, 2, and 8. At vacuum dwell times of 0 to 5 ms, differences were observed in dewatering, where pulps with the addition of PVAm were more effectively dewatered than the corresponding reference pulps. Comparing the 20 g/m^2 to 100 g/m^2 sheets, the effect was greater when the dewatering resistance was greater. The CTMP pulps showed a greater improvement in dewatering rates than the kraft pulps with the addition of PVAm, possibly as a result of higher flocculation.

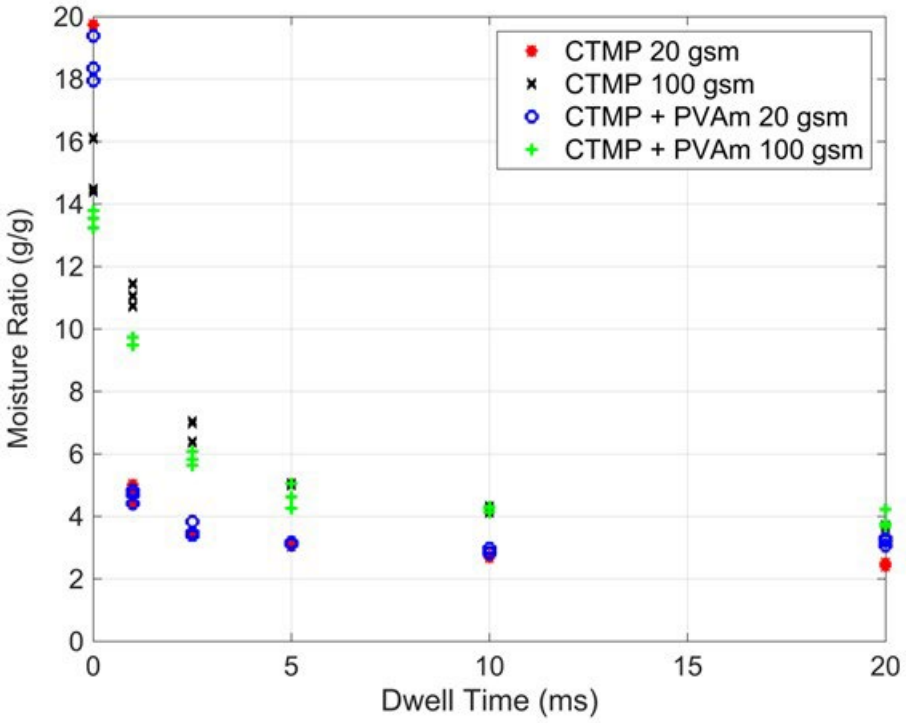

Fig. 6. Moisture ratio (g/g) and vacuum dewatering dwell time (ms) for 20 g/m^2 and 100 g/m^2 CTMP sheets with and without the addition of 0.01 g PVAm/g fiber

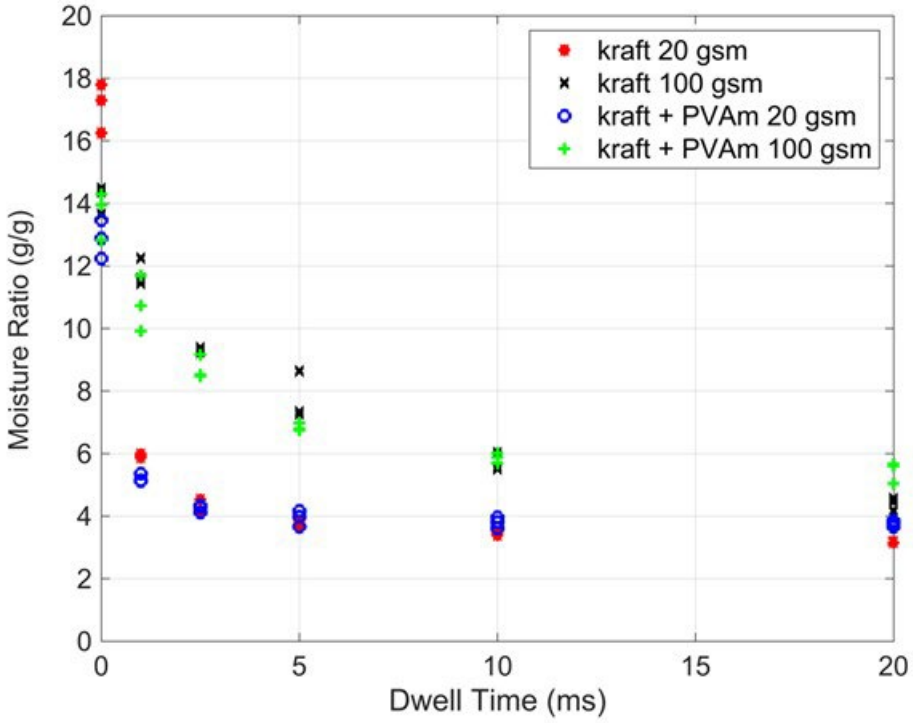

Fig. 7. Moisture ratio (g/g) and vacuum dewatering dwell time (ms) for 20 g/m^2 and 100 g/m^2 kraft pulp sheets with and without the addition of 0.01 g PVAm/g fiber

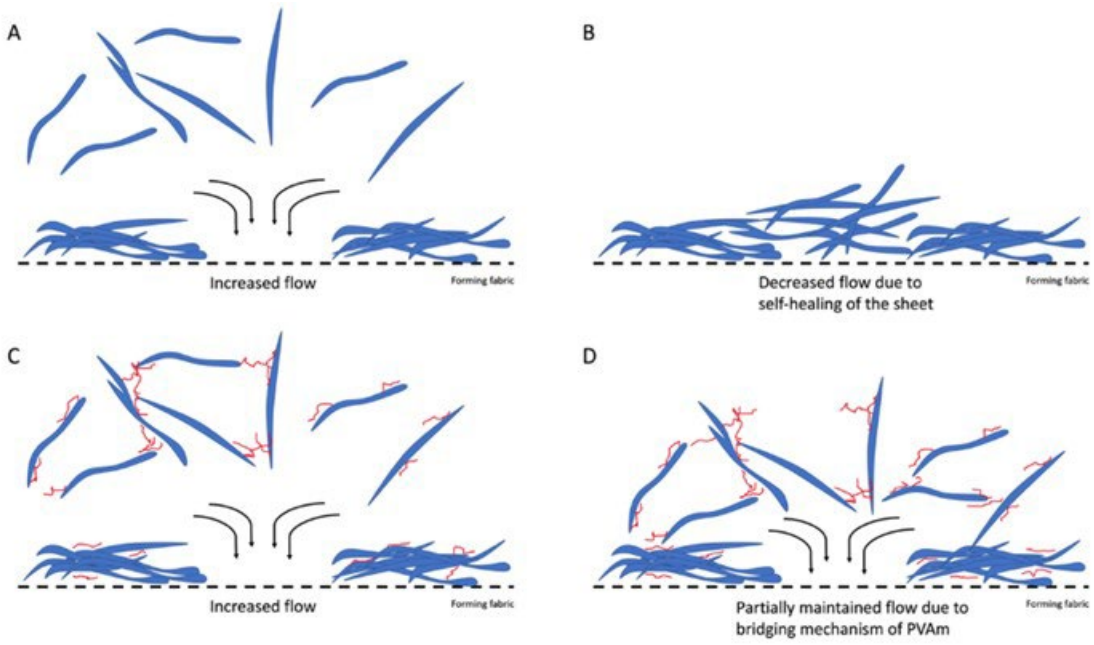

Fig. 8. Diagrams of the self-healing mechanism where a) uncovered patches on the forming fabric increase the outflow of water in those locations, which in turn b) brings more fibers to cover the holes. c) With PVAm present in the early dewatering stage and d) the addition of PVAm is suggested to limit the self-healing and maintain the increased dewatering flow that occur by poor formation and uncovered patches of the forming fabric. Red PVAm molecules not drawn to scale. Redrawn from Hubbe *et al*. (2020).

At longer dwell times (10 to 20 ms), the addition of PVAm resulted in higher dewatering times, which is partly explained by the interaction between the added chemicals and the forming fabrics. At the end of the test series, the forming fabrics were less permeable because of chemical interaction with the PVAm, which naturally resulted in slower dewatering. This does not make the reasoning behind the dewatering mechanisms contradictory, but it is an important observation. When implementing the addition of PVAm in this fashion on an industrial scale, the forming fabrics can be sealed by the chemicals. The loss in permeability can be crucial for production. The sealing behavior of fabrics should therefore be reduced before industrial implementation can be considered. This study did not include such an investigation, but it is possible that the problem can be solved by an extra washing step after the addition of chemicals. Furthermore, the poor formation (Fig. 9), which leads to faster drainage, also could lead to less efficient vacuum dewatering because it will be more difficult to achieve a high vacuum and air will flow more easily through the patches between fiber flocs. This would leave more water in the more flocculated areas. The flocculation behavior causing poor formation due to PVAm addition might be a laboratory effect when the high levels of hydrodynamic shear in the paper machine (Norman 1989) are nonexistent. PVAm additions would be interesting to test at industrial scale to investigate formation issues with high shear forces present.

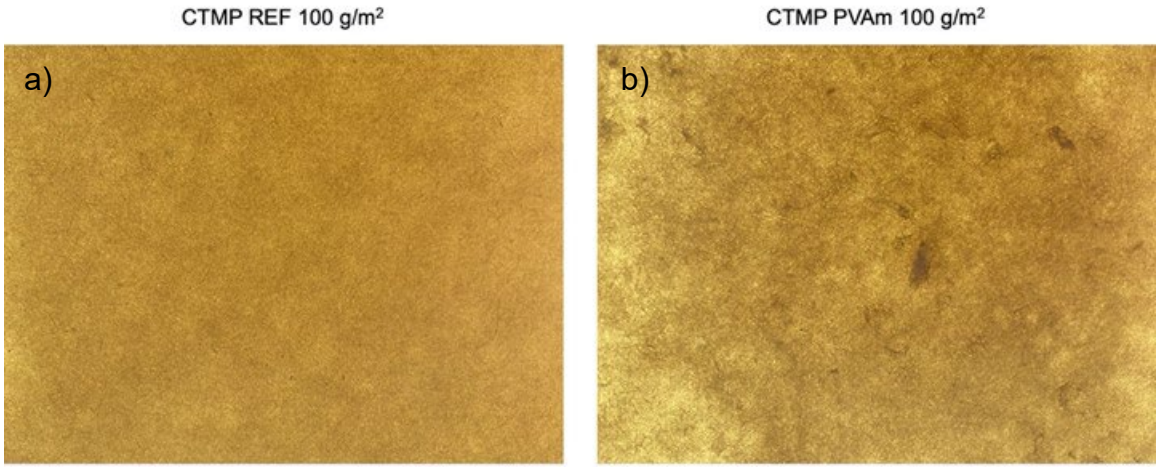

Fig. 9. Photographs of unwashed 100 g/m^2 CTMP sheets a) without PVAm and b) with PVAm to illustrate an example of the formation difference between the reference sheets and the PVAm sheets

The tensile strength results for the kraft and CTMP pulp sheets with and without PVAm are shown in Figs. 10 through 13. No increase in tensile strength index values was found between the reference sheets and the sheets with the PVAm (Fig. 10), even though PVAm is typically considered to be a dry-strength agent. A slight decrease in strength was observed for unwashed CTMP sheets with PVAm. This is explained by the visibly worse formation of sheets with PVAm, which is supported by the large variation in the tensile strength values for the kraft PVAm sheets (Fig. 10). The improved strength properties gained by adding PVAm was counteracted by the worsened formation attributed to maximization of flocculation associated with a charge-neutral system. One of the pulp specimens was washed prior to forming and the resulting sheets had the same tensile stress index as the reference pulp (Fig. 10). The higher tensile strength for the washed PVAm pulp compared with the unwashed is explained by the better formation that is a result of

the washed pulp behaving more similar to the reference pulp than to the unwashed pulp. Washing will remove some of the chemicals in the water that lead to poor formation, but the strength-enhancing properties related to PVAm attached on fibers will remain. The tensile strain decreased for the CTMP with PVAm sheet, but the results stayed within the error margins for the kraft sheets (Fig. 11). The tensile energy absorption index (Fig. 12) and the elastic modulus (Fig. 13) showed similar behavior.

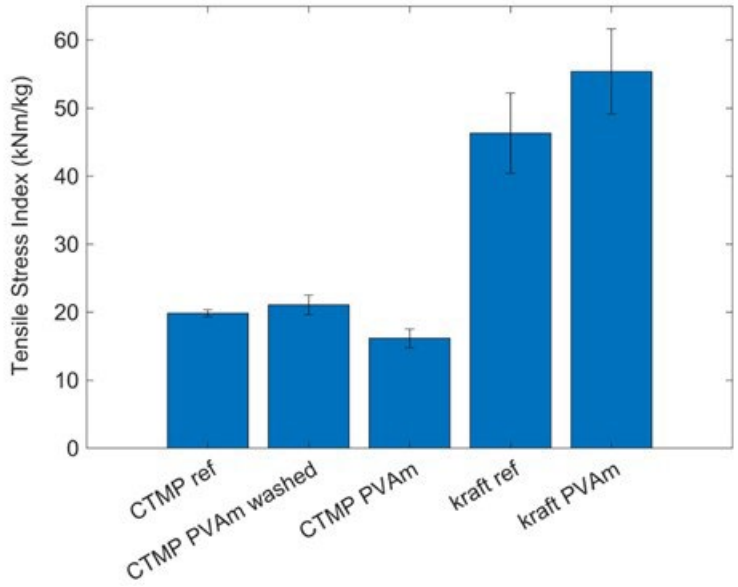

Fig. 10. Tensile stress index (kNm/kg) mean values and 95% confidence intervals for 200 g/m^2 CTMP and kraft sheets with and without the addition of 0.01 g PVAm/g fiber. The CTMP additions were both washed and unwashed according to the description in the Experimental section.

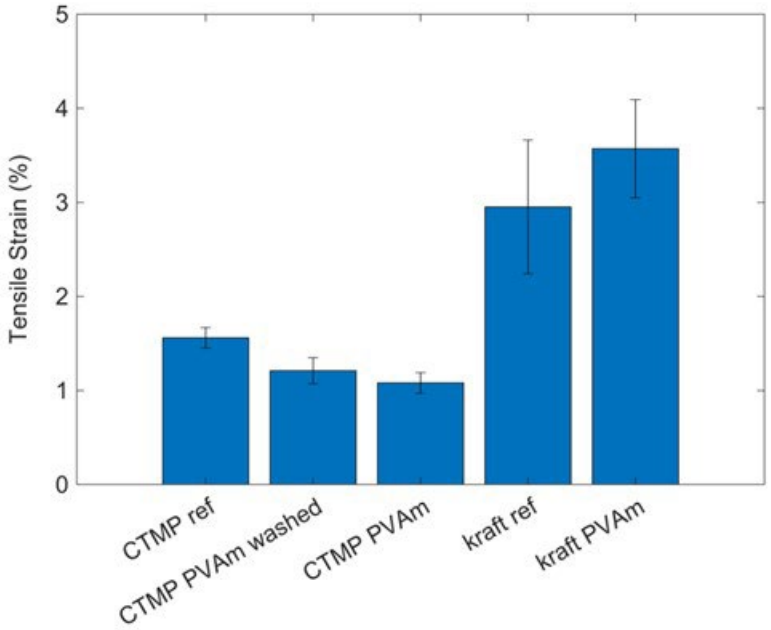

Fig. 11. Tensile strain at break (%) mean values and 95% confidence intervals for the 200 g/m^2 CTMP and kraft sheets with and without the addition of 0.01 g PVAm/g fiber. The CTMP additions were both washed and unwashed according to the description in the Experimental section.

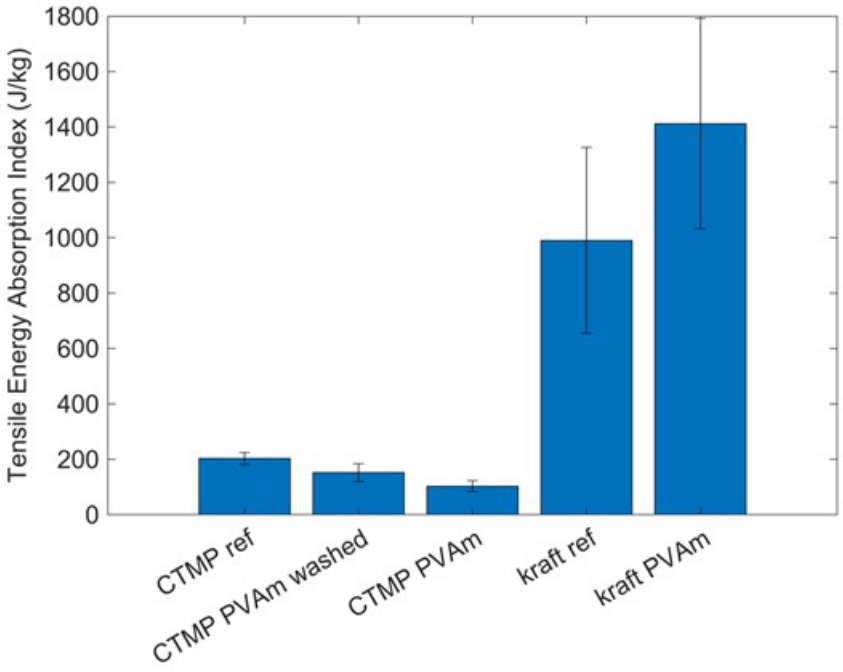

Fig. 12. Tensile energy absorption index (J/kg) mean values and 95% confidence intervals for 200 g/m^2 CTMP and kraft sheets with and without 0.01 g PVAm/g fiber. The CTMP additions were both washed and unwashed according to the description in the Experimental section.

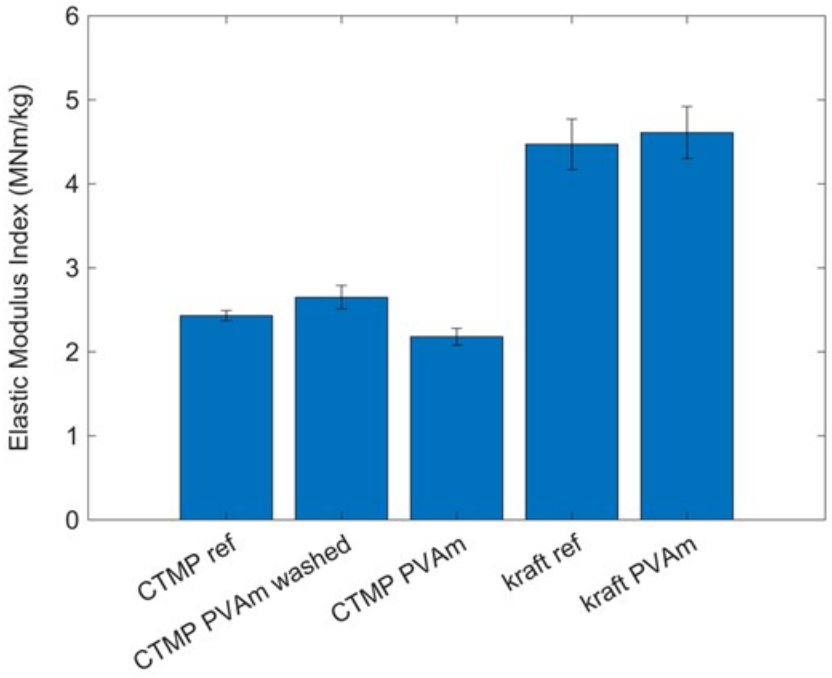

Fig. 13. Tensile stiffness index (MNm/kg) mean values and 95% confidence intervals for the 200 g/m^2 CTMP and kraft sheets with and without the addition of 0.01 g PVAm/g fiber. The CTMP samples were both washed and unwashed according to the description in the Experimental section.

Tables 2 and 3 show the mean values, standard deviations, and confidence intervals for the yield stress, yield strain, elongation at break, and tensile energy absorption.

Table 2. Tensile Strength Measurements of 200 g/m^2 CTMP Sheets with and without PVAm, Washed and Unwashed

CTMP ref 200 g/m^2				
Tensile Property	Tensile strength index (kNm/kg)	Strain at break (%)	Tensile energy absorption index (J/kg)	Tensile stiffness index (MNm/kg)
Mean	19.9	1.56	203	2.43
Standard Deviation	0.86	0.18	33.9	0.09
95% Confidence Interval	0.53	0.11	21.0	0.06
CTMP + PVAm 200 g/m^2 Unwashed				
Tensile Property	Tensile strength index (kNm/kg)	Strain at break (%)	Tensile energy absorption Index (J/kg)	Tensile stiffness index (MNm/kg)
Mean	16.2	1.08	103	2.18
Standard Deviation	2.25	0.17	32.6	0.16
95% Confidence Interval	1.39	0.11	20.2	0.10
CTMP + PVAm 200 g/m^2 Washed				
Tensile Property	Tensile strength index (kNm/kg)	Strain at break (%)	Tensile energy absorption index (J/kg)	Tensile stiffness index (MNm/kg)
Mean	21.1	1.21	152	2.65
Standard Deviation	2.33	0.22	51.6	0.22
95% Confidence Interval	1.44	0.14	32.0	0.14

Table 3. Tensile Strength Measurements of 200 g/m^2 Kraft Sheets with and without PVAm

Kraft 200 g/m^2				
Tensile Property	Tensile Strength Index (kNm/kg)	Strain at Break (%)	Tensile Energy Absorption Index (J/kg)	Tensile Stiffness Index (MNm/kg)
Mean	46.3	2.95	990	4.47
Standard Deviation	9.50	1.14	541	0.48
95% Confidence Interval	5.89	0.71	335	0.30
Kraft + PVAm 200 g/m^2				
Tensile Property	Tensile Strength Index (kNm/kg)	Strain at Break (%)	Tensile Energy Absorption Index (J/kg)	Tensile Stiffness Index (MNm/kg)
Mean	55.4	3.57	1,410	4.61
Standard Deviation	10.1	0.84	613	0.50
95% Confidence Interval	6.27	0.52	380	0.31

The air permeance according to ISO 5636-3 (2013), using 0.7 kPa instead of 1.47 kPa, is shown for single and double sheets in Figs. 14 and 15. The air permeance clearly shows that addition of PVAm in all cases gave a more open sheet structure that allowed more air to flow through.

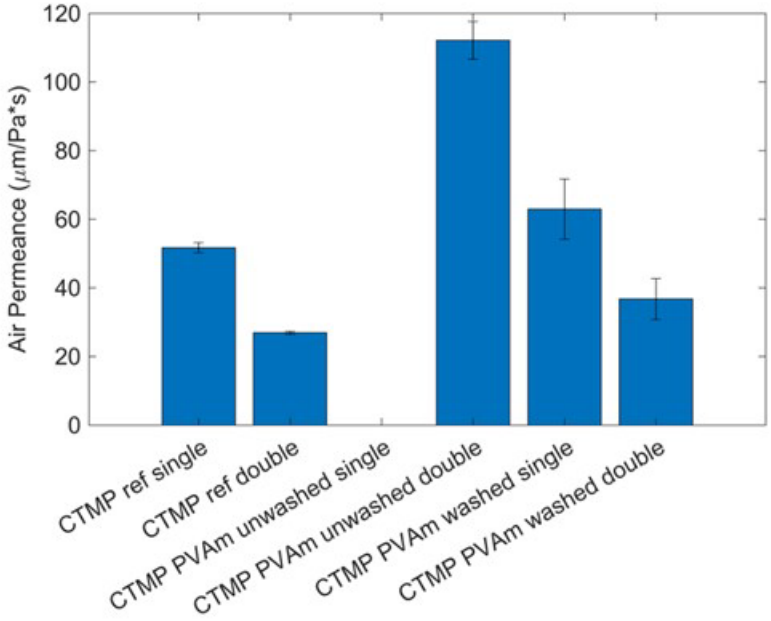

Fig. 14. Air permeance with 0.7 kPa instead of 1.47 kPa for single and double sheets of CTMP with and without 0.01 g PVAm/g fiber (washed and unwashed). The CTMP + PVAm unwashed single sheets were not measurable because the flow in the machine was too high. Error bars represent 95% confidence interval.

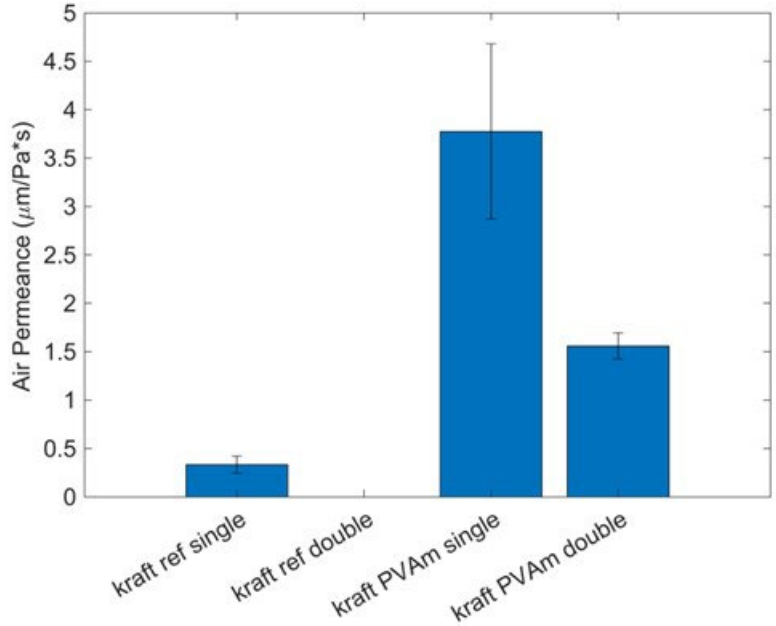

Fig. 15. Air permeance with 0.7 kPa instead of 1.47 kPa is shown for single and double sheets of kraft pulp with and without 0.01 g PVAm/g fiber. The kraft ref double sheet sample had zero flow. Error bars represent 95% confidence interval.

Higher air permeance is presumably strongly linked with increased flocculation, which led to poorer formation of the PVAm sheets compared with the reference sheets, and with bonding of fine materials to the fiber surfaces. In both cases, the resulting sheet would be more open for flowing air. Figure 13 shows that the washed pulp had a smaller effect on air permeance, agreeing with the observations related to formation from tensile testing, in which most of the chemicals in the water were removed. The proposed mechanism in Fig. 8d supports the idea that higher air permeance is connected to PVAm in the water and attached to the fiber surfaces.

CONCLUSIONS

1. The initial dewatering was observed to be faster for pulps with added polyvinylamine (PVAm). This effect was greater for chemithermomechanical pulp (CTMP) and higher basis weights. The enhanced dewatering also depends on the bonding mechanism of the PVAm and its effects on self-healing and plugging of flow channels.
2. The strength was increased when PVAm was added, but only if the pulp was washed before sheet forming. Unwashed pulp with PVAm led to poorer formation, which resulted in lower sheet strength, even if local areas could be stronger because of the presence of PVAm.
3. The air permeance was higher for sheets made from unwashed pulps with the addition of PVAm, as the presence of PVAm limits the self-healing and plugging of flow channel mechanisms during dewatering.
4. The wet-end addition of PVAm worked well for both CTMP and kraft pulps under the conditions used in this study.

The addition of PVAm to the stock suspension in the wet-end of the paper machine has great potential for applications that require high air permeance of the product. Polyvinylamine could also work as a dewatering enhancing agent, but caution must be taken regarding the potential of formation problems.

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