Discrete Element Method to Predict the Mechanical Properties of Coatings

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Extended Abstract:

The mechanical properties of many coatings and paints are important in a number of applications. In many paint applications, scuff or rub resistance is important. For coated papers, the resistance to "picking" during the printing operation is critical as well as the ability for the sample to be converted or folded without cracking of the coating layer. If the coating layer is a homogenous material, such as a specific polymer, the mechanical properties of that layer can be estimated from the bulk properties of that material. However, when the coating layer is a composite of pigments and binder, the mechanical properties are difficult to predict.

Finite element methods can be used to predict the mechanical properties of a group of particles connected by polymeric bridges, but these are limited by the need to mesh fine gaps between a large number of particles (Barbier *et al.* 2005, Alam *et al.*, 2009, Rätto, 2004). Key insights into the mechanical properties of paper coatings have been obtained with this approach (Toivakka *et al.* 2015). Discrete element methods (DEM) can be based on the pigment length scale and have potential to reveal particle level mechanisms in the study of these systems. DEM has been used to study the compression of paper coatings during the calendaring event (Azadi *et al.* 2008). Here, we propose to use DEM to understand the tensile and bending behavior of coating layers.

The experimental methods and data are reported in Najafi *et al.* (2018). Latex and starch mixtures were used as a binder between ground calcium carbonate pigments. The mechanical properties of the binder layer were characterized by casting pure binder films followed by tensile tests. The combination of latex and starch in relationship to the pigment is the Pigment Volume Concentration (PVC). Films of the composite materials were cast. These films where subject to tensile and three-point bending tests. The bending results are of interest here. Tensile results and predictions have been reported previously by Varney and Bousfield (2018).

Model Description

When two pigments move relative to each other, a restoring force is calculated to pull them together based on the local strain of the polymer between them. The force equation used here takes on the form

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

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where *F* is the tensile force between particles, *A* and *B* are parameters that depend on the pure binder properties, 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 particle depends on the pigment volume fraction (PVC) that 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 the data of Zhu *et al.* (2014), the model provides a non-linear response as others have reported. If particles move closer to each other compared to the initial gap, a repulsive force is applied to keep the particles from overlapping. This repulsive force is linear and depends on the compressive strain. The model in the current form neglects the viscous effects and shear effects, but these can be incorporated in a straight forward way if needed.

For the 2D model, spheres are assumed to be confined to a monolayer, as depicted in Figure 1. Spheres on the two sides of the simulation are not allowed to move in the vertical direction, but they are allowed to slide in the horizontal direction. Spheres are "pressed" into the region keeping the minimum separation of spheres to be around 0.5% of the radius. In the pull up zone, a group of particles are assigned an upward velocity. For the results here, the up zone has a width of 20 units which is smaller than depicted in the figure. Similar conditions are imposed for the 3D case. The size of the holding zone and pull up zone has minimal influence on the results as long as the distance from the zones is large compared to the zones themselves. Similar conditions are set for the 3D model: the bending of a 3D case is shown in Figure 2.

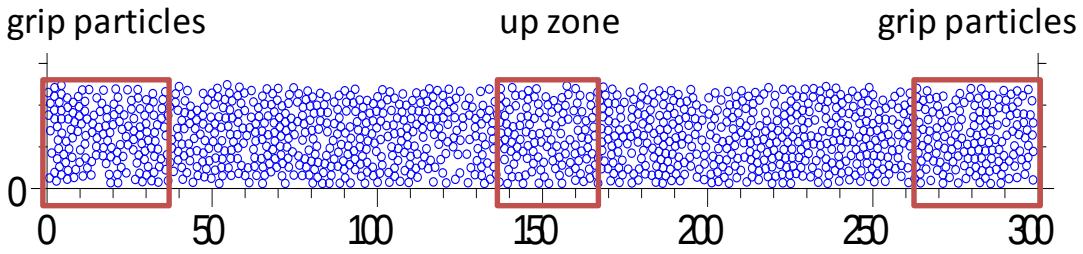

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Figure 1. Simulation set up for the 2D model for the three point bending case.

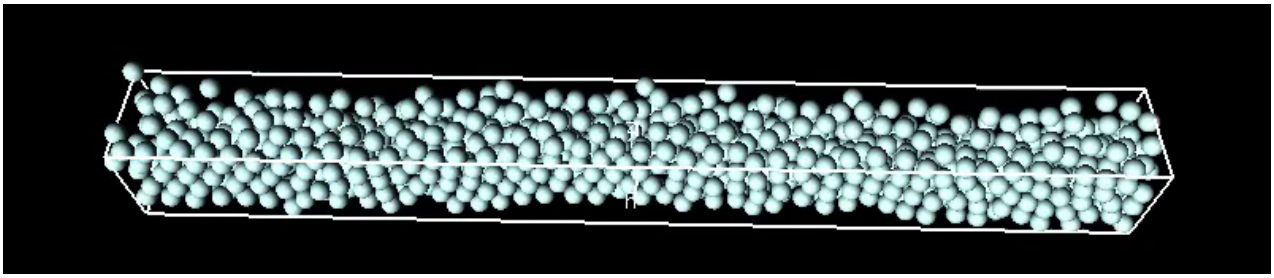

Figure 2. Three dimensional situation for uniform spheres packed in a 10x10x100 cell. Particles here have undergone upward deflection.

In both cases, as some particles are forced to move from their equilibrium position, a force is calculated from Eq. (1). This force is used to update neighboring particle velocities and positions with a numerical integration. The net force on the particles that are forced to move upward balances the sum of the forces on the particles that are not allowed to move. From these forces, the flexural stress and strain can be calculated as

$$\sigma_f = \frac{3PL}{2bd^2} \tag{2}$$

$$\varepsilon_f = \frac{6Dd}{L^2} \tag{3}$$

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 strain, σ_f, reported here is made dimensionless with the elastic modulus of the binder, and the same applies for the stress, ε_f. The goal is to predict the bending behavior of these systems and to predict the crack propagation. For the 2D case, the width of the sample is one particle diameter.

A typical result is shown in Figure 3. As a group of particles moves from the initial position, the forces are transmitted through the particles to generate a force throughout the sample. At some point, the local strain of the sample exceeds the strain to failure of the binder, a crack propagates, and the sample breaks. This general behavior and the shape of the response is quite similar to the experimental data. The model predicts the elastic modulus of the coating layer from the initial slope of the response as well as the maximum stress and the strain to failure.

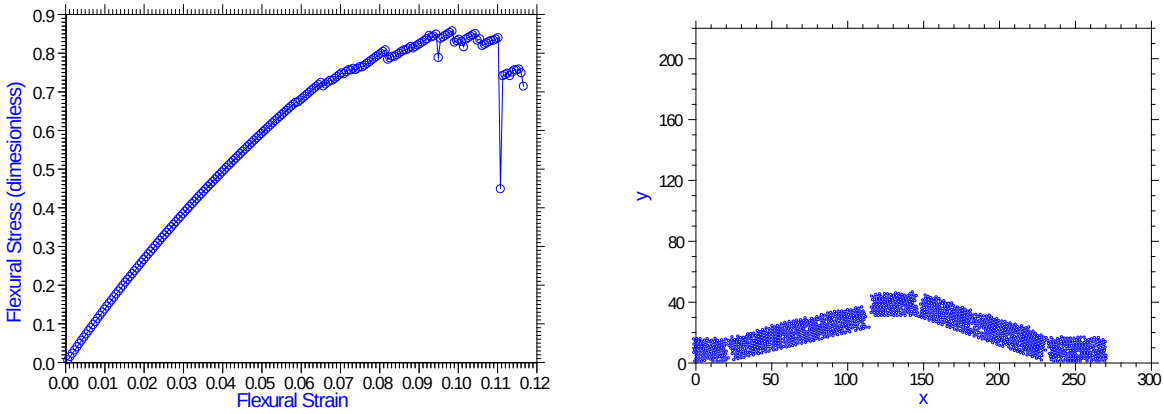

Figure 3. Flexural strain and stress predicted by the simulation (left) and crack of the coating layer (right).

The deformation and local forces in the 3D case are shown in Figure 4 for a typical case. In the region that is forced upward, a tensile force is generated. Also, near the region where particles are only allowed to slip in the horizontal direction, a tensile force is generated between particles.

Results

The predicted flexural modulus, maximum stress, and strain at failure are shown in Figures 5-7 for the 2D and 3D cases, as well as the experimental data for various latex content of the binder system, for a PVC of 63% in all cases. The different ratio of latex and starch results in different values of A and B in Eq. (1) as well as a different strain to failure of the binder itself. This PVC should be near the critical case where there is just enough binder to fill all of the pores in the system.

Both the 2D and 3D models predict the correct trends: as the latex content decreases, the coatings become more brittle. The 2D model over predicts the elastic modulus and the maximum stress to some

degree, but the predictions are in the correct range. This difference may be in the method within the code to determine the particles that are considered connected by binder bridges or by the way the particles are packed within the initial structure. Both models over predict the strain to failure, in Fig. 7: this may be due to minor imperfections in the coating layers in the experiments causing the sample to failure earlier than they would in theory.

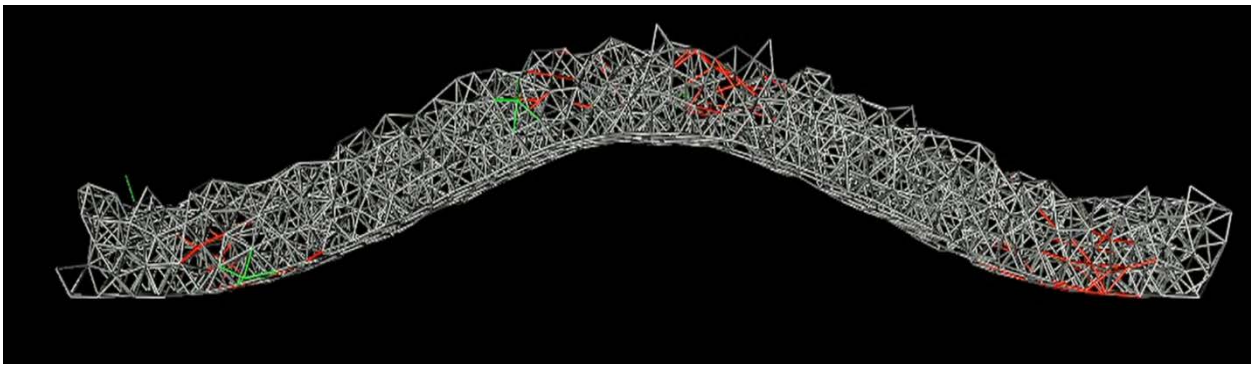

Figure 4. Connections between spheres in the 3D case. Red shows tensile forces and green compression.

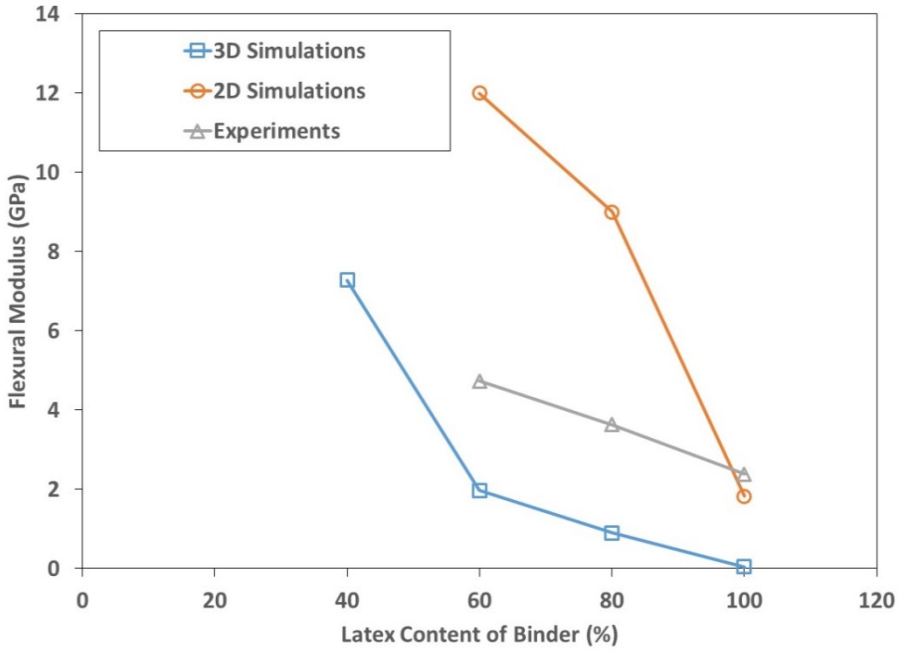

Figure 5. Flexural modulus predicted by the 2D and 3D models compared to the experiments.

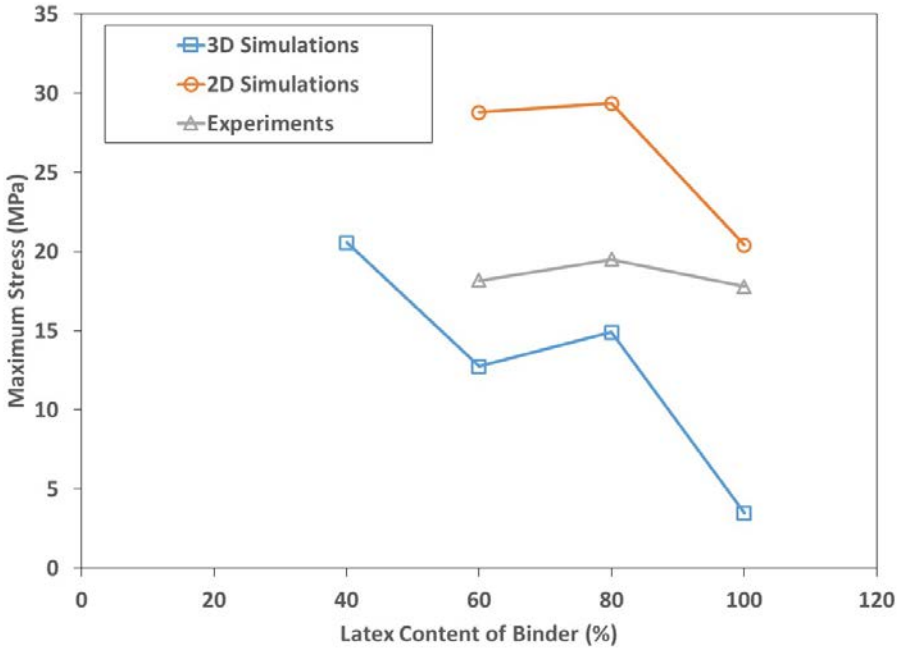

Figure 6. Predicted maximum stress of the systems compared to the experiments.

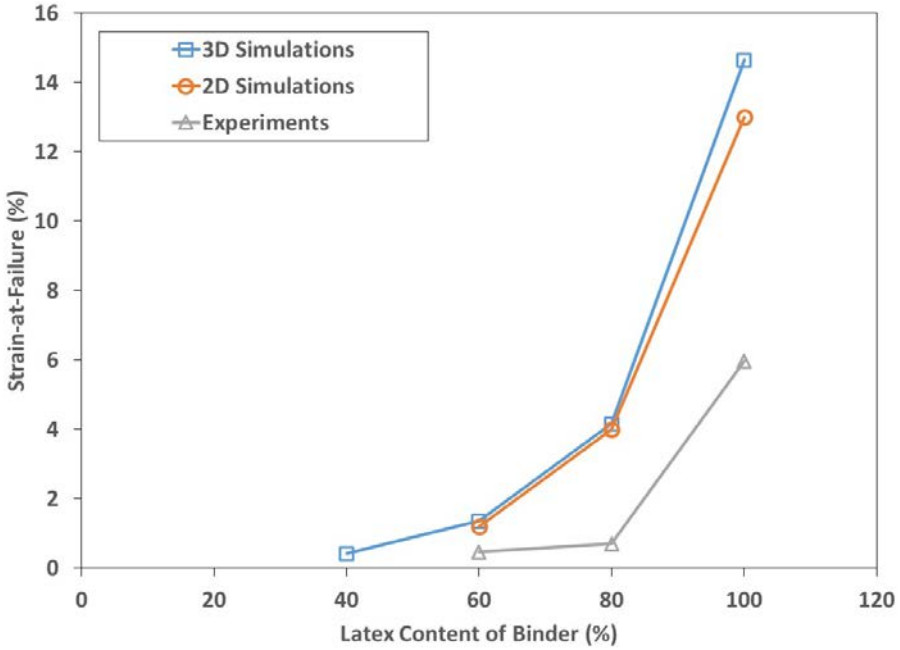

Figure 7. Predicted strain to failure compared to the experiments.

A number of assumptions are used these simulations such as perfect adhesion between the binder and the pigment, the initial packing of the particles is similar to that of the real case, and that the starch and latex are forming a uniform material. In addition, these results are for a uniform spherical case while

pigments in the experiments have a wide distribution. Work is continuing to understand these various issues.

Acknowledgements

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References

Alam, P., Toivakka, M., Carlsson, R., Salminen, P., and Sandas, S., "Balancing Between Fold-Crack Resistance and Stiffness", Journal of Composite Materials, Vol. No. 43, No. 11, pp. 1265 – 1283, 2009

Azadi, P., Farnood, R., and Yan, N., "Discrete Element Modeling of the Mechanical Response of Pigment Containing Coating Layers Under Compression", Computational Materials Science, Vol. 42, pp. 50 – 56, 2008.

Barbier, C., Larsson, P-L, Ostlund, S., "Numerical Investigation of Folding of Coated Papers", Composite Structures, No. 67, pp. 383 – 394, 2005.

Najafi, S.M.H., Tajvidi, M. and Bousfield, D.W., 2018. Production and mechanical characterization of free-standing pigmented paper coating layers with latex and starch as binder. *Progress in Organic Coatings*, *123*, pp.138-145.

Ratto, P., "Mechanical Properties of Coating Layers", Journal of Pulp and Paper Science, Vol. 30, No. 12, pp. 335 – 340, 2004.

Toivakka, M.., Alam, P, Touaiti, F., Oravilahti, A., Oravilahti, T., Knuutinen, J., Nilsson, R., Pahlevan, M., Ahokas, M. and Wilen, C. Understanding coating strength at the molecular and microscopic level. *TAPPI J.* 14(6):373-383, 2015.

Varney D. and D.W. Bousfield, "Discrete Element Method to predict coating failure mechanisms", TAPPI J., 17(1), 21- 30 (2018).

Zhu, Z., Salminen, P., Chen, G., and Toivakka, M., "Mechanical Properties of Pigment Coating Composites Containing Starch", 13th TAPPI Advanced Coating Fundamentals Symposium, October 7 – 9, 2014, Minneapolis, Minnesota.