

Cracking at the fold in double layer coated paper: the influence of latex and starch composition

SEYYED MOHAMMAD HASHEMI NAJAFI, DOUGLAS W. BOUSFIELD, AND MEHDI TAJVIDI

ABSTRACT: Cracking at the fold of publication and packaging paper grades is a serious problem that can lead to rejection of product. Recent work has revealed some basic mechanisms and the influence of various parameters on the extent of crack area, but no studies are reported using coating layers with known mechanical properties, especially for double-coated systems.

In this study, coating layers with different and known mechanical properties were used to characterize crack formation during folding. The coating formulations were applied on two different basis weight papers, and the coated papers were folded. The binder systems in these formulations were different combinations of a styrene-butadiene latex and mixtures of latex and starch for two different pigment volume concentrations (PVC). Both types of papers were coated with single and double layers. The folded area was scanned with a high-resolution scanner while the samples were kept at their folded angle. The scanned images were analyzed within a constant area. The crack areas were reported for different types of papers, binder system and PVC values.

As PVC, starch content, and paper basis weight increased, the crack area increased. Double layer coated papers with high PVC and high starch content at the top layer had more cracks in comparison with a single layer coated paper, but when the PVC of the top layer was low, cracking area decreased. No measurable cracking was observed when the top layer was formulated with a 100% latex layer.

Application: By understanding how the mechanical properties of two coating layers interact to generate cracking, coatings can be designed to minimize the cracking issue. This work suggests that cracking could also be minimized if, during converting, a latex rich overcoat was applied to regions that are folded.

Coated paper or paperboard is subject to different mechanical stresses, such as tension, during printing and bending during folding. One serious defect is called crack at the fold, where cracks in the coating layer form along the folding region of a folded product. While the understanding of this defect has improved in recent years, more work is needed to understand the influence of the mechanical properties of the paper and the coating layer on the cracking problem.

Many studies have been reported that evaluate the mechanical properties of paper coatings. For example, the shape of pigment particles influences the in-plane tensile and the z-direction strength of the coating layer [1-2]. The effect of latex binder content on the mechanical properties of coating is reported by other researchers [3-4]: increasing the pigment volume concentration (PVC) in the coating formulations showed tensile strength increased up to the critical PVC (CPVC), beyond which tensile strength decreased. These changes showed different mechanisms are involved before and after CPVC. Composite models were not able to predict the influence of binder content on mechanical properties. Studies show the influence of using starch and latex in the coating formulation; increases in starch content improve the

tensile strength of the coating but decrease the elongation at break [5]. In other studies, paper coating elastic moduli in tension and compression were evaluated [6]. In tensile modulus of coating formulations, there was a maximum value when hard or soft latex was used, but in compression, this trend was not observed, which can be attributed to the role of pigments in resisting load. The amount of latex still had an important role in the compression mode. Using some models to predict these mechanical properties also showed that compression properties can be predicted by a Voigt model, but the rule of mixture for tension was not applicable. In a study, Alam [7] suggests mixing rules that seem to predict these mechanical properties. The influences of binder and dispersants are summarized by other researchers [8].

The role of pigment type, amount of latex, and folding direction on cracking at the fold have been documented [9-11]. Kaolin-based pigmented coating formulations had larger cracks in comparison with ground calcium carbonate (GCC) pigments. For kaolin-based coatings, cracks propagate at an angle through the coating layer, while GCC coatings had cracks that were perpendicular to the coating layer. Paper folded in cross machine direction showed larger cracking compared with those folded in machine direction.

CRACKING

Finite element method (FEM) has been used to understand the folding event and to optimize the crack performance of multilayer coatings. In one study [12], it was found that shear, tension and compression stresses are present in the folding areas of a coated paper. Others [13,14] optimized the balance between cracking at the fold and stiffness by the use of triple coating layers: a thin stiff layer as a base coating layer, a second flexible and thick layer, and a thin top coating layer that was stiff was found to be the optimum combination. For modeling purposes, the coating layers were assumed to be perfectly elastic materials in both tension and compression modes. The results were compared with pilot trials of another study [15]. In these models, the coating layer is treated as a continuum material.

Discrete element methods (DEM) were used to predict the failure in the coating layer model, considering binder properties and pigment volume concentration [16]. The binder properties are used to predict the pigment-pigment force transmission as regions undergo tension or bending.

Cracking is predicted if the local strain exceeds the strain at failure of the binder system.

In all past work related to cracking at the fold, the mechanical properties of the coating layers were not quantitatively reported. In this study, the degree of cracking of two types of paper coated at two different binder contents with binder systems that include different ratios of latex to starch is quantified. The mechanical properties of these coating layers are known and given in a recent paper [17]. The cracking area of the folded papers was evaluated within single layer coated papers and double layer coated papers. Cracking area of different coating formulations is compared and reported for different basis weight papers. Effort has been made to explain

observations using the mechanical properties of the coating layers, as well as those of the base paper.

MATERIALS AND METHODS

Coating formulations, with different pigment and binder ratios, were prepared to apply on the base papers. These coating formulations were combinations of GCC (Covercarb HPL 70.5, Omya; Florence, VT, USA) as coating pigment and styrene-butadiene copolymer latex (Genflo 5086, Omnova; Green Bay, WI, USA) with low glass transition ($T_g = 5^\circ\text{C}$) and ethylated starch (Ethylex 2025, Tate & Lyle; Houlton, ME, USA) as binders. Papers were coated at two different PVC ratios: 78% PVC (90% pigment and 10% latex based on weight) and 62% PVC (80% pigment and 20% latex based on weight) when the binder was pure latex. Similar coatings were formulated as the same PVCs with starch replacing part of the binder system. These were 78% PVC (90% pigment, 8% latex, and 2% starch or 90% pigment, 6% latex, and 4% starch based on the weight) and 62% PVC (80% pigment, 16% latex, and 4% starch or 80% pigment, 8% latex and 12% starch based on the weight).

The starch was cooked prior to the mixing, by using a standard starch cooker Sterco#4 (Sterling, IL, USA) for 60 min at 90°C at 20% solids content. The pigment, latex, and starch were mixed for 20 min using a shear mixer at 300 rpm prior to application. The mechanical properties of free-standing coating layers produced from these formulations obtained in a three-point bending test are given in **Table I**. Details of the coating layer preparation and testing methods are presented in [17]. In these coating layers, as starch content increases, the elastic modulus, flexural modulus, and stress at failure all increase, but the strain at failure decreases.

These coatings were applied on two types of base paper,

PVC, %	Starch Content, %	Flexural Modulus, GPa	Stress at Failure, MPa	Strain at Failure, %
78	0	1.8 (0.35)	9.2 (1.1)	3.1 (0.23)
78	10	2.1 (0.75)	9.3 (0.9)	0.46 (0.06)
78	20	3.13 (0.64)	9.85 (1.26)	0.58 (0.09)
78	40	1.38 (0.43)	3.8 (0.83)	0.49 (0.06)
62	0	3.04 (1.46)	20 (8.7)	2.74 (1.3)
62	10	3.1 (0.85)	18.5 (2.1)	0.89 (0.91)
62	20	3.7 (0.03)	18.8 (3.04)	0.63 (0.1)
62	40	5.15 (0.45)	18.52 (0.74)	0.41 (0.06)

I. Flexural properties of coating formulations for 78% and 62% PVC [17]. Starch content is based on weight of the total binder weight. Numbers in brackets are the standard deviation of the measurement.

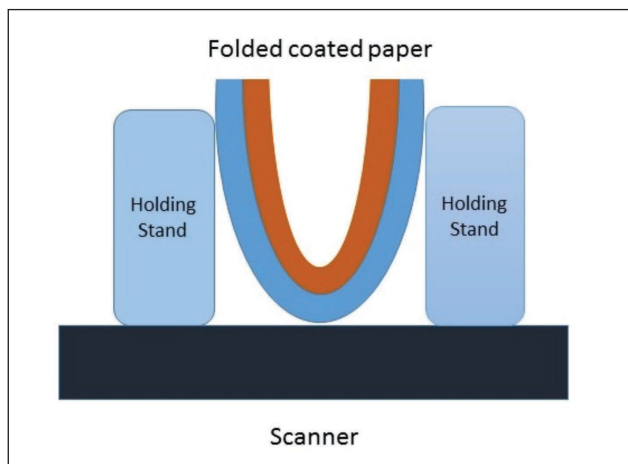
Paper	Base Weight, g/m ²	Thickness, μ m	Porosity, Gurley s	Tensile Modulus, MPa	Tensile Strength, MPa	Max Tensile Strain, %
A	200	400	143	115.5	5.5	11
B	120	240	62	167.7	6.4	7.5

II. Physical and mechanical properties of base papers.

Paper A (200 g/m² basis weight, air resistance of 143 Gurley s) and Paper B (120 g/m² basis weight, air resistance of 62 Gurley s). The mechanical properties of these two base papers are presented in **Table II**. Papers were coated with a BYK automatic film applicator (Rochester Hills, MI, USA). Each coating layer was about 25 g/m². After applying a coating layer, papers were dried in a lab oven at 105°C for 10 min. After a single layer of coating, some samples were coated with a second layer on top of the first coating layer; the second layer coat weight was also about 25 g/m². The double-coated papers were also dried in the lab oven for another 10 min. For comparison, a second layer of pure latex was applied on single layer coated papers with 78% or 62% PVC. The coat weight of a second layer of pure latex was approximately 16 g/m².

To study the effect of coating composition on cracking at the fold, for each base paper and coating formulation, four samples of 50 mm-by-80 mm were cut out in both machine direction (MD) and cross direction (CD). All coating layers, first or second layer, were applied in the machine direction of papers to minimize any effect on coating layout formation.

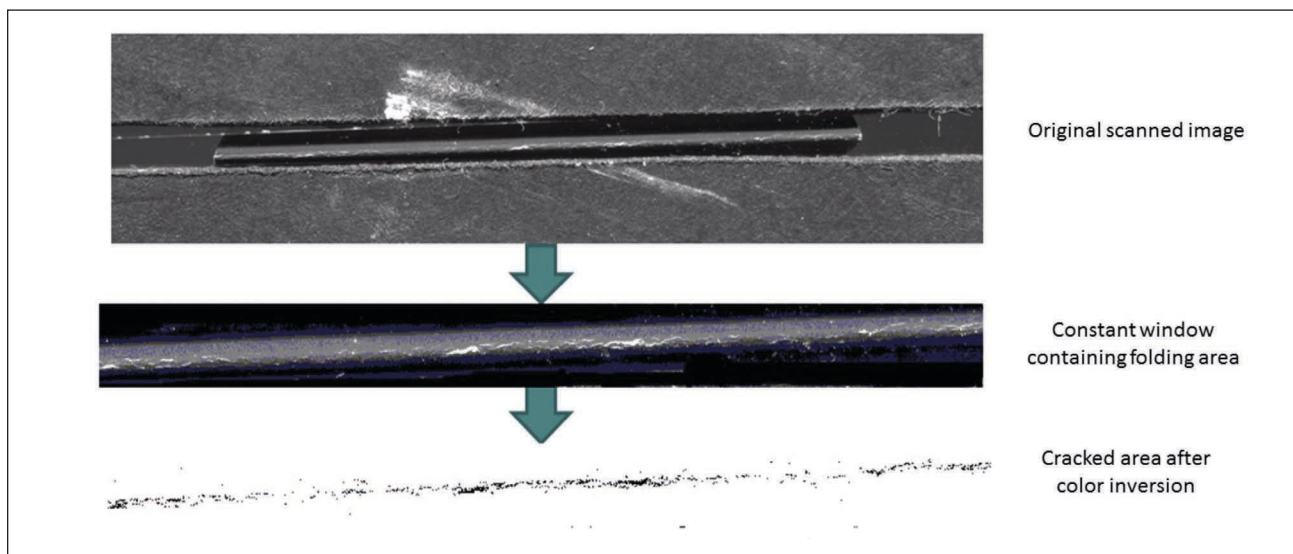
As other researchers have suggested [10-11], in order to have a better observation of cracking at the folding line, the samples were spray painted with a black color to improve the contrast of the cracks in the folding area, without affecting the properties of the coating layer. All painted samples were folded with a lab folding machine (NPA 6, Nancy Plowman Associ-



1. A schematic drawing of the stand used to keep the folded samples at the folding angle while scanning.

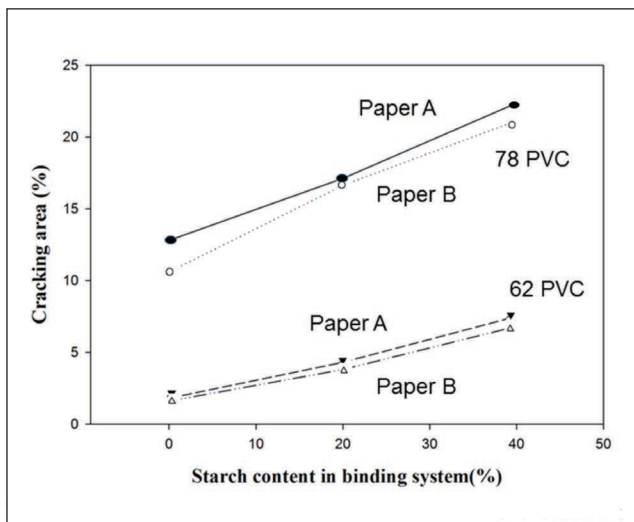
ates; Burlington, MA, USA) at 180° at room temperature.

All the folded samples, in MD and CD directions, were scanned with a high resolution EPSON V370 scanner (J232D, Suwa; Nagano, Japan). To assure the cracking area and openings were still in their place while scanning, a stand depicted in **Fig. 1** was used to hold the samples at the folding angel during scanning. Images were analyzed in a constant area with the help of Image-J software. **Figure 2** shows a typical



2. Image analysis to quantify the cracking area. The image belongs to the folded sample of 78%–62% pigment volume concentration (PVC) double layer coating on base paper B in the machine direction (MD).

CRACKING



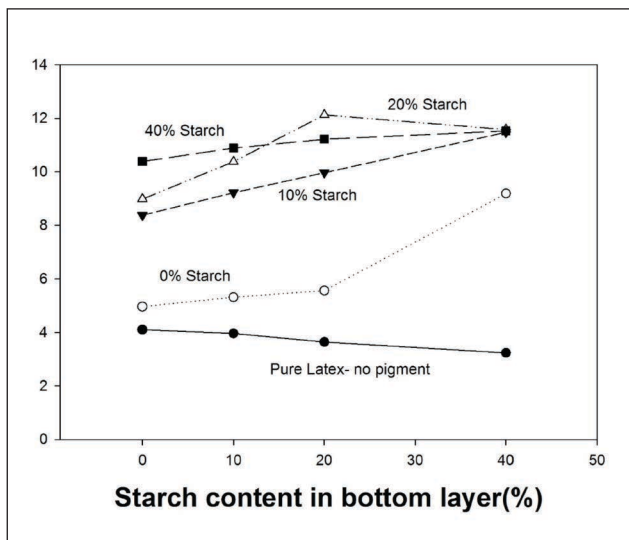
3. Effects of PVC, starch content, and paper type on cracking area in MD for single layer coated papers.

image where multiple cracks are formed in the coating layer, and the steps for crack area calculations. Images of cross sections and cracked areas in the folded samples were also taken after folding, while kept at their folding angle, using a HITACHI Tabletop TM 3000 scanning electron microscope (SEM) (Hitachi America; New York, NY, USA.)

Bending stiffness of all coated papers was also evaluated with a Taber stiffness tester V-5 (Taber Industries; North Tonawanda, NY, USA). All samples were conditioned (60% relative humidity [RH] and 25°C) for 24 h prior to the test. Four samples were cut from each coated paper formulation and tested.

RESULTS AND DISCUSSION

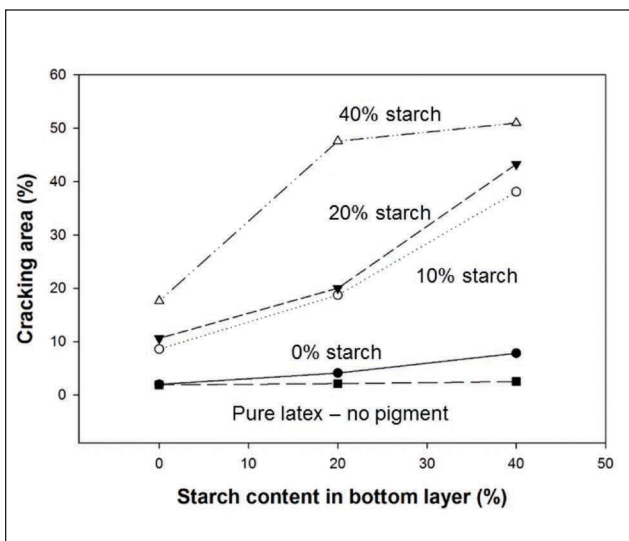
Figure 3 summarizes the effects of PVC, starch content, and paper type on cracking area of single layer coated samples. There is a considerable difference between two PVC levels for both types of coated papers, but the difference between papers A and B is small. In general, paper B showed less cracking area compared with paper A. Paper A is a thicker paper than paper B and has a higher base weight. Studies on the effect of base paper on cracking [10-11] showed that thicker papers have a larger radius at the folding curvature, resulting in a larger strain at the folding area. This increase in strain at the folding area would result in more cracking of the coating layer. With increasing the starch amount in the binding system within the same PVC, the cracking area increased significantly for both paper A and paper B and showed the same trend. However, at 78% PVC, because it is a stiffer coating film, it was expected to have more cracking [10,13]. The binding system of these formulations were combinations of different latex and starch ratios. The latex that has been used in these formulations (S/B latex) had a low T_g , and part of that was replaced with starch in some formulations. It was found that cracking area increased at the folding line with increasing the starch content of the coating formulations: this agrees well with re-



4. Comparison of results in cracking area between two types of papers, A and B, with different amounts of starch in their double layer coating formulations: 78% PVC bottom layer, and 62% PVC top layer.

cent results of others [10]. As starch is included in the binder system, the coating layers increase in elastic modulus and maximum stress in the three point bending tests, given in Table I. However, this increase in properties does not improve crack resistance. Instead, the decrease in the strain at failure with increase in starch content explains the results. Strain-based cracking can be used to explain this increase of cracking in coated papers. There are fewer cracks in the folding line but the cracks became larger, and this pattern was also observed in other studies [18].

For double-coated samples that had 78% PVC in the top and bottom layers, the cracking is serious and the crack areas are

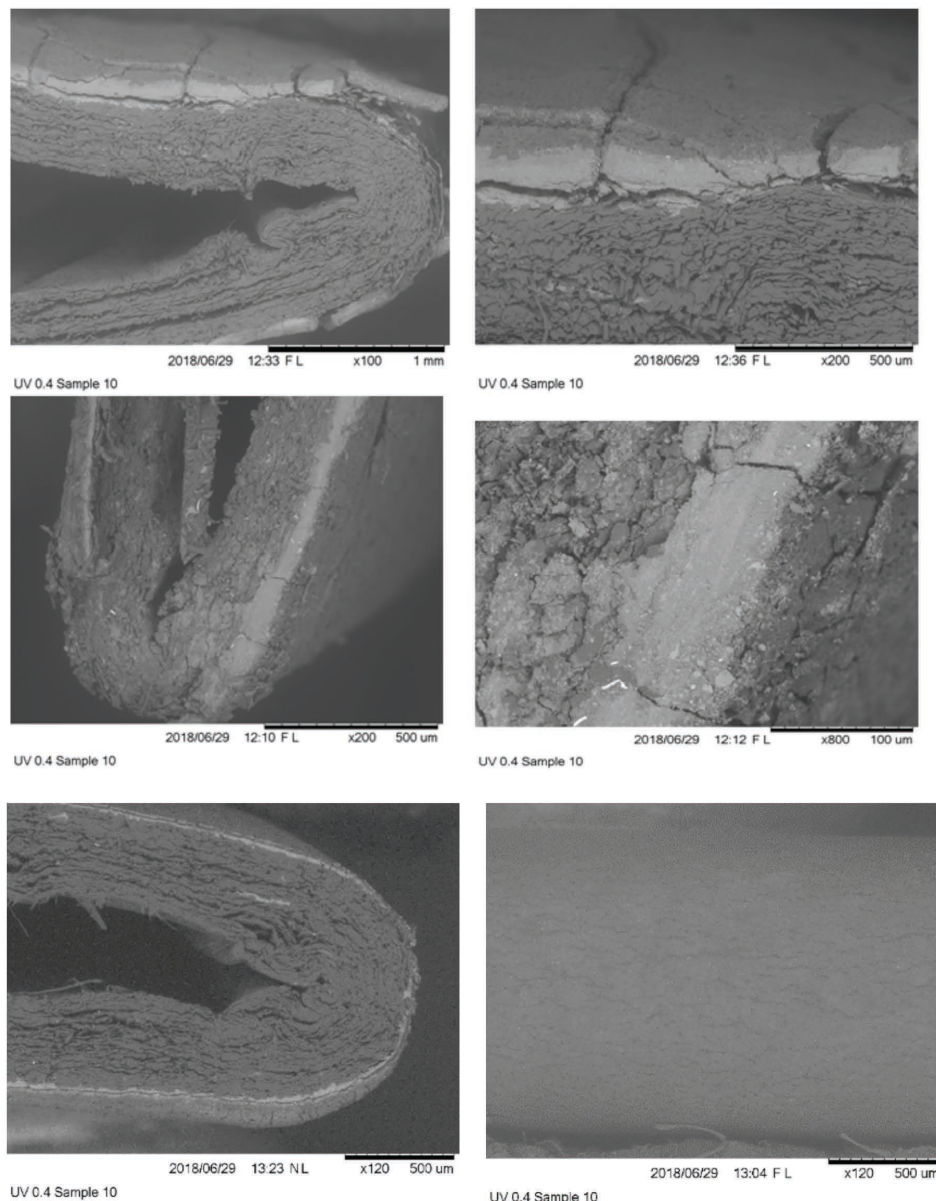


5. Comparison of results in cracking area between two types of papers, A and B, with different amounts of starch in their double layer coating formulations: 62% PVC bottom layer, and 78% PVC top layer.

large, especially when starch is present in the coatings. The crack areas were generally more than the single-coated systems. The quantitative results are not reported here. In double layer coated papers, for both paper A and paper B, when the papers were coated with a 78% PVC formulation as the first coating layer and either a pure latex or 68% PVC second layer, the crack area has a complex behavior, which is shown in **Fig. 4**. Changes in the starch content of the binding system for the first coating layer did not show a considerable change in the cracking area, but change in the starch content

of the second layer had a large effect. This result is the same as what is reported by other researchers [10]. Again, the decrease in the strain-to-failure of the top coating's starch-rich coating layer correlates with the increase in crack area.

Comparing Figs. 3 and 4, the crack area of a high starch content 78% PVC decreases a significant amount by coating a second layer that has a 62% PVC, even if it contains a significant amount of starch. This decrease is even more pronounced when the second coating layer is low in starch. For example, the crack area of 40% starch at 78% PVC is around 20%. When



6. Scanning electron microscope (SEM) images of double-coated samples. Top: 78%–62% PVC samples at 100X on the cross section of folded paper (left) and at 200X on the cracking area (right); delamination occurred in these samples. Middle: 62%–62% PVC samples at 200X on the cross section of folded paper (left) and at 800X on the cracking area (right); no delamination is observed but the coating layer has cracked after folding. Bottom: 62% PVC with a second layer of pure latex on top at 120X on the cross section of the folded sample (left) and 120X on the folded area from top that shows a scarifying layer cracking underneath (right).

CRACKING

this coating layer is covered by a 62% PVC coating that contains only latex, the crack area is reduced to 9%.

Figure 4 also demonstrates that coating with a high starch containing first layer, when coated with a pure latex layer, reduces the crack area even further. The flexural properties of the pure latex film could not be obtained in the bending tests, but in tensile, the strain-to-failure is over 300%. Therefore, this high strain at failure of the top coating layer reduces crack area to a large extent.

The results for the 62% PVC as the bottom layer, which is coated with 78% PVC as a top layer, are shown in **Fig. 5**. When the top coating layer is pure latex or a latex rich coating layer, the crack area is low. However, once starch is added in the second coating layer, the crack area increases to a great extent. For example, the crack area of the first layer increases from around 7% for a single layer at 62% PVC and 40% starch, to 40% crack area for the same coating covered with a 78% PVC coating that contains 20% starch. Again, the increase in the flexural modulus would indicate a stiffer top layer, but the decrease in the strain-to-failure of the top layer, with the starch addition, correlates well with the results.

Similar to Fig. 4, Fig. 5 also shows the decrease in crack area to almost nothing for the pure latex coating. This suggests one method to reduce cracking would be to apply extra latex to regions that are to be folded. This application potentially could be done in the converting operation. Therefore, instead of using latex everywhere in the top coating layer, it can be applied only to regions where folds are expected to occur.

In the double layer coated samples with 62% PVC as the first coating layer and 78% PVC as the second coating layer, cracking area increased as the starch-latex ratio increased in the first layer of coating, but for the reverse situation, the crack area did not depend a great deal on the starch content of the first layer. The 78% PVC coating is porous and when cracking occurs, it likely can fragment in compression. The 62% PVC coating is

near the CPVC and has minimal pores. When the 62% PVC coating is compressed and cracks, there is no region to absorb energy and hence the top layer is forced to deform and crack.

This behavior can be better explained with some SEM images taken from the samples shown in **Fig. 6**. Having 62% PVC as the first coating layer and applying a ductile layer on top of it, the image analysis did not show much cracking area in the fold line, but the SEM images show that the first layer can play the role of a scarifying layer that cracks although the second layer helps hold together the layer that has many cracks. In other words, although cracks are present in the top layer, they are not large and visible to the optical images.

CONCLUSIONS

Increasing starch content and PVC leads to a decrease in the strain at failure of coating layers and an increase in cracking area for both types of papers for single layer systems: crack area correlates with strain at failure more than elastic modulus or stress at failure. In double layer coatings with 78% PVC in both layers, the cracking was severe. However, when a latex-rich top coating layer is applied on top of a high starch content lower layer, the cracking is reduced to a large extent: the high strain to failure of the top layer helps keep cracking contained in the first layer and holds the system together. When the second layer coating is pure latex, cracking decreases to a low level, even with a high starch content first layer. Coatings with 78% PVC as the first layer showed delamination in the samples, but for 62% PVC samples, delamination did not happen, although the coating layer cracked in the folded area. **TJ**

ACKNOWLEDGEMENTS

The authors wish to thank the Paper Surface Science Program of the University of Maine for discussion and support of this project. We especially thank Gary Fugitt, WestRock, for his input and advice on this work.

ABOUT THE AUTHORS

We chose this topic for study because it remains an important issue for the sponsors of our program. It advances the work of Oh et al. [9,10] and provides critical data for the work of researchers D. Varney and D.W. Bousfield.

The most challenging aspect of this study was measuring the crack area. To address this, we built a device to hold paper in a folded position.

We discovered that a latex-rich top coat can solve much of the cracking issue. It was interesting to find that a top layer latex concentration could cover high starch.

This study shows that mills that double coat have a significant potential to use more starch.

Hashemi Najafi is a graduate student in the School of Forest Resources; Bousfield is professor, Paper Surface



Hashemi Najafi



Bousfield



Tajvidi

Science Program, in the Department of Chemical and Biomedical Engineering; and Tajvidi is professor in the School of Forest Resources at the University of Maine, Orono, ME, USA. Email Bousfield at bousfld@maine.edu.

LITERATURE CITED

1. Husband, J., Preston, J., Gate, L., et al., *TAPPI J.* 5(12): 3(2006).
2. Husband, J., Preston, J., Gate, L., et al., *TAPPI J.* 6(12): 10(2007).
3. Prall, K., Shaler, S., and LePoutre, P., *Nord. Pulp Pap. Res. J.* 15(5): 564(2000). <https://doi.org/10.3183/NPPRJ-2000-15-05-p564-571>.
4. Raman, K., Bousfield, D.W., and Shaler, S., "Analyzing the effect of binder content on the mechanical properties of paper coating," *TAPPI Adv. Coat. Fundam. Symp.*, TAPPI Press, Atlanta, GA, USA, 1998.
5. Chen, G., Zhu, Z., Salminen, P., et al., *Adv. Mater. Res.* 936: 74(2014).
6. Alam, P., Qian, X., Toivakka, M., et al., "The elastic modulus of paper coating in tension and compression," *TAPPI Adv. Coat. Fundam. Symp.*, TAPPI Press, 2007.
7. Alam, P., *Mech. Res. Commun.* 37(4): 389(2010). <https://doi.org/10.1016/j.mechrescom.2010.04.002>.
8. Toivakka, M., Alam, P., Touaiti, F., et al., *TAPPI J.* 14(6): 373(2015).
9. Oh, K., Sim, K., Bin, Y., et al., *Nord. Pulp Pap. Res. J.* 30(2): 360(2015). <https://doi.org/10.3183/NPPRJ-2015-30-02-p360-367>.
10. Oh, K., Seo, D., Jung, Y.H., et al., *Nord. Pulp Pap. Res. J.* 31(2): 347(2016). <https://doi.org/10.3183/NPPRJ-2016-31-02-p347-353>.
11. Rättö, P., Hornatowska, J., Changhong, X., et al., *Nord. Pulp Pap. Res. J.* 26(4): 485(2011). <https://doi.org/10.3183/NPPRJ-2011-26-04-p485-492>.
12. Barbier, C., Larsson, P., and Ostlund, S., *Compos. Struct.* 67(4): 383(2005). <https://doi.org/10.1016/j.compstruct.2004.01.024>.
13. Alam, P., Toivakka, M., Carlsson, R., et al., *J. Compos. Mater.* 43(11): 1265(2009). <https://doi.org/10.1177/0021998308104227>.
14. Saliman, P., Carlsson, R., Sandas, S., et al., "Modeling and experimental studies to optimize the balance between fold crack resistance and stiffness for multilayered paper coatings—part 2: Pilot coater experimental studies," *PaperCon*, TAPPI Press, Atlanta, 2008.
15. Saliman, P., Carlsson, R., Sandas, S., et al., "Modeling and experimental studies to optimize the balance between fold crack resistance and stiffness for multilayered paper coatings—part 1: introduction and modeling studies," *PaperCon*, TAPPI Press, 2008.
16. Varney, D. and Bousfield, D.W., *TAPPI J.* 17(1): 21(2018). <https://doi.org/10.32964/TJ17.01.21>.
17. Hashemi Najafi, S.M., Tajvidi, M., and Bousfield, D.W., *Prog. Org. Coat.* 123: 138(2018). <https://doi.org/10.1016/j.porgcoat.2018.07.009>.
18. Rättö, P. and Hornatowska, J., "Dynamic aspects of crack development in coating layers," *TAPPI Adv. Coat. Fundam. Symp.*, TAPPI Press, Atlanta, 2010.

ABOUT THIS PAPER

Cite this article as:

Najafi, S., Bousfield, D., and Tajvidi, M. *TAPPI J.* 18(2): 93(2019). <https://doi.org/10.32964/TJ18.2.93>

DOI: <https://doi.org/10.32964/TJ18.2.93>

ISSN: 0734-1415

Publisher: TAPPI Press

Copyright: ©TAPPI Press 2019

[About this journal](#)