

Discrete element method to predict coating failure mechanisms

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ABSTRACT: The mechanical properties of coating layers are critical for post-application processes such as calendering, printing, and folding. Discrete element methods (DEM) have been used to simulate basic deformations such as tensile and compression, but have not been used as a tool to predict cracking-at-the-fold (CAF) or picking. DEM has the potential to increase our understanding of these failure mechanisms at the particle level. We propose a method to model the three-point bending of a coating layer and also the out-of-plane picking event during printing (using a z-direction scenario and an approach involving a moving force/velocity). Properties of the binder and the binder concentration are input parameters for the simulation. The model predicts the crack formation of the layer, the flexural modulus, and the maximum flexural strain during bending. The model also predicts the forces required for picking to occur. Results are compared with those of complimentary studies.

Application: Mills can use the findings of this study to better understand the mechanical properties of these coatings and to adjust coating formulations to better meet product needs.

Coated paper and board undergo various process steps after coating application which, depending on the coating mechanical strength, can negatively affect final quality. Such post-application steps include calendering, printing, and folding. The potential quality problems that can result from these processes are cracking, picking, and cracking-at-the-fold (CAF), respectively. These issues result from compression (calendering), z-direction tension (printing), and bending (folding). The proper balance of the mechanical properties of the coated substrate must be achieved to avoid such problems.

The discrete element method (DEM) and the finite element method (FEM) have been used extensively for modeling free-standing coating layers. DEM has been used for modeling coating layers under tension [1-4], under compression [5-7], and, just recently, during bending [8]. FEM has been used to model all three types of events [9-11]. Lyons and Peshave [12] used a unique approach to model the bending of coated paper. They proposed calculating coating stiffness by using a three-point bending technique in conjunction with modeling the coated paper as a multilayer construct as opposed to an I-beam. Although this work has great potential to help us understand the macroscopic behavior of these systems, that does not extend to an understanding of behavior at a pigment level.

Because the finite element method normally treats the coated paper as a composite material, it does not make the “connection” between events on the micro-scale and the responses on a macro-scale (as can the DEM). FEM can be used to develop particle-level models [13], but the computational complexity and the computing time increase significantly as the number of particles goes up.

This current paper will build on the recent work of Varney and Bousfield [4,8]. The nonlinear tension model will be applied to out-of-plane (z-direction) tension, which is a modification to the in-plane tensile modeling of the first paper referenced. This effort will simulate picking. Two picking simulations will be studied: the traditional out-of-plane z-direction approach and a novel moving force/velocity situation. In addition, the same tension and compression equations outlined in the second paper will be applied to a three point bending simulation for latex and latex/starch binder systems. For both the picking and bending scenarios, the model will be compared to the experimental data of Hashemi-Najafi et al. [14].

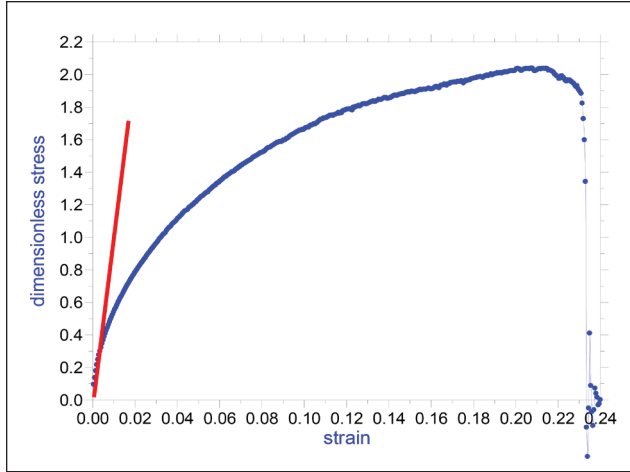
MODEL DEVELOPMENT

As outlined in two earlier papers [4,8], a set of equations were developed to provide a better fit with the nonlinear stress-strain results seen during tension experiments [15-17]. The force equation evolved into the following:

$$F = A(1 - e^{-B\varepsilon})\pi R_b^2 \quad (1)$$

where F is the tensile force between particles, A and B are parameters that depend on the pure binder, ε is the local strain between particles, and R_b is the radius of the binder bridge between particles. The bridge radius and the spacing of the particles depend on the pigment volume concentration (PVC), which is defined as the ratio of total volume of pigments to the volume of pigments plus the volume of binder. When the local strain between particles is larger than the strain-to-failure of the pure binder, the binder is assumed to fail cohesively and the force is set to zero. As confirmed with

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1. Typical nonlinear response from tension simulation [4].

the data of Zhu et al. [17], the model provides a nonlinear response, as depicted in **Fig. 1**.

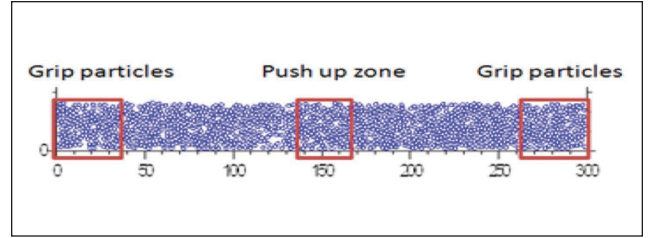
For compression, a repulsive force is calculated as:

$$F = \frac{Eh_i}{h} \varepsilon \pi R_b^2 \quad (2)$$

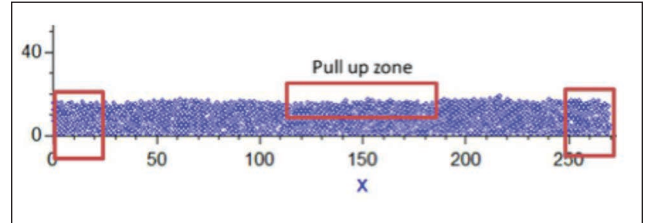
where b and b_i are the current and initial gaps between particles, respectively, and E is the elastic modulus of the binder. The ratio of initial to current gaps is needed in this expression to give a large force as the current gap goes toward zero. This large force would come from the compression of high points on the particles and is important to keep particles from overlapping. The difference between tension and compression is that, in compression, the force has to go to a large value as the gap is small to keep particles from overlapping each other while, in tension, the force follows the behavior seen in tensile tests and goes to zero when the critical strain is reached.

The bending simulation is done by holding two groups of particles in place, not allowing motion, while another group of particles is set to move upward (**Fig. 2**). This arrangement simulates a three-point bending test. Other types of deformation are also possible with the model, such as moving a group of particles at one end upward and not allowing some particles on the other end to move, as in a cantilever. Particles in the “push-up” zone are moved upward with a dimensionless velocity of 1. The grip particles can be either not allowed to move at all or not allowed to move in the vertical direction. This latter condition lets the particles in the grip zones slide and simulates a three-point bending test where the sample is supported loosely with a support structure.

The flexural force on each particle is calculated as it moves relative to its neighboring particles. If particles move away from each other, Eq. 1 is used to calculate the tension force between these particles. If particles are pushed together, then a repulsive force is applied to keep the particles from overlapping, as in Eq. 2. The net vertical force on the left and right



2. Boundary conditions for a bending simulation. Particles in push-up zone are set to move upward.



3. Picking simulation by applying an out-of-plane velocity in the pull-up zone. Either bottom layer of particles is held stationary or the side particles are not allowed to move.

grip particles are summed. This upward force should equal the net downward force on the particles that are moving upward to balance the forces. The flexural stress and strain are defined respectively as:

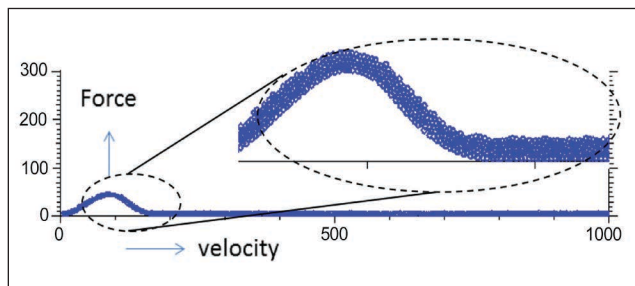
$$\sigma_f = \frac{3PL}{2bd^2} \quad (3)$$

and

$$\varepsilon_f = \frac{6Dd}{L^2} \quad (4)$$

where P is the sum of the forces on the grip particles (or the load force), L is the distance between grips, D is the displacement of the upward moving particles at the center of the sample, b is the width of the sample, and d is the thickness of the sample. The stress, σ_f , reported here is made dimensionless with the elastic modulus of the binder. The goal is to predict the bending behavior of these systems and to predict the crack propagation.

Similarly to the bending simulation, the picking model sets the velocity of the top couple of layers of particles to an upward direction in the “pull-up” zone (**Fig. 3**). Two conditions are compared for other particles: 1) the particles along the bottom of the region are assigned zero velocity or 2) the particles on the sides are not allowed to move. The first condition does not let the paper flex or bend during printing, as when printing on both sides of the sample. The second condition would represent single-sided printing, where the paper might bend slightly at the nip exit as ink tack forces pull on the paper. This condition might be similar to standard tests, such as the IGT pick test that prints an ink layer on one side



4. Conditions to simulate the exit of a printing nip. A force is applied to a region of particles and moves along the web. Upper right part of the figure is an enlargement of the region that is experiencing an upward force or velocity.

of the paper. The net force on the bottom particles predicts the force event that the coating will see as it fails.

The length scales for the simulations for bending and for picking are scaled with pigment radius. If the particles have a radius of 1 μm , then the length in Fig. 3 would represent a $270\ \mu\text{m} \times 20\ \mu\text{m}$ region; the height of this region would be typical of a paper coating layer thickness. The length scale is much smaller than typical bending tests or production scales, but representative paper and board deformations of these sorts can be modeled by increasing the number of particles in the matrix. Regardless of the length scale, the results should be similar because the parameters input into the model would be the same.

Another way to simulate picking takes a long layer of particles with a force or velocity dynamically applied to various regions of this sample. **Figure 4** illustrates this condition. The force is applied to a region of the particles. The position of this applied force or velocity then moves from left to right at some known velocity. This would represent the paper moving from right to left. This scenario should be close to the condition where the tack force is applied to a small region of the paper for a short amount of time as the web exits the printing nip.

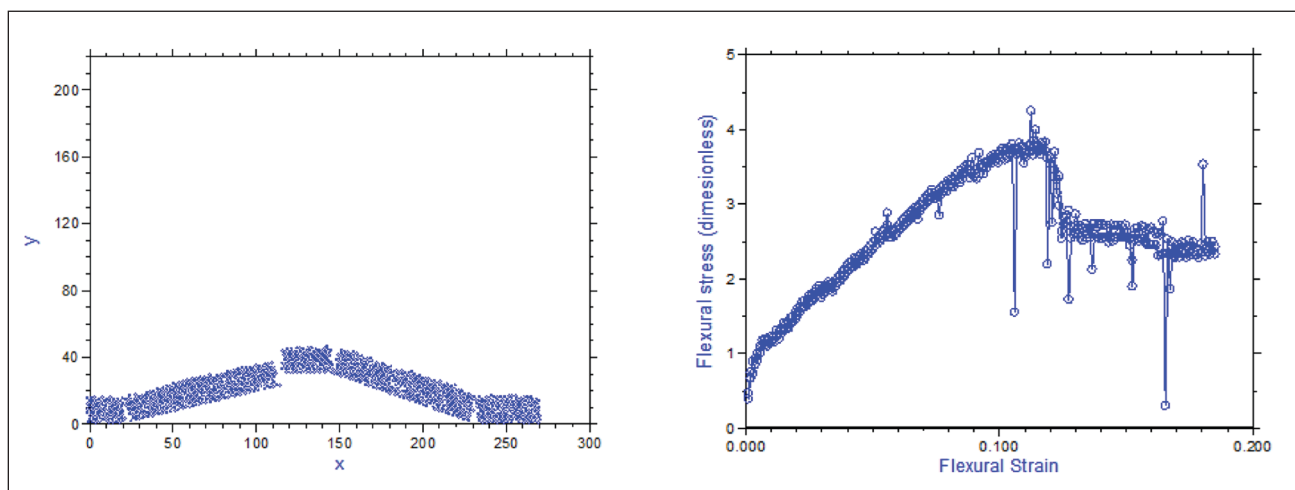
A model of this nature involves a number of assumptions. The results presented here are limited to equal-sized spheres that are confined in a mono-layer. The spheres are assumed to be rigid particles and all of the deformation is either compression or tension of the latex bridges between particles. The model in this current form neglects the rotation and shear between particles; they can be included if these types of deformation are found to be important.

Figure 5 shows the results of a bending simulation for a system near the critical pigment volume concentration for a binder package of 40% latex and 60% starch. As expected, the coating layer fractures earlier than the latex-only binder properties at around 10% strain. The flexural stress increases to a maximum and drops sharply when a crack develops. The shape of these predictions is similar to the experimental results.

Figure 6 shows typical results for the picking simulations, where the bottom particles are held stationary. A group of particles are forced to “pick” from the coating layer. In the real situation, the top surface of a coating layer would be subject to a normal-acting tack force instead of a velocity. If the force is less than the maximum shown in Fig. 6, the particles will deform some amount but then remain as part of the coating layer.

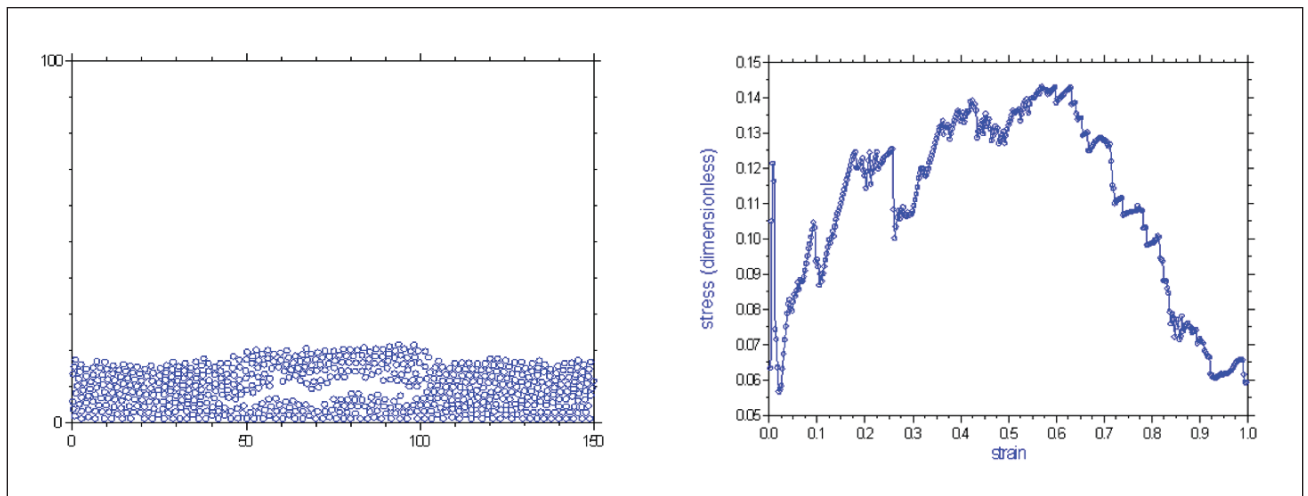
Figure 7 shows results obtained when the picking simulation is done by allowing the bottom particles to move but by holding the ends stationary. Again, this situation should represent the case where the web is allowed to deflect a small amount during printing, as would happen with one-sided printing. Note that quite a different behavior is obtained in that the coating layer cracks through the entire depth instead of a small region being picked from the coating layer. In addition, the strain-to-failure is larger in this case because the bottom layers of the particles are not being held stationary, which allows some level of movement.

Figure 4 shows typical results of the moving force boundary conditions. For the conditions used, the coating layer de-

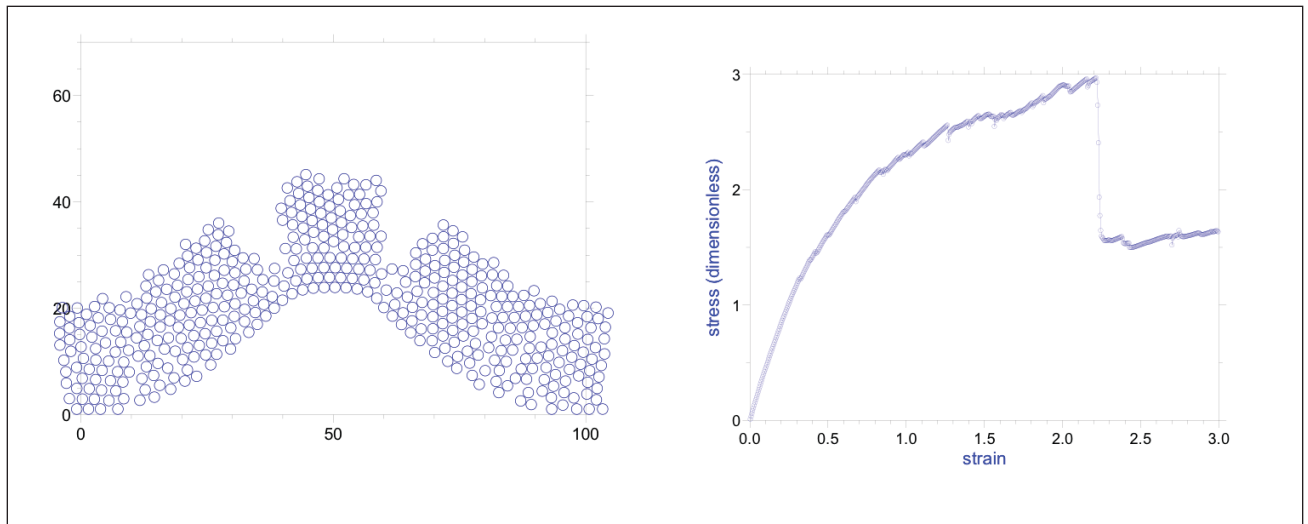


5. Typical results for 62 pigment volume concentration (PVC; 20 pph binder) for binder properties that resemble 40% latex and 60% starch. Left are the particle positions near the end of the simulation and right is the stress-strain prediction.

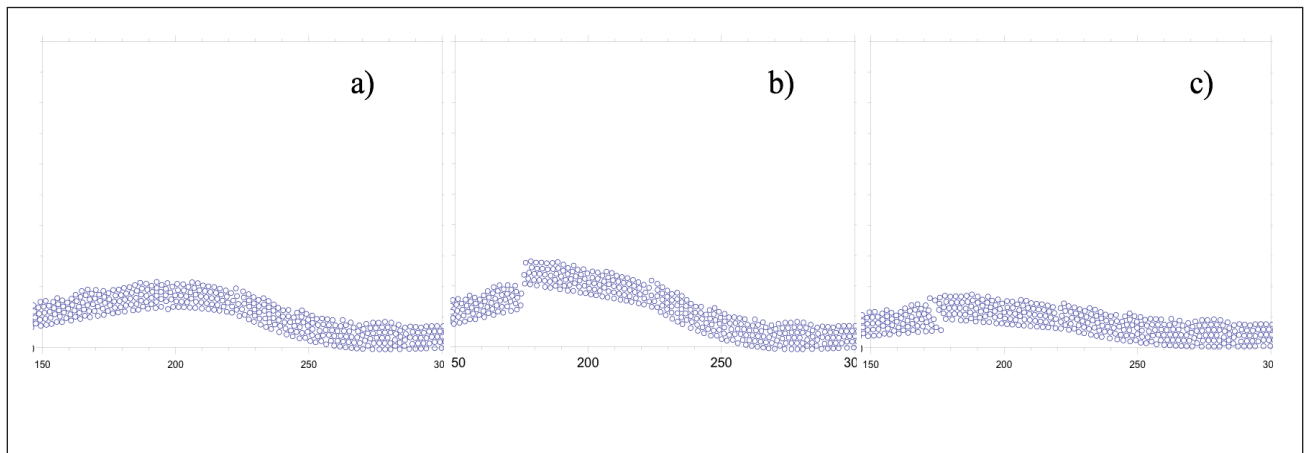
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6. Typical results of the picking simulation for pure latex binder properties and PVC of 62 (20 pph binder). Left are the particle positions near the end of the calculation and right is the stress-strain prediction.



7. Typical picking results when the ends of the coating layer are fixed for similar binder properties as in Fig. 6. Left are the particle positions near the end of the calculation and right is the stress-strain prediction.



8. Results for binder properties that contain 80% latex and 20% starch at 20 pph binder content with a) net displacement of 10 units, b) net displacement of 12.5 units, and c) binder composition of 60% latex and 40% starch and a net displacement of 10 units.

forms and does not crack. In general, the model predicts that as the elastic modulus of the binder becomes large, the force needed to crack the layer increases. This behavior does not agree with the common experience and the companion study [14] in that as starch is added, the coating layer elastic modulus increases, but the picking velocity decreases.

Figure 8 shows another key result for the moving velocity condition. In this case, a region of the coating layer is forced to deflect a certain distance because an upward velocity boundary condition is set on a region of the coating layer. The location of this condition moves from left to right (Fig. 8). Regardless of the elastic modulus of the coating, the layer must deflect. Figure 8 shows that coating layers that contain starch are more prone to crack. Therefore, even though coating layers become stronger with the addition of starch, they also become more brittle. The cracking shown in Figs. 8b and 8c illustrate the crack formation that occurs.

COMPARISON WITH EXPERIMENTAL DATA

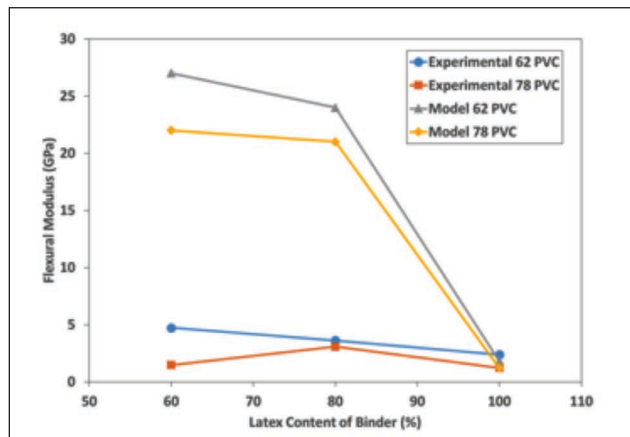
We compared the results of the model simulations with the experimental work of Hashemi-Najafi et al. [14,18]. Their efforts involved three-point bending and picking experiments. The pigment used in their coatings was a ground calcium carbonate (GCC) (60 w/w%, <2 μm) and various ratios of a latex/starch binder system (latex/starch ratios were 100/0, 80/20, and 60/40). The GCC used in this study would approximate the aspect ratio of the spheres used in the model but would not have the same particle size distribution. The properties of binder-only films were characterized by tensile tests. **Table I** lists the parameters that fit the nonlinear binder deformation, where A and B are the parameters in Eq. 1, E is the elastic modulus of the binder, and $A^*=A/E$. As expected, as the starch level increases, the elastic modulus increases, but the strain to failure decreases. Hashemi-Najafi et al. [18] report some tensile properties of the latex-only and coating layers with latex as the binder.

Bending simulations

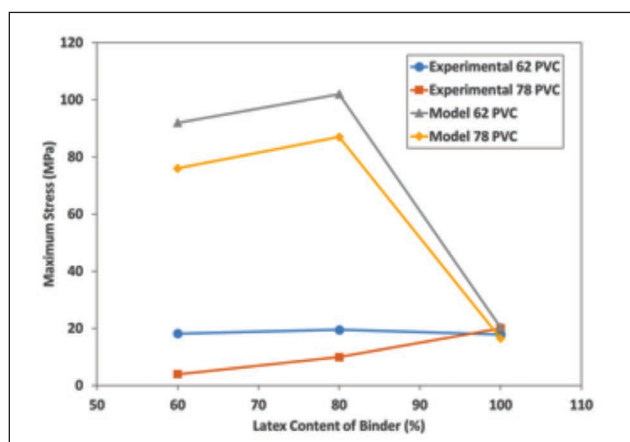
Figures 9-11 compare the model predictions with the bending experiments. For the 100% latex cases, the model predicts

Parameters	Pure Binder			
	Pure Latex	80L-20S	60L-40S	40L-60S
A	1.5	4.9	4.8	11.0
B	2	15	35	60
E, Mpa	3	73.5	168	660
STF, %	200	80	22	5
A^*	0.5	0.067	0.029	0.017

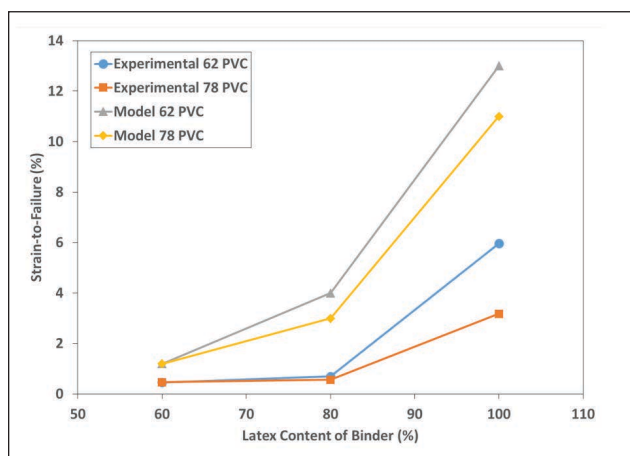
I. Properties of the pure binder films from tensile tests. Strain-to-failure (STF) is given as a percent. 80L-20S is 80% latex and 20% starch on a weight basis from companion paper.



9. Flexural modulus as a function of the latex content of the binder system for two pigment volume concentrations (62 PVC corresponds to 20 parts of binder and 78 PVC corresponds to 10 parts of binder).

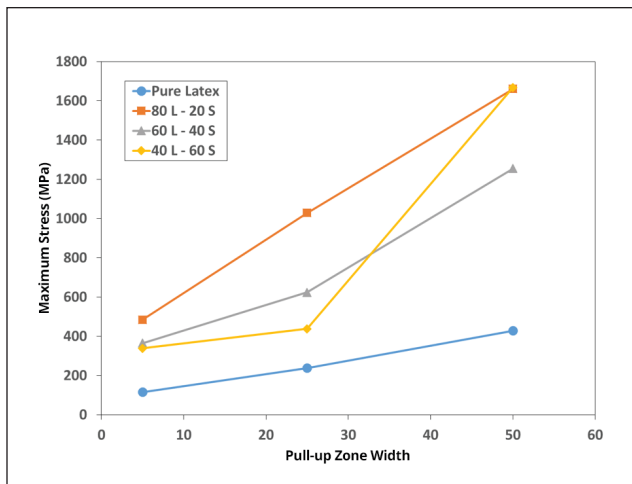


10. Maximum stress as a function of the latex content of the binder system for two pigment volume concentrations (62 PVC corresponds to 20 parts of binder and 78 PVC corresponds to 10 parts of binder).



11. Strain-at-failure as a function of the latex content of the binder for two pigment volume concentrations (62 PVC corresponds to 20 parts of binder and 78 PVC corresponds to 10 parts of binder).

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12. Maximum dimensional picking stress against pull-up zone width for four binder systems. The 80L-20S is 80% latex and 20% starch on a weight basis.

the flexural elastic modulus within 20% and maximum stress within 25%. The strain at failure is overpredicted by the model by up to 100%. This overprediction of the model can easily come from its failure to predict minor defects in the experimental samples that lead to early cracking. The model overpredicts the elastic modulus, the maximum stress, and the strain at failure for all of the starch-containing binder systems by a large amount. This result is similar to the predictions for tensile properties given by Varney and Bousfield [4,8]. The starch is a likely cause of this overprediction by:

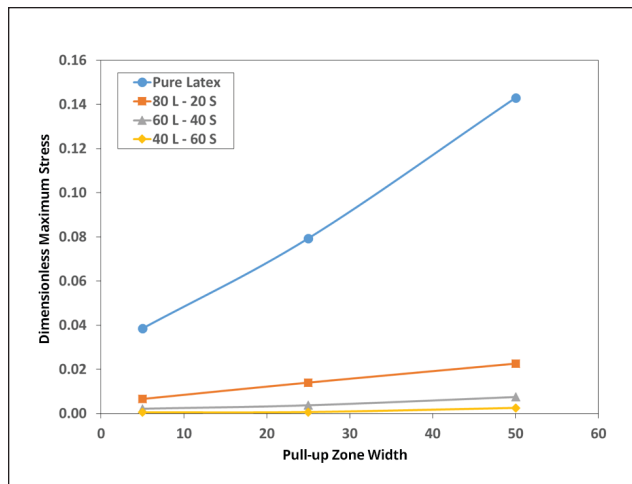
- acting to reduce the adhesion of the binder to the pigments,
- altering the packing properties or the initial separation of the particles (induced flocculation), or
- causing a number of fine scale defects (shrinkage during drying).

The maximum stress predictions are similar to the elastic modulus.

The overprediction of the elastic modulus and the stress at failure is at first surprising, but can be expected from looking just at the experimental results. As the pure binder films are changed from all latex to 60% starch, the elastic modulus of the binder increases by a factor of 50, but the elastic modulus of the coating only increases by a factor of 1.5. Similar results are found in the data of Zhu et al. [17] where the elastic moduli, in tension, of the pure binder films increase by a factor of 100, yet the moduli of the coating layers, even below the critical pigment volume concentration (CPVC), increase by a modest amount.

As discussed by Zhu et al. [17], starch seems to be acting more like a pigment than a binder. Adjusting the PVC calculation to take this idea into account results in improved predictions, but a good match with the experiments is lacking. The strain-to-failure still overpredicts the data.

Another issue that caused the overpredictions at high



13. Maximum dimensionless picking stress against pull-up zone width for four binder systems. The 80L-20S is 80% latex and 20% starch on a weight basis.

starch levels might be the adhesive strength of the binder to the pigments. Starch might reduce this adhesive property. Therefore, as the binder elastic modulus increases with starch addition, the adhesive properties of the binder to the pigment might decrease. This adhesive behavior could be studied by measuring the adhesion of binder to calcium carbonate crystals. The adhesive parameter has been incorporated into the current model, but the correct value of this parameter needs to be determined from adhesive type tests.

The best predictions are obtained by assuming that the starch acts as a pigment, by using the binder properties of pure latex, and by using the strain-to-failure properties of the starch-latex binder film. This method of using the pure binder properties underpredicts the elastic modulus of the starch-containing coating by around 20%, underpredicts the maximum stress by 15%, and overpredicts the strain at failure by 80%.

Picking simulations

Figures 12 and 13 show results of the picking simulations when the bottom layer of pigments are held stationary. These simulations were done using the four binder packages from the experimental work of Hashemi-Najafi et al. [1] and three “pull-up” zone widths in the model. Changing the widths would represent different ink halftone dot sizes in an offset press, for example. The model particle size is such that the range of up-zone widths chosen in this simulation would represent a typical halftone dot size of about 20–50 μm . This set of data is for a pigment volume concentration at the critical value (CPVC).

The maximum stresses plotted in Fig. 12 were taken from the peak on the stress/strain curves, such as those in Figs. 6 and 7 (right side), and were converted from dimensionless stress to dimensional stress. Figure 12 shows how the maximum stress at failure would increase directly with the width of the pull-up zone. This plot also shows that the latex-only

binder system had the lowest stress values while the trends for the starch-containing systems generally increased in stress indirectly with starch levels. Figure 13 is similar to Fig. 12, except that the dimensionless stress is plotted instead. In this case, the latex-only binder system had the highest values while the addition of starch caused the maximum dimensionless stress to decrease. The predicted stress before the coating cracks is well above the ink tack stress values measured by Harrison and Bousfield [19]. This result indicates that these coating layers would be strong enough to withstand normal offset printing forces.

The picking results of Hashemi Najafi et al. [14] show that the IGT velocity at picking increases with the binder level and decreases with the addition of starch. The model also predicts that increasing binder level increases the force required to cause picking. However, the increase in starch content, corresponding to the amount used in the experiments, should also increase the force required to generate picking. This result is opposite to what was found in the experiments, where increasing starch decreases the pick velocity.

The ability of the coating layer to deform, even a small amount, to a bending motion explains this outcome. As seen with the moving force condition and the picking simulation that allows bending of the coating layer, the addition of starch in the binder system decreases the strain at failure even if the coating layer is stronger.

CONCLUSIONS

A discrete element model is proposed to describe the bending and picking events for a coating layer. This model has the potential to predict cracking events when the sample is bent or picking phenomena during printing. The model incorporates the mechanical properties obtained from binder-only films.

For the bending results, the model well predicts the behavior of coatings that contain only latex as the binder in terms

of the elastic modulus, of the maximum stress, and of the strain at failure. For coatings that contain starch, the model overpredicts the elastic modulus and maximum stress. If starch is treated as a pigment, and the properties of latex are used for the binder, the predictions are improved.

In the picking simulations, the model predicts the correct trend in terms of binder level. The model also predicts that starch-containing coatings should be stronger than coatings that contain only latex, but the experiments show that the latex-only coatings are more resistant to picking. If a deflection or bending of the coating layer is imposed in the model, the cracking of the coating layer is found to increase with starch content. This result agrees with the experiments. **TJ**

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LITERATURE CITED

1. Alam, P., Toivakka, M., Lazarus, E., et al., "On the mechanisms of wet and dry strength of paper coatings: Part II – modelling and simulation," *PaperCon*, TAPPI Press, Peachtree Corners, GA, USA, 2012.
2. Toivakka, M. and Bousfield, D., *Adv. Coat. Fundam. Symp.*, TAPPI Press, Peachtree Corners, 2001, pp. 197-211.
3. Toivakka, M., Alam, P., Touatti, F., et al., "Molecular and microscopic level understanding of coating strength," *PaperCon*, TAPPI Press, Peachtree Corners, 2014.
4. Varney, D. and Bousfield, D., "Improved discrete element method model to predict coating layer mechanical properties," *PaperCon*, TAPPI Press, Peachtree Corners, 2016.
5. Azadi, P., Farnood, R., and Yan, N., *Comput. Mater. Sci.* 42(1): 50(2008).
6. Azadi, P., Farnood, R., and Yan, N., *Comput. Chem. Eng.* 32(12): 3084(2008).

ABOUT THE AUTHORS

We chose this topic to study because cracking at the fold and cracking of barrier layers are important topics for the industry. This study builds on our past modeling work. It connects with work by others who have studied the mechanical properties of coating layers.

The most challenging part of the study was trying to make accurate predictions for starch-containing coatings. Those predictions were not so good, and the reasons were hard to explain. This was an interesting finding, in that starch-containing coatings do not act as one would expect. However, we did discover a pattern to coating flexing properties and tension.

Mills can use the findings of this study to better understand the mechanical properties of these coatings and to adjust coating formulations to better meet product needs. Future work will focus on two-layer and three-dimensional modeling.



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7. Ma, D., Carter, D., and Bousfield, D., "Prediction of coating structure change during calendaring," *TAPPI Adv. Coat. Fundam. Symp.*, TAPPI Press, Peachtree Corners, 2008.
8. Varney, D. and Bousfield, D., "Discrete element method to predict bending and cracking," *Adv. Coat. Fundam. Symp.*, TAPPI Press, Peachtree Corners, 2016.
9. Alam, P., Toivakka, M., Carlsson, R., et al., *J. Compos. Mater.* 43(11): 1265(2009).
10. Barbier, C., Larsson, P-L, Ostlund, S., *Compos. Struct.* 67(4): 383(2005).
11. Salminen, P., Carlsson, R., Sandas, S., et al., "Combined 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, Peachtree Corners, 2008.
12. Lyons, A. and Peshave, C., *TAPPI Adv. Coat. Fundam. Symp.*, TAPPI Press, Peachtree Corners, 2014, pp. 11-17.
13. Barbier, C., Rättö, P., and Hornatowska, J., "Coating models for an analysis of cracking behavior between folded paper and creased board," *TAPPI Adv. Coat. Fundam. Symp.*, TAPPI Press, Peachtree Corners, 2012.
14. Hashemi-Najafi, S.M., Tajvidi, M., and Bousfield, D.W., "Measurement of tensile and bending properties of free standing coating layers," *PaperCon*, TAPPI Press, Peachtree Corners, 2017.
15. Prall, K., Shaler, S., and LePoutre, P., *Nord. Pulp Pap. Res. J.* 15(5): 564(2000).
16. Raman, K., Bousfield, D.W., and Shaler, S.M., "Analyzing the effects of binder content on the mechanical properties of paper coatings," *TAPPI Coat. Conf.*, TAPPI Press, Peachtree Corners, 1998.
17. Zhu, Z., Salminen, P., Chen, G., et al., "Mechanical properties of pigment coating composites containing starch," *TAPPI Adv. Coat. Fundam. Symp.*, TAPPI Press, Peachtree Corners, 2014.
18. Hashemi-Najafi, S.M., Tajvidi, M., and Bousfield, D.W., "Mechanical properties of free standing coating layers: bending and tensile properties," *TAPPI Adv. Coat. Fundam. Symp.*, TAPPI Press, Peachtree Corners, 2016.
19. Gates, H. and Bousfield, D., "Pressure pulse measurements for coating and printing," *PaperCon*, TAPPI Press, Peachtree Corners, 2015.

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