

Suspension-polymerized Latex as an Additive for Surface Sizing and Its Effect on Fold Cracking of Coated Paper

Araz Rajabi Abhari,^a Hak Lae Lee,^{a,b,*} Kyudeok Oh,^a Wanhee Im,^a Jee-Hong Lee,^a Sooyoung Lee,^c and Songju Kim^d

As an approach to decrease the fold cracking of coated paper, suspension-polymerized (SP) latexes were developed and tested as a surface sizing additive. Styrene, ethyl acrylate, butyl acrylate, acrylic acid, and itaconic acid were used as monomers for the SP latex, and oxidized starch and polyvinyl alcohol were used as stabilizers. The SP latexes were found to be more stable against the charge neutralization by salt solutions and flocculation by cationic polyacrylamide than conventional styrene-butadiene and styrene-acrylate latexes, and they were highly compatible with the conventional surface sizing solution. The effect of using SP latexes as a surface sizing additive on the mechanical properties of the paper was examined. The SP latexes had greater tensile strength and extensional properties than the emulsion-polymerized latexes, which suggested their potential applicability for reducing the fold cracking of coated paper. A mill trial was performed to test the use of SP latexes as a surface sizing additive, and the results showed that they had a positive effect in reducing the fold cracking of coated paper.

Keywords: Latex; Suspension polymerization; Surface sizing; Additive; Fold cracking

Contact information: a: Program in Environmental Material Science, Department of Forest Science; b: Research Institute of Agriculture and Life Sciences, College of Agriculture and Life Sciences, Seoul National University, 08826, Seoul, Korea; c: Songkang Industrial Co. Ltd., Samseong-myeon, Eumseong-gun, Chungbuk-do, Korea; d: Moorim Paper Co. Ltd., 1003 Namgang-ro, Jinju-si, Gyeongsangnam-do, Korea; *Corresponding author: lhakl@snu.ac.kr

INTRODUCTION

Fold cracking of coated paper is one of the most frequent and troublesome problems affecting the use of coated paper and boards. Fold cracking is becoming more common in the paper industry because more filler is being used in papermaking and heavier coatings are applied to substitute the use of expensive fibers with pigment coating (Seo *et al.* 2013). Fold cracking occurs when coated paper and paperboard are folded or creased during the converting operation. The issue of fold cracking most often arises when the tensional or compressional stress or strain exceeds the level that the coated product can withstand.

Many studies have investigated the factors that affect the fold cracking of coated papers, such as the fiber composition, coating pigments and binders, coating layer structure, and folding direction. Sim *et al.* (2012) evaluated the influence of the fiber composition and showed that fold cracking of coated paper was minimized when the base paper consisted of 90% hardwood fiber and 10% softwood fiber. It was also shown that beating exacerbated fold cracking, even though it increased the tensile strength. Clay has been found to result in greater fold cracking than ground calcium carbonate (GCC) (Rättö and Hornatowska 2010). The cracks in clay-based coatings can be initiated anywhere along the

thickness direction in the coating layer and continue at an angle through the thickness direction. In contrast, GCC-based coatings become cracked specifically on the surface of the coating layer and the crack then propagates through the thickness in the Z-direction of the coating.

An increase in the starch content used as a binder increases the modulus of elasticity (MOE) and decreases the strain of the coating (Okomori *et al.* 2001; Rioux *et al.* 2011; Oh *et al.* 2015). This change in mechanical properties influences the fold cracking of the coating layer. The use of starch as a coating binder has been found to increase the fold crack area, while decreasing the total number of fold cracks (Oh *et al.* 2015). This effect contributed to the greater occurrence of strain-based failure of coatings with higher starch binder contents. The importance of the extensional strain property of the coating layer in decreasing the fold cracking has been shown by Okomori *et al.* (2001) and Oh *et al.* (2015). The use of latex with a high glass transition temperature or starch as the binder increased the MOE, while decreasing the strain of the coating layer, which eventually led to a greater length, width, and area of the cracks in the coating after folding.

To minimize fold cracking, a method to improve the extensional property and tensile strength of the base sheet and coating layer is necessary. Although many studies have been done to improve the coating layer properties and make coatings more resistive to fold cracking, few approaches have tried to make the base stock more forgiving to the folding and converting operation. Starch is widely used for surface sizing of coating base stocks because it has many advantages for improving the tensile strength, burst strength, and stiffness of the coating base stock. However, the use of starch as a coating binder is one of the major causes of fold cracking for coated products because of its stiffness and low resilience properties. A question has been raised as to whether the use of starch in surface sizing and not as a coating binder would have a detrimental effect during the folding operation of coated papers. If this is so, another question is raised as to whether or not it is possible to reduce the fold cracking problem by using an additive that provides a good extensional property for surface sizing.

Various materials, including water soluble polymers and latexes, have been used as additives for surface sizing. Polyvinyl alcohol (PVOH), sodium alginate, polyacrylamide (PAM), carboxymethyl cellulose, and other compounds have been used along with starch to improve the film-forming properties and modify the viscosity of surface sizing solutions. Seo *et al.* (2013) showed that low-molecular weight PVOH was effective at improving the folding endurance and internal bond strength without causing an increase in the viscosity that is associated with high-molecular weight polyDADMAC. Cationic PAM (Jeon and Lee 2003; Seo *et al.* 2015) and cationic starch (Lee *et al.* 2002) have been used to increase the interaction of the surface sizing solution with papermaking fiber, and thereby limit the penetration of surface sizing solutions into paper. Dextrin-poly(acrylic acid) copolymer has been examined as a surface sizing additive to improve paper properties (Kim *et al.* 2017; Rajabi Abhari *et al.* 2017). The use of water soluble polymers often results in an increasing starch solution viscosity, which limits the solids level of surface sizing solutions (Jeong *et al.* 2013).

Latexes have advantages as surface sizing additives because they do not increase the solution viscosity, which allows for an increasing solids level in sizing solutions. It is also possible to impart a good extensional property to the latex by using appropriate monomers. There are some prerequisites for latex as a sizing additive. First, it should be compatible and stable for use with a starch solution (Bacquet and Isoard 1997). Furthermore, it should provide sufficient strength and extensional properties to resist stress

and strain during folding and cracking. A latex binder with a low glass transition temperature has been employed to reduce the fold cracking of the coating layer because latex with a low glass transition temperature increases the stretch of the pigment coating layer (Okomori *et al.* 2001; Oh *et al.* 2015). It has been determined that the use of styrene-acrylate (S/A) latex decreases the fold cracking of coated paper because it has a greater extensional property than styrene-butadiene (S/B) latex (Oh *et al.* 2016). Good stability against changes to the pH and conductivity is important as well because the contact or immersion of paper web with a sizing solution causes changes in the pH and conductivity of the surface sizing solution.

In this study, suspension-polymerized (SP) latexes were prepared and tested as surface sizing additives. The stability of the sizing solutions with a change in the salt concentration and with the addition of cationic PAM were investigated and compared with that of conventional emulsion-polymerized latexes. Moreover, the effect of the SP latexes on the mechanical properties of the surface sized papers was examined. Finally, a mill trial was performed to examine the effect of using SP latex as a surface sizing additive to decrease the fold cracking of coated paper.

EXPERIMENTAL

Suspension-polymerized Latex

The SP latexes were synthesized using styrene, ethyl acrylate, butyl acrylate, acrylic acid, and itaconic acid as monomers. Oxidized starches and PVOH were used as stabilizers for suspension polymerization. The details of the polymerization method are beyond the scope of this paper. Two SP latexes, namely SP-1 and SP-2, prepared in the laboratory and a SP-M latex synthesized in a pilot scale reactor were compared with two commercial emulsion-polymerized latexes, *i.e.*, S/B and S/A latexes (LG Chemical, Daejeon, Korea). The properties of the latexes used in this investigation are shown in Table 1.

A commercial oxidized starch (C 3011) was provided by Samyang Genex (Seongnam, Korea). Cationic PAM in powder form with a molecular weight of approximately 6 Mg/mol was provided by BASF (Seoul, Korea) and used as a flocculant. Fine paper with a basis weight of 85 g/m² from Moorim Paper (Jinju, Korea) was used as a base paper for surface sizing.

Table 1. Properties of the Latexes

Latex	Solids (%)	pH	Average Particle Size (nm)	Viscosity (cP)
S/B	49.5	7.9	122	190
S/A	49.5	7.3	111	366
SP-1	34.3	8.1	81	27
SP-2	35.4	6.8	95	18
SP-M	35.0	7.4	70	23

Methods

The effect of the salt concentration on the stability of the latex dispersions was studied by measuring the zeta potential and particle size distribution at different NaCl concentrations. The zeta potential and particle size were measured using a Malvern Zetasizer (Nano-ZS, UK). For this experiment the latex dispersions were diluted with deionized water to 0.1% in the NaCl solutions, which had concentrations that ranged from 0 M to 4 M. To study the effect of polyelectrolytes on the stability of the latexes, a cationic PAM was used. A solution with 1% cationic PAM was prepared and added to the latex dispersion at dosages of 0.005%, 0.01%, 0.05%, and 0.1% based on the weight of the latex dispersion. The latex and cationic PAM were mixed at a stirring speed of 400 rpm for 5 min and the low-shear viscosity (Brookfield viscometer, DV-2, USA), and particle size were measured.

A surface sizing solution at 16 wt.% was prepared by cooking the starch slurries at 95 °C for 30 min. Then, the oxidized starch paste was cooled down to 65 °C and 3 parts or 5 parts of latex per 100 parts of starch (pph) were added to the starch solution. Surface sizing was done using an automatic bar coater (GIST Co. Ltd., Daejeon, Korea) on the base paper. The sized paper was dried in a hot-air drying oven at 120 °C for 2 min. The surface sized papers were conditioned at a constant temperature of 23 °C and a relative humidity of 50% for at least 24 h. The tensile strength, bending resistance, and internal bond strength were determined according to TAPPI standards (T 1009 om-92, T 556 pm-95, T 569 pm-00).

To investigate the effect of the latex addition on the tensile property of the starch film, an oxidized starch solution at a concentration of 16 wt.% solids was placed in a polystyrene dish and dried in a drying oven at 50 °C under vacuum at 0.009 MPa. Then, the dried starch film with a thickness of 100 µm was carefully separated and cut for the tensile strength measurement. The width and length of the samples were 15 mm and greater than 40 mm, respectively. The span length for the tensile testing was 30 mm, and the testing speed was 3 mm/min. Additionally, dried starch films containing 5 pph of S/B or SP latexes were prepared and tensile tested.

Mill Trial

A mill trial was conducted to study the effect of using the SP-M latex as a surface sizing additive. This work was done at Moorim Paper Co. (PM1, Jinju, Korea) on a Fourdrinier type paper machine with a conventional inclined size press. The fiber composition of the base paper was 84% bleached hardwood kraft pulp, 4% bleached softwood kraft pulp, and 12% bleached chemi-thermomechanical pulp. The oxidized starch solution was cooked in a jet cooker and 2.9 pph of SP-M latex was added to the cooked starch solution and applied to the base paper with a basis weight of 198 g/m². A high-grammage grade was used for the trial run because this would show the fold cracking phenomena more clearly. The mechanical properties of the sized papers were measured according to TAPPI standards. To investigate the distribution of the surface sizing starch in the Z-direction of the sized papers, cross-sectional samples of the sized papers were prepared using razor blades. Then, the samples were immersed in a 0.1-M iodine potassium iodine solution for staining according to the method described by Lipponen *et al.* (2004). The cross-sectional images of the samples were obtained using a video microscope (i Megascop 1080 P, Alphastec Co., Anyang, Korea). The precoating color was prepared using coarse-grade GCC as a pigment. Additionally, 6 pph of S/B latex and 5 pph of starch were used as binders. The top coating color was prepared using 90 parts of fine-grade GCC

(Setacarb HG, Omya Korea) and 10 parts of clay (HYDROGLOSS® 90, KaMin) as the pigments. The S/B latex was used as a binder, along with some additives. The addition rate of the S/B latex was 7 pph. The coat weight was 26 g/m² for each side. After offset printing (Mitsubishi Offset Press, Mitsubishi Heavy Industry, Tokyo, Japan), the coated papers with a black ink, a commercial folding machine (Stahlfolder Ti52, Heidelberg, Heidelberg, Germany) was used for folding. Images of the folded paper stack were taken for analysis. The fold cracked area of the paper was analyzed using Image Plus Pro software (Ver. 4.1, Media Cybernetics, Inc., Rockville, MD, USA).

RESULTS AND DISCUSSION

Chemical Stability

Emulsion polymerization is a method widely used for the preparation of S/B and S/A latexes. Because surfactants with charged functional groups are used for the stabilization of emulsion-polymerized latexes, it is possible to keep latex particles dispersed for several months. However, the stability derived from electrostatic repulsion would be lost if the latex dispersion is exposed to an environment containing counter ions, which neutralize electrostatic charges. It was shown that the stability of emulsion-polymerized latex is lost and coagulation or gelation occurs when the pH of the latex is decreased to acidic conditions. In contrast, SP latexes maintain their stability over a wide range of pH conditions because they are stabilized not by electrostatic repulsion, but through steric hindrance by the adsorbed polymers. The chemical stability of the latex particles is affected not only by the pH change, but also by the addition of polyelectrolytes.

The flocculation phenomena of the emulsion-polymerized latex have been investigated by many research groups. Eriksson *et al.* (1993) have shown that latex particles are flocculated by cationic polymers that are adsorbed either through bridging between the latex particles or by charge neutralization. Feng *et al.* (2015) investigated the rate of flocculation of emulsion-polymerized latex particles and reported that the maximum flocculation rate was obtained immediately after the onset of mixing, and the flocculation rate gradually slowed down as adsorption progressed. They also showed that the initial rate of flocculation was minimized as the ionic strength increased.

The effect of the cationic PAM addition on the particle size and viscosity of the SP-1 and S/B latexes is depicted in Fig. 1. The addition of cationic PAM increased the viscosity of both latexes. In the case of the S/B latex, the viscosity increase was so substantial that it was not possible to measure the viscosity when the cationic PAM addition level was greater than 0.01%, which was attributed to the extensive flocculation of the latex by polymer bridging. The addition of cationic PAM to the SP-1 latex dispersion also resulted in flocculation, but the steric stabilization of the SP latex prevented the formation of very large flocs. It has been known that steric stabilization provides the advantages of pH insensitivity and improved freeze-thaw stability (Ottewill *et al.* 1987).

The same phenomena occurred when the salt concentration increased. The effect of the salt concentration on the zeta potential of the latex is depicted in Fig. 2. The zeta potential of the SP latexes was less negative than that of the S/B and S/A latexes when there was no salt addition because the stabilization by low-charged polyelectrolytes was the dominant mechanism for the SP latexes. With the addition of salt, the zeta potential decreased and approached the isoelectric point.

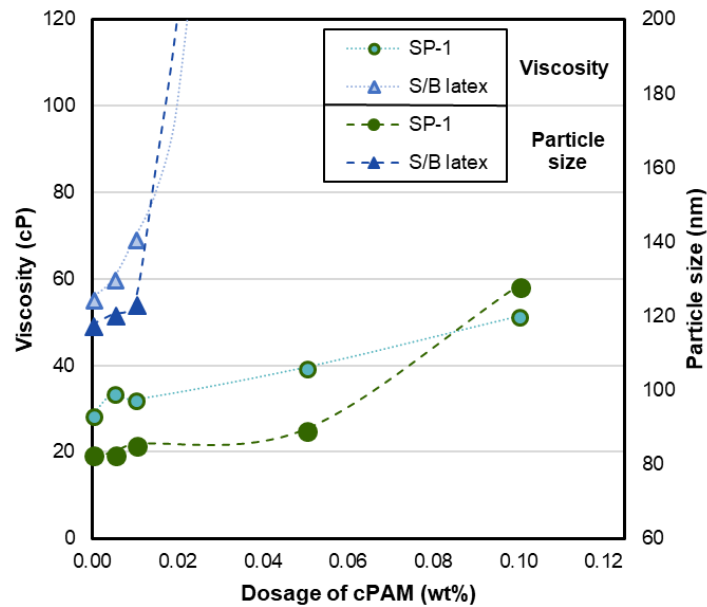


Fig. 1. Effect of the cationic PAM on the viscosity (open symbols) and particle size (closed symbols) of the S/B (▲) and SP-1 (●) latexes

The change in the zeta potential was more abrupt for the SP latexes, which indicated that a lesser amount of counter ions is required for charge neutralization. The particle size distribution data of the S/B and S/A latexes showed that severe agglomeration of these latexes occurred at 4 M NaCl (Fig. 3). However, the particle size distribution curves for the SP latex showed that less agglomeration occurred (Fig. 4), which was attributed to the steric stabilization mechanism of the SP latex.

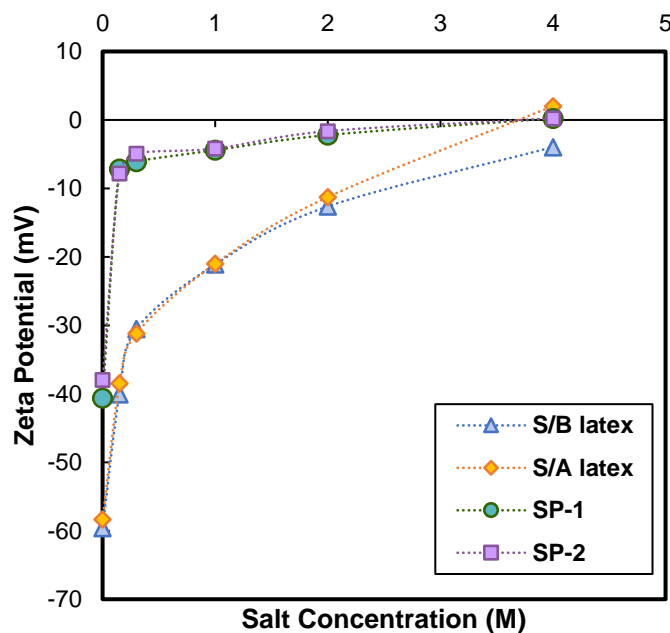


Fig. 2. Zeta potential of the latexes as a function of the salt concentration

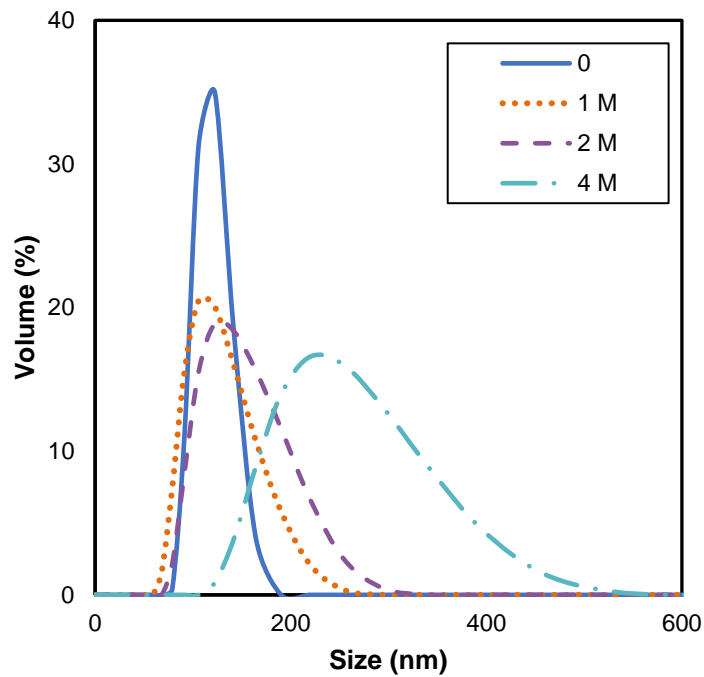


Fig. 3. Particle size distribution (PSD) curves for the S/B latex at four different salt concentrations

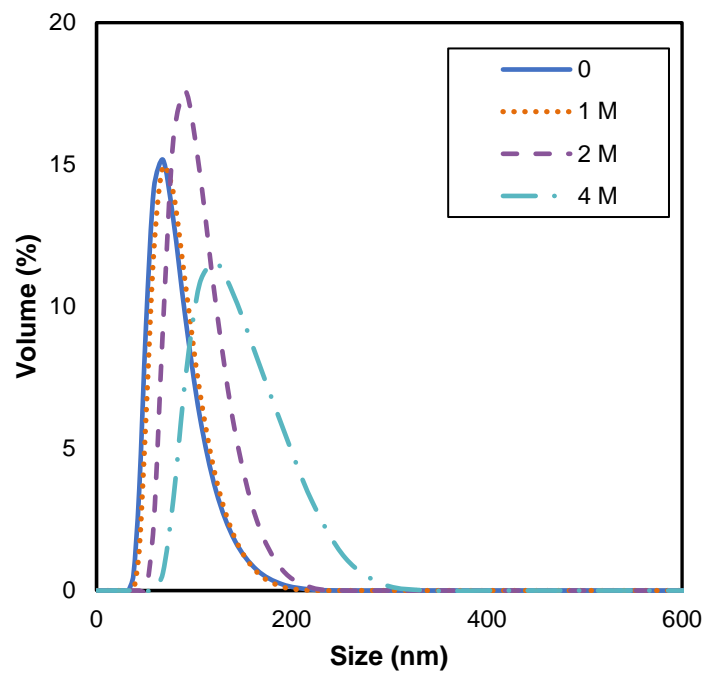


Fig. 4. Particle size distribution (PSD) curves for the SP-2 latex at four different salt concentrations

Ishikawa *et al.* (2005), who investigated the effect of pH and salt concentration on the zeta potential, also showed that the total potential energy of interaction between pairs of latex particles changed by altering the salt concentration and pH and found that the steric stabilization provided greater stability for the latex. The chemical stability of the latex particles is important because the salt or polyelectrolytes contained in the base paper dissolves into the sizing solution when the hot paper web passes through the hot sizing solution or comes into contact with the surface sizing films.

Film Properties

The solution stability and film properties of the surface sizing agent are critical to obtain the desirable effect of surface sizing. To examine the strength and stretch properties of starch films without the influence of the base paper, films were prepared by drying the sizing solutions in petri dishes. By doing this it was possible to compare the film properties without variability caused by the base sheet. Figure 5 shows the tensile stress, elongation at break, and MOE of the starch films prepared with and without the latex additive. The addition of 5 pph of the S/B and SP-M latexes increased the tensile strength and elongation at break. The SP-M latex showed greater improvement in both the tensile strength and elongation at break. This showed that the use of SP-M latex would have a positive effect in improving the tensile and elongation properties of paper after surface sizing, which was attributed to the monomer composition of the SP-M latex. Oh *et al.* (2016) found that S/A latex has a greater extensional property than the S/B latex, which is advantageous in decreasing the fold cracking of coated paper.

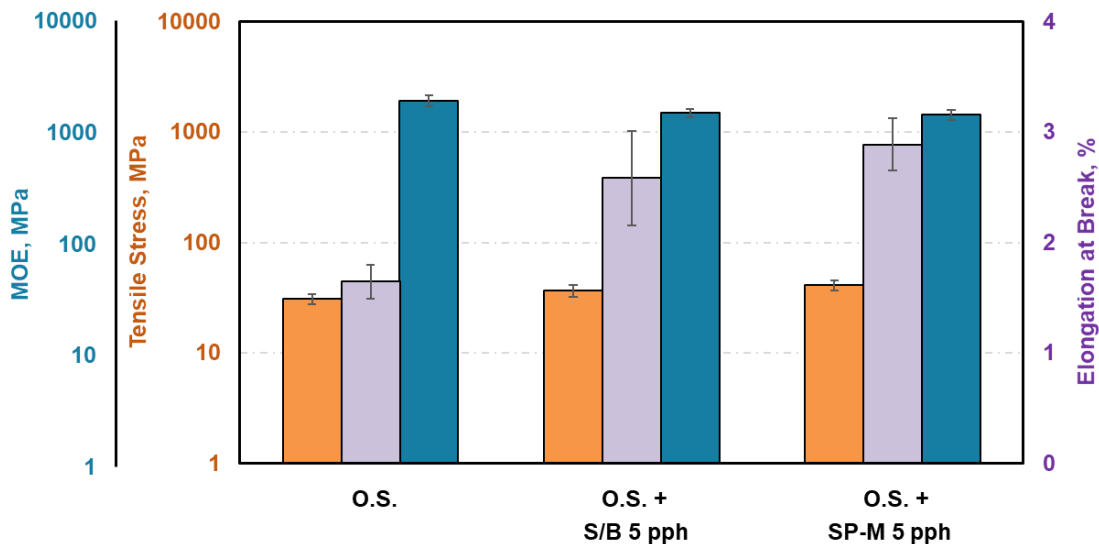


Fig. 5. Tensile stress, elongation at break, and MOE of the starch films prepared with 5 pph of S/B and SP-M latex; O.S. – oxidized starch

Surface Sized Paper

To test the effect of latex addition on the paper properties, 16% starch solutions with and without the latex additive were prepared and applied onto a base paper using the laboratory bar coater. The pickup weight was 3.3 g/m² for all of the samples. The viscosity of the starch solutions was 30.1 cPs, and it changed to 28.0 cPs and 26.5 cPs when 5 pph

of the S/B and SP-M latexes were added, respectively. Figure 6 shows that the tensile and elongation properties of the surface sized paper (machine direction) increased after surface sizing with starch. When 5 pph of SP-M latex were used, the tensile stress increased further. In contrast, the addition of S/B latex decreased the tensile stress. The same trend was obtained for the elongation at break. The MOE of the sized papers decreased with the use of SP-M latex, which indicated that the SP-M latex improved the flexibility of the paper after surface sizing. This was because the SP-M latex, which had a lower gel content, stretched more under tension. The lower gel content of the SP latex was attributed to the substitution of butadiene monomer with acrylate monomer. Increases in the tensile stress and elongation at break are highly favorable for decreasing fold cracking (Oh *et al.* 2015).

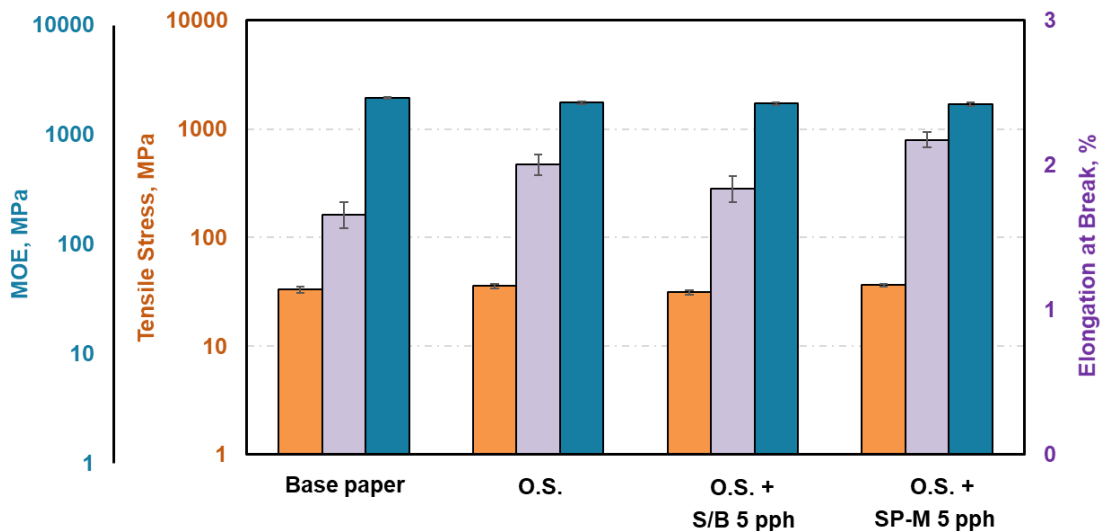


Fig. 6. Tensile stress, elongation at break, and MOE of the base paper and sized papers with O. S. (oxidized starch) and 5 pph of the S/B and SP-M latexes

Because the SP latex showed substantial improvement in the tensile strength and elongation at break at the 5-pph addition rate, the possibility of reducing the SP latex dosage was tested. Table 2 shows the tensile index, bending stiffness, and internal bond strength of the paper after surface sizing with 3 pph and 5 pph of SP-M latex as the surface sizing additive. The 3-pph dosage showed similar levels of improvement in the tensile strength and bending stiffness as at 5 pph.

Table 2. Properties of the Surface Sized Paper with SP-M Latex

	Oxidized Starch Only		O.S. + 3 pph SP-M		O.S. + 5 pph SP-M	
	MD	CD	MD	CD	MD	CD
Tensile Index (Nm/g)	51.1	33.9	55.9	34.6	54.2	33.7
Bending Stiffness (mNm)	1.7	1.0	1.8	1.0	1.8	0.9
Internal Bond Strength (J/m^2)	-	176.4	-	181.6	-	205.3

MD – machine direction; CD – cross-machine direction

Mill Trial and Fold Cracking

The laboratory experiments showed that the addition of SP-M latex did not negatively affect the viscosity, chemical stability, and foaming. Table 3 shows the properties of the coating base stocks that were produced from PM1 by Moorim Paper Co. The paper was surface sized either with conventional oxidized starch or with a starch solution containing 2.9 pph of SP-M. The bulk density of the surface sized papers with and without SP-M were 1.27 cm³/g.

The tensile strength and stretch increased when SP-M was used as a surface sizing additive, which decreased the stiffness in both the machine direction (MD) and cross-machine direction (CD). The internal bond strength also increased when SP-M was added to the sizing solution. This indicated that some reduction in the fold cracking may be achieved by using SP-M as a surface sizing additive.

Table 3. Properties of the Base Paper Surface Sized with and without SP-M Latex

Property	Oxidized Starch Only		2.9 pph of SP-M	
	MD	CD	MD	CD
Tensile Strength (kN/m)	7.93	5.57	9.10	5.38
Stretch (%)	2.54	7.95	3.55	8.06
Stiffness (g·cm)	52.5	34.7	46.9	26.1
Internal Bond Strength (J/m ²)	118.6	124.9	122.7	127.3

The cross-sectional images of the papers sized with oxidized starch only and oxidized starch with 2.9 pph of SP-M latex were obtained using the microscope imaging system (Fig. 7). The addition of SP-M to the oxidized starch resulted in deeper penetration of the sizing solution into the base paper. This was attributed to the reduction in the viscosity of the sizing solutions with the latex addition. Deeper penetration of the sizing solution usually increases the tensile and internal bond strengths (Lipponen *et al.* 2003; Rajabi Abhari *et al.* 2017).

The fold cracking was evaluated for the paper double-coated on an off-machine coater. The same coating color, coating condition, and coat weight were used in the trial production. The only difference was the use of SP-M latex as a surface sizing additive for the base paper. Solid printing with black ink was done on the double-coated paper using the offset printing machine, and then the folding of the printed papers using the commercial folding machine was done. Images of the folded areas were obtained, and the fold crack area was determined.

Table 4 shows that the trial samples, which were surface sized with 2.9 pph of SP-M latex as a surface sizing additive, showed less fold crack areas in both the MD and CD. When 2.9 pph of SP-M latex was used, the fold crack area in the MD decreased by 16% from 20.6% to 17.3%. In the case of CD folding, the reduction in the fold cracking was 27.5%.

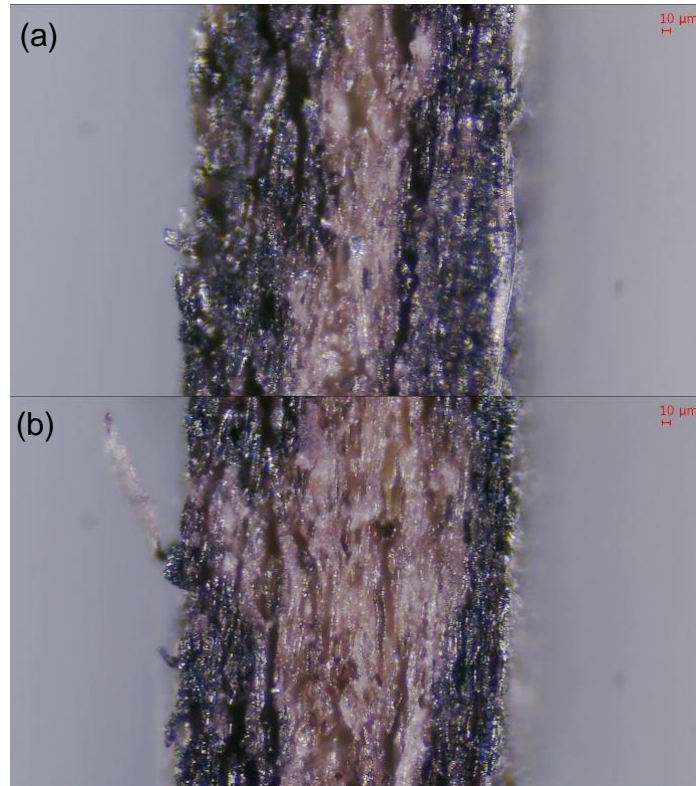


Fig. 7. Microscope images obtained from the cross-section of papers sized with only oxidized starch (a) and oxidized starch containing SP-M latex (b)

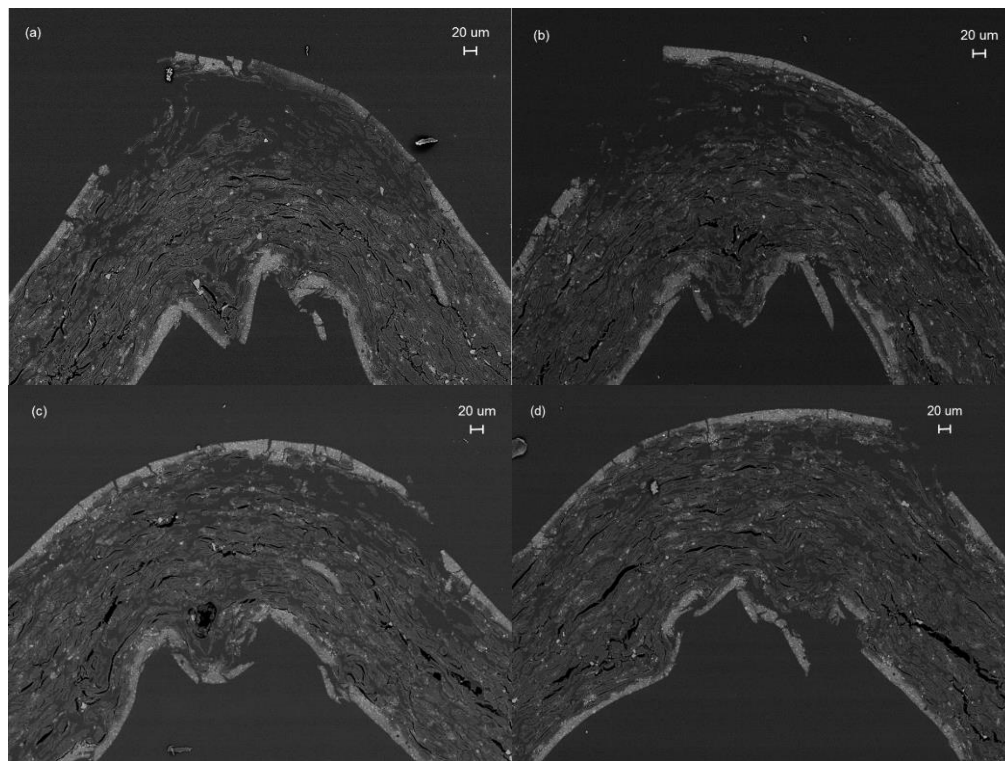


Fig. 8. SEM images of the fold cracked papers: (a and b) folded coated papers that were surface sized with starch only; (c and d) folded coated papers that were on the trial base stock surface sized with starch and 2.9 pph of SP-M latex

Table 4. Percentage of Fold Crack Areas and Reduction Percentage in the MD and CD

Property	MD		CD	
	Control	Trial	Control	Trial
Fold Crack Area (%)	20.6	17.3	15.3	11.1
Reduction (%)	16.0		27.5	

The cross-sectional images of the folded samples were obtained and are shown in Fig. 8. Large coating layers were detached from the base stock surface sized with starch only. In contrast, the coating layer on the base stock surface sized with SP-M latex as an additive showed no large detachment of the coating layer. This was likely because of the improved flexibility of the starch layers on the base paper.

CONCLUSIONS

1. Suspension-polymerized latexes were stabilized with a protective layer containing starch and polyvinyl alcohol (PVOH). These hydrocolloids provide a favorable chemical stability against polyelectrolytes and salts through steric hindrance.
2. The suspension polymerized (SP) latexes were prepared and tested as surface sizing additives. The trial results suggested that an improvement in the tensile strength and extensional property could be achieved with the use of SP latex as a surface sizing additive.
3. A mill trial was performed to verify the effect of using SP latex as a surface sizing agent. The tensile strength and extensional property were found to improve, while the stiffness decreased after surface sizing with SP-M latex (see Table 1).
4. It was indicated that the use of SP-M would provide an opportunity to reduce the fold cracking after coating. This was confirmed by a trial coating, in which coated paper surface sized with SP-M greatly improved the resistance to fold cracking.

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