NOTE

On the Cohesion of Clay and CaCO₃ Coatings

Despite its importance in printing, folding, or gluing, surprisingly little work has been reported on the subject of the mechanical strength of a paper coating, no doubt on account of its complexity. A recent publication¹ lists the many factors that influence coating strength, among them: pigment and binder mechanical properties; adhesion between pigment, binder and fiber; and the structure of the coating. In this short communication we would like to contribute information regarding the latter, specifically, the effect of the porous structure of the coating on its strength in the transverse direction.

The mechanical strength of a material decreases when voids, which are not load-bearing, are introduced. Accordingly, one might expect, at a given binder level, denser coatings to be stronger. Indeed, long ago, Cobb^2 and Willet and Marchetti³ showed that bulk was an important component of coating strength.

Ground $CaCO_3$ coatings, at a given binder level, have a better IGT pick resistance than clay coatings.⁴ There are two main differences between clay and calcium carbonate coatings. One is, of course, a difference in particle shape: Clay is plate-like; ground calcium carbonate particles are isometric. The other is a difference in density. Even though very few data have been published, one may speculate that $CaCO_3$ coatings are denser than clay coatings since they can be formulated at higher solids, an indication of better packing ability.

Can one separate the effect of coating void fraction from that of particle shape on the z-direction strength? We have attempted to do so.

EXPERIMENTAL

Coatings made of # 1 clay (Hydrafine, Huber) or/and ground $CaCO_3$ (Hydrocarb 65, OMYA) and an s/b latex (P-756, Polysar) with no other additives were applied at 60% solids on mylar film and dried at room temperature. The binder content was varied from 0 to 20 parts per hundred parts (pph) of pigment.

The void volumes of the coatings were measured with the oil absorption technique described in Ref. 5. Light-scattering coefficients were calculated from measured reflectances at 458 nm. Coating cohesion in the transverse direction was measured on an inclined plane apparatus (Fig. 1) similar to the rolling cylinder tackmeter of Voet.⁶ The mylar coating is secured with double-sided adhesive tape to a wedge-shaped holder that can be raised or lowered. Another adhesive tape covers the coating and is attached to the roller. As the roller rolls down over the coated surface it delaminates the coating and loses an amount of kinetic energy equivalent to the energy of delamination of the coating, plus a small friction loss which was neglected.

RESULTS AND DISCUSSION

Figure 2 confirms that there exists a significant difference in void fraction between clay and ground $CaCO_3$ coatings. This can also be concluded from Figure 3 which shows the variation in light-scattering coefficient with binder content. The light-scattering efficiency of a paper coating is much influenced by its porous structure.⁵ Since the two pigments have similar particle size distribution (Fig. 4) and refractive index, it would appear that the difference in light-scattering comes mainly from a difference in pore size.

In Figure 5, the rupture energy is seen to increase with binder level at a faster rate for $CaCO_3$ than for clay. Finally, the rupture energy values are plotted as a function of the void volume fraction in Figure 6. We see that a common curve at the lower void fraction end (higher binder content) separates into two different curves at higher void fraction (lower binder content). This indicates that, above a binder level sufficient to provide a cohesive composite structure, the void



Fig. 1. Inclined plane apparatus. (A) roller; (B) Sample holder (adjustable wedge in order to bring sample surface in contact with roller surface).

fraction determines the strength of the composite. Void fraction can therefore be considered to explain part of the difference in cohesive strength between clay and $CaCO_3$ coatings.

Microscopical examination of the ruptured coatings provides another clue. First, we see in Figures 7 and 8 that failure was not by uniform delamination into two layers as we expected. Rather, failure took place in successive, and presumably impact-type, jumps appearing as streaks or grooves perpendicular to the peeling action. The groove frequency increased with roller speed but there was no obvious systematic correlation with speed. The same phenomenon was observed when IGT tests were performed on these coatings with a high viscosity oil.



Fig. 2. Void fraction of clay and $CaCO_3$ coatings vs. binder content: (•) clay; (•) $CaCO_3$.



Fig. 3. Light-scattering coefficient vs. binder content: (•) clay; (•) CaCO₃.

Examination at higher magnification revealed a significant difference between the failures in the clay and in the carbonate coatings. In the clay coatings, the failure line was rather shallow. In CaCO₃ coatings, failure went deep down, near the mylar substrate. When formed from a well-stabilized coating suspension, the clay plates tend to orientate themselves in the plane of the coating as can be seen on the micrograph. One may therefore expect the stress concentration to cause rupture along x-y planes. With the more isometric CaCO₃ particles, random stress distribution and random failure might be expected. It appears that the plane of failure is roughly at 45° to the z-direction down close to the mylar interface.

Our observations, in particular the in-plane failure of clay coatings under transverse stress, support the model proposed by Parpaillon et al.⁷ Analyzing results of in-plane strength measurements, they proposed a model for clay coatings where the binder accumulates at the plate ends rather than between the plate faces.



Fig. 4. Particle size distribution of clay (\bullet) and CaCO₃ (\blacktriangle) pigment used.



Fig. 5. Rupture energy vs. binder content for clay (\bullet) and CaCO₃ (\blacktriangle) coatings.





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Fig. 7. Scanning electron micrographs of ruptured clay coatings at two magnifications (10 pph latex).

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Fig. 8. Scanning electron micrographs of ruptured calcium carbonate coatings at two magnifications.



Fig. 9. Rupture energy of coatings made from pigment blends.



Fig. 10. Rupture energy vs. void fraction: (\bullet) clay; (\blacktriangle) CaCO₃; (\blacksquare) blend 5 pph; (\blacklozenge) blend 10 pph.

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At this point one may be curious to know what happens if the two pigments are blended. Figure 9 shows that the rupture energy decreases monotonically as $CaCO_3$ is replaced by clay while the relationship between rupture energy and porosity is confirmed (Fig. 10).

SUMMARY

The lower cohesive strength of clay coatings at a given binder level, compared to $CaCO_3$ coatings is due partly to a higher void fraction that makes the coating more fragile, and partly to the different failure mechanisms that result from the different pigment shape.

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P. Lepoutre T. Hiraharu*

Pulp and Paper Research
Institute of Canada
570 St. John's Boulevard
Pointe Claire, Quebec, Canada H9R 3J9

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*Visiting scientist from Japan Synthetic Rubber Co., Ltd.