Experimental investigations into fold cracking of double coated barrier dispersion coatings

JANET PRESTON, GAVIN BUTLER-LEE, CHRIS NUTBEEM, AND CHRIS DISBERRY

ABSTRACT: The trend for replacing single-use plastics with fiber-based barrier coated board packaging has prompted a significant amount of research. There are many proposed ways of providing suitable packaging for applications like food service. Among these are dispersion coated barriers on board, as well as laminated boards that can be produced using conventional polyethylene (PE) or new biodegradable plastics. Minerals have also been shown to be suitable additives to these coatings for improving barrier performance through surface chemistry and by increasing the tortuosity of the pathway through the barrier layer. They also improve the cost effectiveness of the layer by lowering the material cost and raising the solids content, and by improving hold out of the functional layers, leading to a reduction in the amount of barrier coating needed to meet a given performance requirement. Minerals can also aid in the barrier handling in terms of rheology and reduced "stickiness," as well as blocking of the films. When incorporated as fillers into extruded films, improved adhesion of the film to the board has been reported.

One of the remaining challenges is the potential for cracking at the fold during converting and the loss of barrier performance that this can lead to. In this work, we systematically looked at the impact of mineral type and level in a dispersion coating. We assessed the differences in performance resulting from different coating application methods for the precoat layer by looking at the cracking tendency and loss of barrier functionality after folding for both the precoat alone and the final double coated sheets. Barrier results include moisture vapor transmission rate (MVTR), viscous vegetable oil, and the fluid blue stain in industrial methylated spirits (IMS) and Cobb water absorption, both before and after folding.

Application: This work continues to build on previous work that has shown some benefits in barrier performance by including minerals, due to a longer tortuous path for the oil or water vapor to move around. However, we found that it was not just important to look at the sheet as coated before folding or creasing. The process of converting puts additional strain on the sample, and we found that a moderately platy kaolin outperformed an extremely platy one after the folding was carried out and the barrier properties assessed. Therefore, the converting step should also be considered during the formulation development. This work should help guide development teams in their choice of mineral if it is added, as well as a suitable amount.

In the development of fiber-based packaging, application of a suitable barrier layer to prevent migration of moisture vapor, grease, oil, and water is a key requirement if the substrate is to replace single-use plastic packaging material [1]. Water-based barrier dispersions have the advantage over polyethylene (PE) layers applied to paper and paperboard substrates in that they are more easily recycled and re-pulped and can be applied as thinner layers. Mineral addition to the dispersion gives benefits in barrier performance, solids increase, less blocking of the final surface, and lower cost. Devisetti et al. [2] showed that higher shape factor kaolin, when incorporated into a biopolymer, gave superior barrier performance to lower shape factor kaolin. It may often be the case that a combination of polymers and minerals are able to give a good barrier when the coating is flat. However, other constraints must be fulfilled; for example, maintaining barrier functionality after folding.

Cracking at the fold is an important aspect of magazine finishing and has been investigated extensively [3,4]. Important properties were found to include pigment type and ratio, the binder level, coat weight, and calendering pressure. The base structure was also an extremely important aspect of the folding. Optimization of these properties led to a decrease in unsightly white cracks after folding, which can destroy the visual impact of the converted product. However, the visual aspects of cracking in graphic articles are less critical than those occurring in a barrier paper [5], where prevention of migration of water vapor, oil and grease, and oxygen are vital for the sheet functionality.

Zhu found that plate-shaped pigments gave the best moisture vapor transmission rate (MVTR) results before folding, but tended to generate large cracks and poor water vapor barrier properties after folding [6]. Spherical plastic pigments yielded smaller cracks. It has also been found that the correct alignment of the particles is vital to achieve the required barrier properties [7]. Lower elastic modulus (more flexible) pigments (e.g., talc) improved crack resistance compared to coatings made with higher modulus pigments (e.g., kaolin) [8]. Adding a flexible latex reduced the crack formation and led to good barrier performance even after folding.

There has also been a considerable amount of work published that has looked at the different coating structures achieved with different coating technologies and varying pigment morphologies. In the case of blade coating of platy kaolin, the platelets are well aligned and generally give a smooth surface, although the coating thickness distribution can be poorer, with areas of thick and thin coating layer. Conversely, a coating application with a roller such as a metered size press results in a rougher surface and less aligned particles, but a more even coating distribution. This study includes some measurements of precoats applied by a rod compared to those applied with a blade.

This work was carried out to have a systematic data set to better understand the properties of mineral type, loading level in the formulation, and coater application system on the barrier performance and the barrier properties after folding.

METHODS

Precoating

One set of coating precoats was applied with a blade using a trademarked Helicoater (English China Clay International; St. Austell, UK) [9] at a speed of 500 m/min and a blade angle of 45° to obtain coat weights of 7 g/m² ± 0.2 g/m². These were applied to a freesheet base sheet of ~45 g/m². The coatings were dried using 30 s of hot air and infrared (IR) drying, and 30 s of cold air.

The second set of precoats was applied to the same 45 g/m² base, but this time by using rod coating as simulated by wire wound rods from RK Printcoat Instruments (Royston, UK) with a hand draw down (HDD) technique. The precoats were applied at similar coat weights to the blade application and were oven dried at 80°C for 10 min. All samples were conditioned at 21°C and 50% relative humidity (RH) before being tested.

A functional barrier dispersion coating formulation topcoat was subsequently applied at 7 g/m² ± 0.2 g/m² coat weight onto all the precoats by the rod hand draw down technique.

Folding

The sheets were initially evaluated in the flat, unfolded condition. When samples are creased (either 90° or 180°), an inside fold implies the fold brings the coated surfaces together. An outside fold is the opposite, bringing the uncoated sides together. This means an outside fold should be more strain on the coating than an inside fold and be more likely to fail. A custom metal frame was used to ensure a consistent 90° fold (paper folded over stainless steel angle iron "L" shape and then another "L" pressed on top). The 180° fold was made by hand, with the same metal "L" pressed over the fold to ensure a sharp crease.

Cracking tests

Panek and Hart provided a review of coating cracking and barrier integrity, which contained a summary of methods used to study cracking of paperboards [10].

In this study, the sample failure after folding was evaluated using three different methods. The first was the traditional black printed surface that gives a visual indication of cracking in the coating layer. The second and third evaluation methods involved more stringent tests for the integrity of the fold for barrier purposes, which was determined by using two stains: Sudan Blue dye in industrial methylated spirits (IMS), and Sudan Blue dye in vegetable oil.

To carry out second and third methods, three drops of the test liquid, either Sudan Blue in IMS or in vegetable oil, were applied to the surface using a plastic disposable pipette. After 3 s, this was wiped off with a cotton wool pad and the penetration assessed. Due to the Sudan Blue dye in both the IMS and the vegetable oil, any penetration into the surface is visible as a blue stain. The amount of staining (e.g., pinholes or complete penetration) gives an idea of the cracking present. No staining implies no penetration. The MVTR evaluation was then carried out on the samples following TAPPI Standard Test Method T 464 "Water vapor transmission rate of paper and paperboard at high temperature and humidity." Finally, the ability to hold out water was assessed using a 60 s Cobb test according to ASTM D3285-93 "Standard test method for water absorptiveness of nonbibulous paper and paperboard (Cobb test)."

MATERIALS

Minerals and application

The minerals in **Table I**, which are commonly seen as components of barrier formulations, were evaluated in some simple latex/mineral combinations that were applied as precoat layers to the base sheet.

The three different barrier minerals were combined with a specialty latex (a copolymer formed from ethylene and an unsaturated carboxylic acid monomer), and the mineral:latex ratio was selected as 20:80, 40:60, and 60:40. A mineral-free control sample was applied to the base using the polymer component alone and with no pigment addition. However, this polymer application was only possible using the HDD rod coating technique, and it was not possible to successfully apply using the Helicoater. Application of the topcoat was carried out using a proprietary barrier formulation (a heat sealable moisture barrier combining the previously-mentioned latex and a phyllosilicate mineral) on top of all the precoats using the rod HDD technique.

Mineral Type	% < 2 µm Content by Sedigraph	Shape Factor
Moderately Platy Kaolin	83	20
Platy Kaolin	48	40
Platy Talc	46	100

I. Minerals used in the precoat layer.

	Pigment:Binder	Solids, wt%	pH	Initial Viscosity before Thickener Addition, Brookfield 100 rpm viscosity/mPas
Binder	0:100	37	9	164
Moderately Platy Kaolin	20:80	41	9	98
	40:60	48	9	86
	60:40	55	9	120
Platy Kaolin	20:80	40	9	106
	40:60	48	9	92
	60:40	57	9	92
Platy Talc	20:80	42	9	128
	40:60	49	9	206
	60:40	59	9	792

II. Precoating coating color properties.

Coating color properties

Portions of a synthetic thickener were added to all of the precoats to raise the coating viscosity to around ~1000 mPas to allow good coating on the Helicoater, as **Table II** describes.

RESULTS

The traditional cracking at the fold evaluation carried out on the printed surfaces showed no visible cracking for either the precoats alone or after the topcoat application. This is likely to be due to the relatively high polymer level in the formulations compared to traditional graphic coating colors.

However, when the Sudan Blue dye in IMS and the Sudan Blue dye in vegetable oil were applied to the precoated paper after folding, there was penetration of both these fluids into all of the samples. **Table III** describes the degree of penetration assessed. Differences in penetration into the final double coated sheets were observed for each of the various folding configurations and coating recipes, as described in **Table IV** and **Table V**.

Samples with the binder component only (0:100) were applied successfully using the HDD rod technique. As with the mineral-filled samples, the binder layer alone as a precoat gave no barrier properties and resulted in a full penetration of the stains. However, after application of the topcoat, they showed no cracking or stain penetration after folding.

1	No penetration.
2	Slight fail, with one or two tiny pinholes.
3	Slightly more substantial failure, with more pinholes.
4	Complete failure, with penetration all along the crease.

III. Legend for penetration rating of blue stain in IMS used in Table IV and Table V.

These results (Table IV) indicate that there was no penetration of the blue stain or vegetable oil on either the flat coated surfaces or after the 90° fold. However, some penetration differences were observed after the more critical 180° folding, especially with the challenging blue stain in IMS. The pigment is less flexible than the binder, and so increasing the pigment gives slightly poorer performance (seen with the 60 parts per hundred [pph] pigment), especially after the severe folding.

			Blue Stain/IMS					Blue Stain/Vegetable Oil				
			Flat	90°		180°		Flat	90°		180°	
				Inside Fold	Outside Fold	Inside Fold	Outside Fold		Inside Fold	Outside Fold	Inside Fold	Outside Fold
Moderately Platy Kaolin	20 pph	PC1	1	1	1	1	3	1	1	1	1	1
	40 pph	PC2	1	1	1	2	4	1	1	1	1	2
	60 pph	PC3	1	1	1	2	4	1	1	1	1	2
Platy Kaolin	20 pph	PC4	1	1	1	1	4	1	1	1	1	2
	40 pph	PC5	1	1	1	3	4	1	1	1	1	2
	60 pph	PC6	1	1	1	4	4	1	1	1	1	2
Platy Talc	20 pph	PC7	1	1	1	2	2	1	1	1	1	2
	40 pph	PC8	1	1	1	2	2	1	1	1	1	2
	60 pph	PC9	1	1	1	2	3	1	1	1	1	3

IV. Stain penetration results for blade coated precoat (PC) with the Helicoater, after the barrier topcoat application.

			Blue Stain/IMS					Blue Stain/Vegetable Oil				
			Flat	90°		180°		Flat	90°		180°	
				Inside Fold	Outside Fold	Inside Fold	Outside Fold		Inside Fold	Outside Fold	Inside Fold	Outside Fold
Moderately Platy Kaolin	20 pph	PC1	1	1	1	1	1	1	1	1	1	1
	40 pph	PC2	1	1	1	1	3	1	1	1	1	2
	60 pph	PC3	1	1	1	1	4	1	1	1	1	2
Platy Kaolin	20 pph	PC4	1	1	1	1	2	1	1	1	1	1
	40 pph	PC5	1	1	1	1	3	1	1	1	1	1
	60 pph	PC6	1	1	1	2	4	1	1	1	1	2
Platy Talc	20 pph	PC7	1	1	1	1	1	1	1	1	1	1
	40 pph	PC8	1	1	1	1	3	1	1	1	1	2
	60 pph	PC9	1	1	1	2	4	1	1	1	1	2

V. Stain penetration results for rod coated precoat (PC) with the hand draw down (HDD), after the barrier topcoat application.

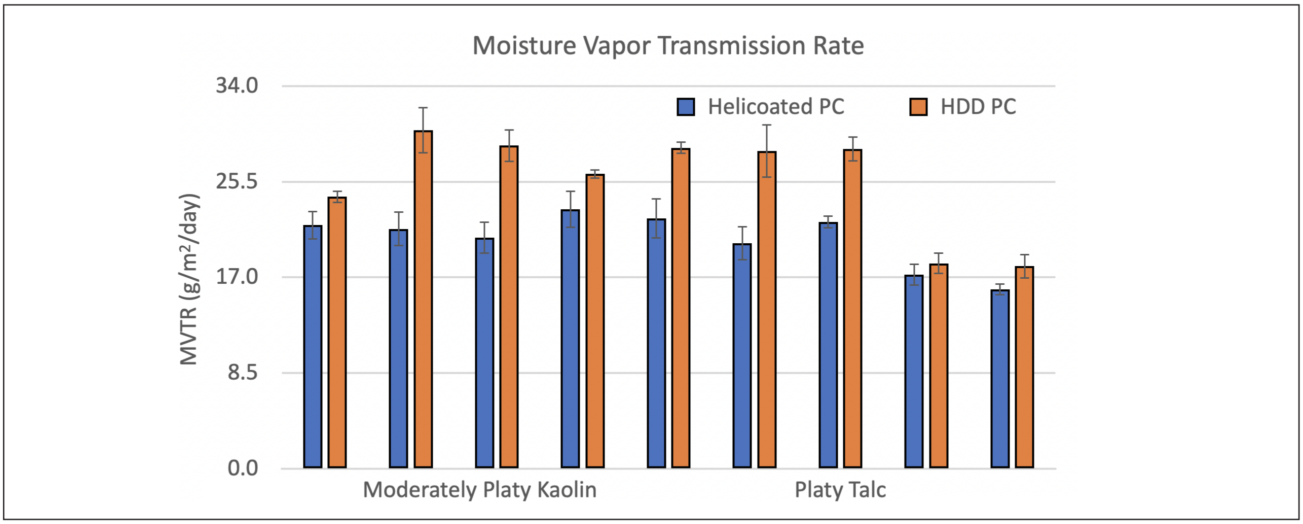
The results (Table IV) show that on the inside fold (where the coating is compressed), there is less stain penetration than when looking at the outside of the fold, where there is coating elongation. It can be seen that the moderately platy kaolin is the best for inside fold with the blue stain. On the outside of the fold, platy kaolin is the worst, and the more flexible talc [8] is slightly better than the moderately platy kaolin.

In Table V, rod application, there were also no issues with any of the flat samples or those folded with the 90° test. The moderately platy kaolin precoat also passes the severe 180° folding when looking at the inside fold. When looking at the outside fold, the penetration of the stains was all more pronounced, and we could see that the platy kaolin is the worst; talc and moderately platy kaolin are the same. However, the precoats applied by the rod technique were better than those applied using a blade with the Helicoater.

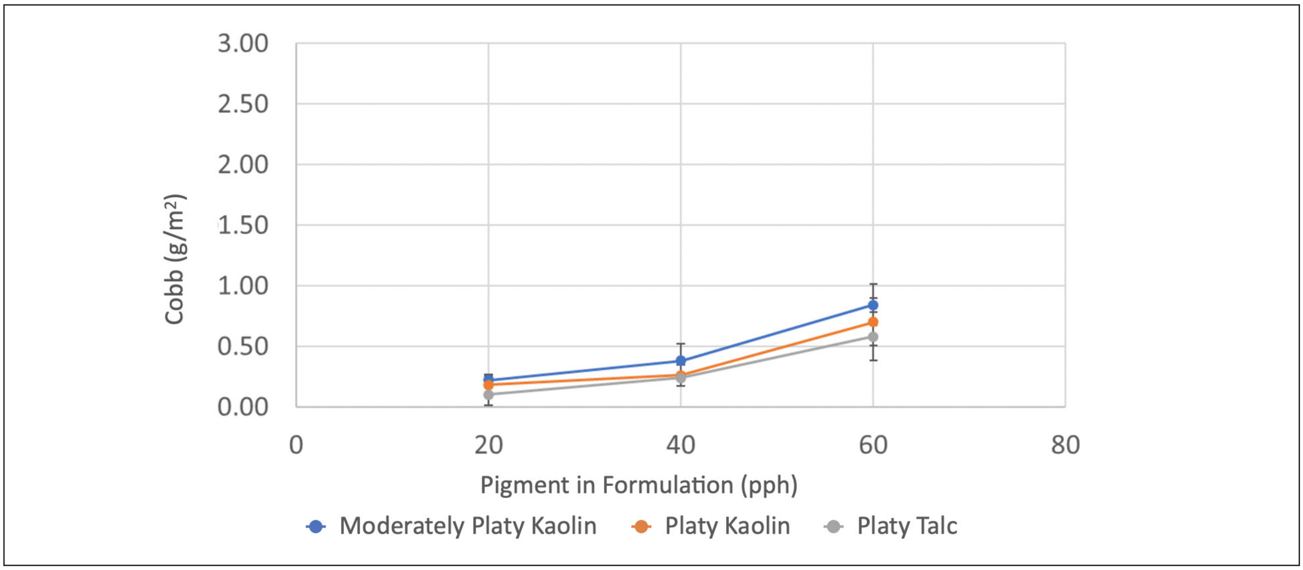
MVTR test results

The MVTR testing was carried out on the unfolded samples, and the results are shown in **Fig. 1**.

The Helicoated sheets with more "aligned" platelets have a generally better MVTR performance (lower). The



1. The moisture vapor transmission rate (MVTR) testing of the final top coated paper with varying precoat (PC) layers (flat samples; HDD: hand draw down, and Helicoated).



2. Cobb test results from Helicoater (blade coated) precoat after top coat application, no folding (uncoated base had a Cobb of 20.5 g/m², and the double coated, but polymer only, precoat layer had a Cobb of 0.98 g/m²).

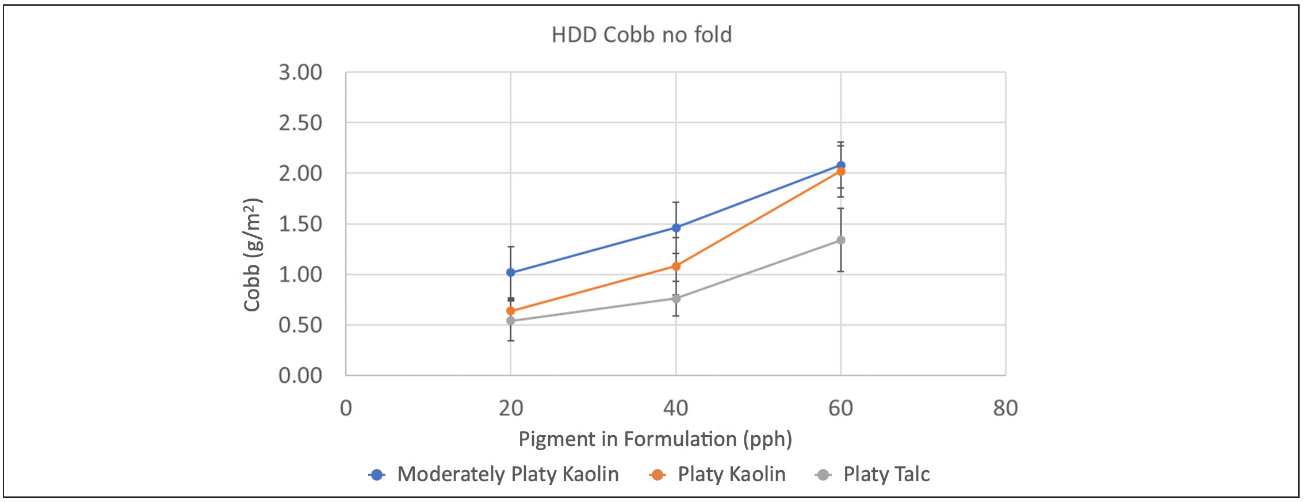
best results are given by the higher levels of talc in the formulation (hydrophobic and platy). In many barrier studies, better barrier properties are observed with higher binder levels. In the case of hydrophobic talc, this is not necessarily so. There is not much difference in the performance of the kaolins at any of the pigment:binder ratios. The unfilled polymer precoat layer applied using the HDD technique and then topcoated had an MVTR of 31.8 g/m²/day, which is slightly worse than when mineral was included.

Cobb test results

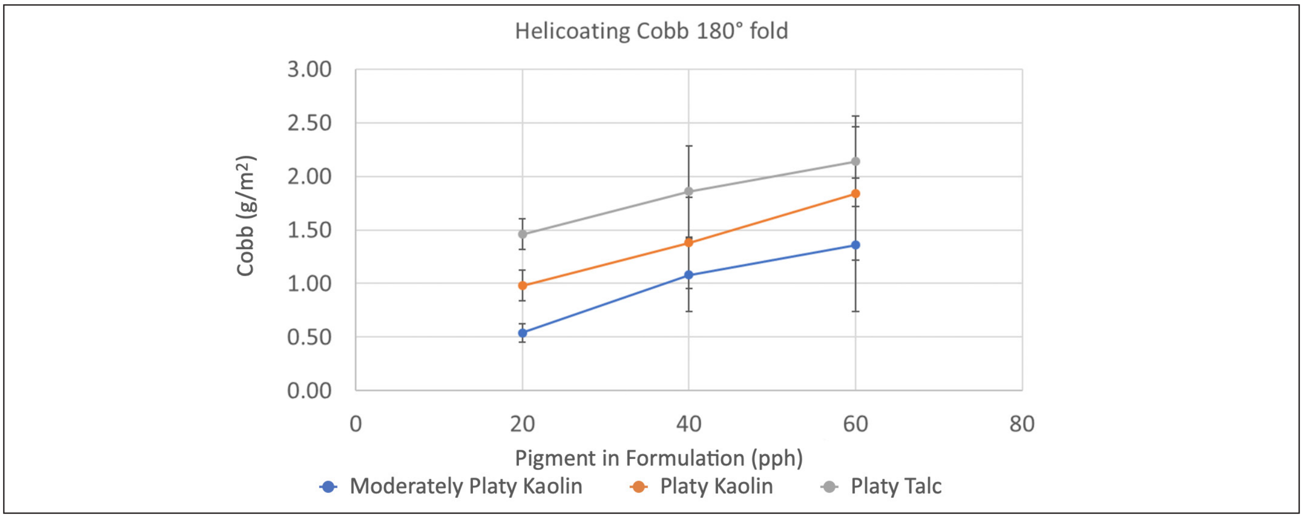
The water absorption tendency was evaluated using the Cobb test after 60 s for both the unfolded samples and those folded at 180°. The results are given in **Fig. 2–5**.

There is generally a better Cobb value (lower) on the flat samples than the folded samples

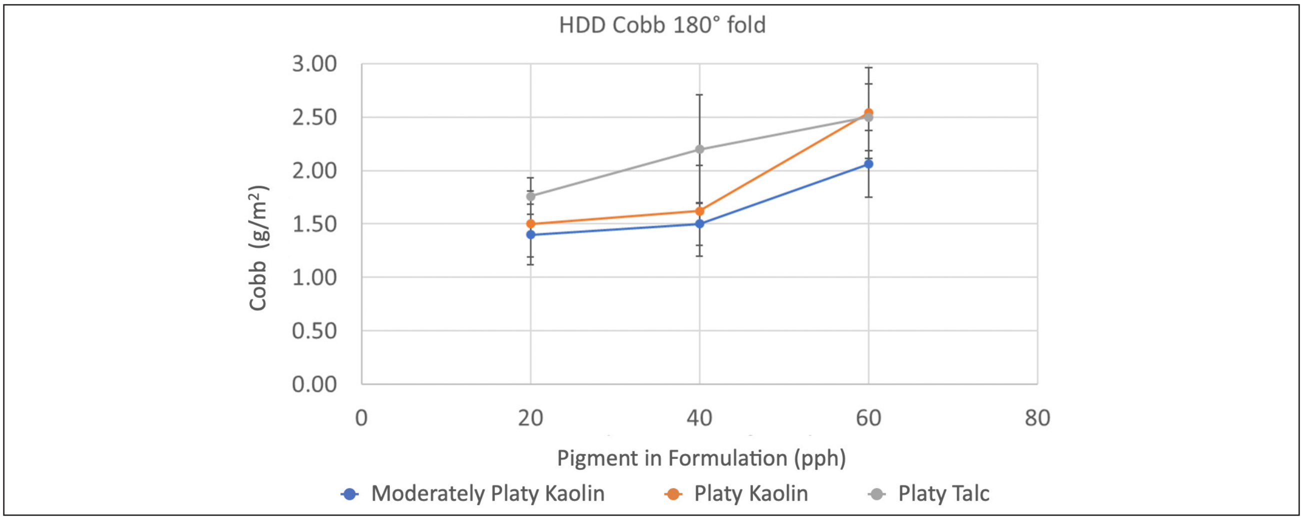
For the flat samples, the blade coated precoats are generally giving a better response than the rod coating, due to better alignment of the pigment particles. As the level of pigment increases, the performance becomes worse. However, it should be noted that the values are still very low compared to the base sheet. The more platy the pigment, the better the Cobb values generally (talc > platy kaolin > moderately platy kaolin) for flat Cobbs. However, the opposite is true after the samples have been creased, and now the best performance is from the more moderately platy kaolin. It is interesting to note that the error bars and variability of the result also increase after sample folding.



3. Cobb test results from rod coated (using hand draw down, HDD, method) precoat after top coat application, no folding (uncoated base had a Cobb of 20.5 g/m², and the double coated, but polymer only, precoat layer had a Cobb of 0.98 g/m²).



4. Cobb test results from Helicoater (blade coated) precoat after top coat application, 180° crease (uncoated base creased had a Cobb of 21.5 g/m²)



5. Cobb test results from rod coated (using hand draw down, HDD, method) precoat after top coat application ,180° crease (uncoated base creased had a Cobb of 21.5 g/m²).

SUMMARY AND CONCLUSIONS

The traditional cracking at the fold evaluation for graphic printing purposes showed that all of the samples visually gave a good performance, and no unsightly white cracking was visible after the folding either on the precoated surface alone or on the final double coated samples. The visual aspects of cracking are therefore much more easily achieved than the more stringent demands of barrier functionality. When the barrier tests were carried out on the precoat layers alone, all failed (instant penetration of blue stain in IMS and blue stain in vegetable oil), showing the importance of having the barrier layer on the top surface.

After the topcoat had been applied, differences were observed in the performance, depending on the folding (degree of folding and inside or outside of the fold); the mineral type and level; and the fluid used in the test and on the precoat application system. The results show:

1. There was a minimal difference in barrier properties with mineral loading. All of the precoats are probably fully bound, and this would be expected if they were beneath the critical pigment volume concentration. The Cobb values were marginally worse at 60 pph pigment.
2. There was good barrier performance on the samples when flat or only creased at 90°.
3. The outside of the fold when the papers have been creased at 180° is the most problematic, and so the stretching of the coating is more critical than compression.
   a. The only coating to pass this was the moderately platy kaolin at 20 pph in the precoat.
4. The MVTR of the flat samples is better (lower values) for the blade coated precoats than for the rod coating, probably due to better particle alignment. However, the rod applicator is superior to the blade when looking at the barrier performance after folding.

ABOUT THE AUTHORS


Preston


Butler-Lee


Nutbeem


Disberry

Barrier coating of fiber based packaging is a hot topic currenty in our industry and an area of growth as people seek to replace single-use packaging. So, our study topic on dispersion coated barriers is of current interest.

There has been quite a lot of work published about making barriers for oxygen, moisture, and grease; however, much of the work in the past has focused on the performance of the flat sheet. In this work, we sought to look a little more into the cracking potential and impact on barrier properties after the product has been folded and converted, which is vital for use in the real world. Other authors have also carried out similar work, and this study agrees with their findings. A particular focus for us was to also look at different coating applicator types, as well as different mineral addition levels and mineral types.

One of the most difficult aspects of this study was in folding the samples accurately and consistently to give a reproducible result. This was achieved by using a device that holds the sheet at the correct angle, with a dedicted and skilled scientist to carry out the folding.

Through this study, it was good to be able to see and understand more about the different variables that impact fold cracking and barrier, as well as the amount of impact each different aspect had on the final result. It was surprising to find that some of the benefits of the extremely platy materials were less apparent when folded samples were considered. In this case, a moderately platy material worked better.

Mills are often developing formulations for barrier coating of paperboard, and this work will help them in their choice of mineral additive. We know from previous work that mineral addition gives benefits in cost effectiveness, technical barrier performance, and ease of handling the formulation. However, using the right mineral at the right loading level is vital for an optimized product.

A next step in this research suggested by the reviewing committee was to further understand the impact of particle size and shape by going to an "extreme" end point of adding a spherical calcium carbonate to the formulation.

Preston is senior scientist, Nutbeem is Science and Technology director for Paper and Board, Butler-Lee is assistant scientist, and Disberry is development engineer for Barrier Coatings at Artemyn UK Ltd. in Par, Cornwall, UK. Email Preston at janet.preston@artemyn.com.

5. In terms of the Cobb test, the platy talc is the best, followed by the platy kaolin on the unfolded sheets. However, after creasing, the opposite is true, and the moderately platy kaolin gives the best performance.

In general, the work has shown that all the aspects of converting and application need to be considered when designing a barrier coated fiber-based product. **TJ**

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