Use of kaolin clay in aqueous barrier coating applications

SIVA DEVISETTI, GUS LEMPSINK, AND PRAKASH B. MALLA

ABSTRACT: Paper-based packaging with barrier effect, as opposed to single use plastics, is gaining more prominence for sustainability reasons. At the same time, latex- or biopolymer-based aqueous barrier coating dispersions are increasingly being adopted as a better alternative to the traditional barrier coating materials, such as wax, surface active chemicals, and polyethylene.

In this work, studies were performed to determine the influence of different kaolin clays in latex binder-based aqueous coatings on barrier properties, namely, oil and grease, water resistance, and water vapor transmission rate, by applying coatings to solid bleached sulfate (SBS) paperboard substrate in the laboratory. The aim was to explore potential benefits of using kaolin clay to replace some of the latex binder in coating and improve or maintain various types of barrier performance and blocking without negatively influencing the other performance attributes, including heat seal.

The delaminated clay with the highest shape factor provided improved barrier properties over the clays of low shape factor. The ultrafine and non-delaminated clays required significantly higher coat weights to reach satisfactory barrier properties. Coatings with different latex levels indicated that a considerably high proportion of coarse delaminated clay can be incorporated to replace latex binder, while still achieving exceptional barrier properties. Furthermore, a change in binder system was found to significantly alter the barrier properties and the role that a mineral pigment can play. The results indicate that a proper selection of binder systems for each barrier property would be required while considering the clay/latex coating systems.

Application: This research suggests that cost effective aqueous barrier coatings can be prepared using various kaolin clays to partially substitute for more expensive latex binder components. The barrier properties can be improved using a wide range of kaolin clays, but the use of high aspect ratio clay provides superior performance at reduced coat weight. This research can be utilized to decide a maximum or minimum clay loading to maintain different types of target barrier performance, as well as blocking and heat seal properties, in a single layer coating system.

An impervious barrier layer may be applied to paper and paperboard substrates for packaging applications in order to protect the contents of the package against oil and grease, moisture, water vapor, or a combination thereof. Uncoated paper and board substrates are recyclable and naturally biodegradable, and their use as a sustainable packaging material is favored, but they lack barrier properties. Traditionally, waxes, extruded polyethylene (PE), or forever chemicals such as PFAS (per- and polyfluoroalkyl substances) have been used as barrier coatings or treatments. Although waxes and extruded polyethylene can provide an excellent barrier, they present problems related to recyclability and biodegradability of the cellulosic substrate [1-4]. Polyethylene has dominated as a barrier coating material in packaging applications because of its low cost and versatile properties. However, it is challenging to recycle the PE coated packages. The PE portion that ends up in landfills is estimated to take decades to breakdown. The PFAS are known to provide good oil and grease resistance and water resistance to many of the packages in the food industry, but they present health hazards. Certain types of PFAS are found to bio-accumulate in the environment [5]. As a result, these materials are being phased out or banned in some cases due to their negative impact on recyclability, repulping, compostability, environment, aquatic lives, and human health. Therefore, sustainable alternative barrier coating materials that are safe for the environment are in favor due to legislative and consumer demands [1-4,6-17].

At an increasing rate in recent years, aqueous dispersion coatings based on latex or biopolymers are clearly emerging as an interesting alternative to the traditional extruded or surface active barrier materials. However, these coatings are either expensive or still need further improvement to be able to fully compete with the traditional extruded coatings. The use of mineral pigments such as bentonite, talc, and kaolin clay in barrier coatings is known to enhance certain barrier properties of the aqueous dispersion coatings, which has been widely published [4,6-10,12]. Most of these studies have focused on a specific barrier property of interest, and some are conceptual in nature. The water resistance of linerboard was improved with a clay and styrene butadiene latex base coat and acrylic latex top coat [1]. Multifunctional coating systems applied by a simultaneous multilayer coating process to achieve more than one barrier property was reported using various functional polymers [14]. The influence of liquid barrier coatings based on polymers was studied on heat seal, blocking, and water resistance [16,17]. The authors in previous works [3,4,7] focused on water vapor transmission rate (WVTR) using clays of various shape factors and different binders. Higher clay loadings improved water vapor transmission all the way up to critical pigment volume concentration, above which point all barrier performance was lost [4]. The coating should have sufficient heat seal properties to ensure short sealing and cooling time from packaging lines. It would be interesting to know how significantly the levels of clay loadings would impact other critical areas, such as heat seal and water resistance. Furthermore, the performance of dispersion coatings (with and without pigments) as of today is still far behind the traditional extruded coatings, and hence, any additional results would complement the existing research.

Incorporating kaolin clays in the aqueous polymer dispersion coatings can bring advantages that reduce cost and contribute to the overall performance. The objective of this work is to develop a cost-effective coating based on kaolin with an equal to or improved performance over the coating based solely on latex-based aqueous dispersion. Some of the challenges encountered with the kaolin-based coating will also be discussed.

MATERIALS AND METHODS

A total of five kaolin clays with different morphologies, particle size distributions, and shape factors were evaluated. The five kaolin (K) clays are No.1 HB ultrafine (K1), special No.2 HB non-delaminated (K2), coarse non-delaminated (K3), regular delaminated (K4), and coarse delaminated (K5). The products were used in coating preparation in their slurry form. The pigment characteristics are given in **Table I**. The study also includes three latex binders, namely, styrene butadiene (SB) latex with a glass transition temperature of ~12°C, acrylic emulsion (acrylic) latex with a glass transition temperature of ~7°C, and styrene acrylate (SA) latex with a glass transition temperature of ~5°C.

The experimental design resulted in a total of 16 coating runs, as shown in **Table II**. The coating formulations were prepared at 54% target solids by adding latex binder and carboxymethylcellulose (CMC) thickener to the pigment slurry. The thickener amount was adjusted to maintain a low shear viscosity of about 300 cP (Brookfield at 20 rpm). The 100% latex coating without clay was applied as received. Coating dispersions were applied onto the back side (rougher surface) of a commercially available solid bleached sulfate (SBS) paperboard (250 gm/m^2 basis weight, PPS roughness of 5.25 µm @10 MPa) at various coat weights (8–16 g/m^2) using a laboratory pilot coater from Dow Modern Metal Craft (Dow; Midland, MI, USA) and wire-wound Mayer rod applicator. All coated substrates were dried using a heated cylinder drum surface (uncoated surface contacting the heated drum) for 1 min unless otherwise specified. The coated sheets were then conditioned at standard TAPPI conditions for about 7–10 days prior to the evaluation. Coat weight (g/m^2) was determined gravimetrically by comparing the grammage of oven dried uncoated and coated substrates tested as per TAPPI Standard Test Method T 410 om-19 "Grammage of paper and paperboard (weight per unit area)." The coated sheets were evaluated for various barrier properties.

Water barrier was measured for 30 min by Cobb test procedure as per TAPPI T 441 "Water absorptiveness of sized (non-bibulous) paper, paperboard, and corrugated fiberboard (Cobb test)." Lower values indicate less water absorptiveness. The oil and grease barrier was measured by Kit test (Kit) procedure following TAPPI T 559 cm-12 "Grease resistance test for paper and paperboard." The Kit test assigns a score rating of 1 to 12 depending on the oil and grease resistance barrier performance, and 12 is the highest ranking. Higher values of Kit test typically indicate better oil and grease barrier.

Water vapor transmission rate was measured by two TAPPI test methods: (1) TAPPI T 448 "Water vapor trans-

Kaolin Clay Type	Particle Size, µm						Shape Factor	Surface Area	Brightness (GE)
	Gravity Method (Sedigraph)							BET (N2)	
	%<2	%<1	%<0.5	%<0.2	Mean	Median	–	m^2/g	%
Ultrafine (K1)	99	97	88	48	0.56	0.22	9	21.0	90.6
Special No. 2 HB									
Non-delaminated (K2)	78	64	47	23	1.73	0.55	22	14.9	90.5
Coarse non-delaminated (K3)	68	56	43	20	2.58	0.71	27	12.6	85.3
Regular delaminated (K4)	86	71	53	23	1.25	0.46	53	15.6	88.2
Coarse delaminated (K5)	65	43	27	12	2.60	1.28	88	13.4	86.4

I. Pigment characteristics.

mission rate of paper and paperboard at 23°C and 50% RH," reflecting standard conditions; and (2) TAPPI T 464 "Water vapor transmission rate of paper and paperboard at high temperature and humidity," reflecting tropical conditions of 90% relative humidity (RH) and 38°C. Lower values indicate less transmission rate of water vapor. The test involves gravimetric determination of the water vapor transmission of the sheet material. A total of 5 sheets were evaluated, and the results were averaged for Kit, while 3 sheets were tested and averaged for WVTR and Cobb tests.

Heat seal tests were performed with Sentinel heat sealer (SencorpWhite; Hyannis, MA, USA) at different temper-

Coating Run No.	Clay Type	Latex Type	Clay/Latex Ratio	PVC, %	CPVC, %
C1	K1	SB	50/50	28.0	59.0
C2	K2				53.3
C3	K3				55.1
C4	K4				48.1
C5	K5				48.3
C6	K5	SB	0/100	0	48.3
C7			20/80	8.8	
C8			35/65	17.3	
C9			50/50	28.0	
C10			65/35	41.9	
C11			70/30	47.5	
C12			85/15	68.7	
C13	K5	Acrylic	0/100	0	48.3
C14			50/50	28.0	
C15		SA	0/100	0	
C16			50/50	28.0	

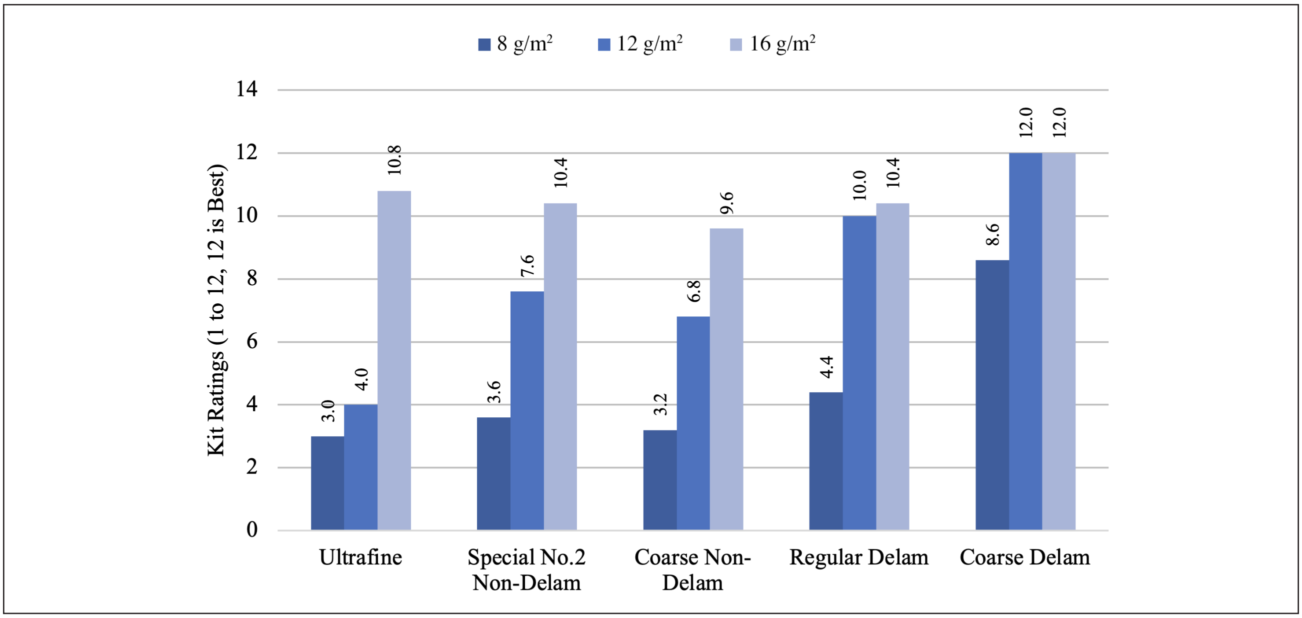
PVC: pigment volume concentration; CPVC: critical pigment volume concentration; SB: styrene butadiene; SA: styrene acrylate.

II. Experimental design showing number of coating runs.

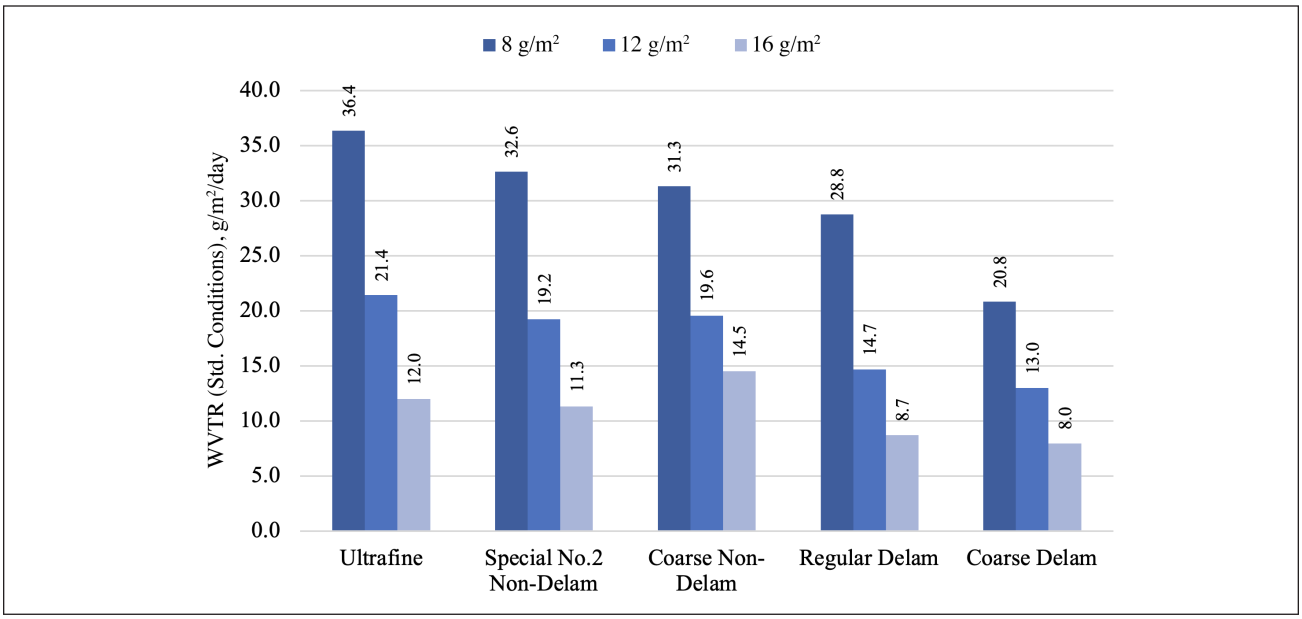
Coating Run No.	Clay Type	Latex Type	Coat Weight	3M Kit Test Rating, 1-12*		WVTR, g/m²/day				Cobb 30 Minutes, g/m²	
						@ 50% RH, 23°C		@ 90% RH, 38°C			
			g/m²	AVG	SD	AVG	SD	AVG	SD	AVG	SD
C1	K1	SB Latex	8.3	3.0	0	36.4	1.2	-	-	22.3	5.3
			12.1	4.0	0	21.4	0.4	327	6.1	11.0	2.1
			16.1	10.8	1.1	12.0	0.1	-	-	10.5	0.9
C2	K2		8.2	3.6	1.3	32.6	3.4	-	-	20.8	3.5
			12.0	7.6	0.9	19.2	0.5	332	13.1	10.5	1.6
			16.3	10.4	1.5	11.3	0.3	-	-	9.5	2.2
C3	K3		8.0	3.2	0.4	31.3	1.7	-	-	20.8	3.7
			12.1	6.8	1.5	19.6	0.5	-	-	12.8	2.8
			16.1	9.6	1.5	14.5	0.3	-	-	10.5	0.6
C4	K4		8.2	4.4	0.5	28.8	3.7	-	-	19.6	3.6
			12.0	10.0	0	14.7	0.2	269	2.8	10.7	0.9
			16.1	10.4	1.1	8.7	0.5	-	-	8.3	2.9
C5	K5		8.0	8.6	1.5	20.8	0.3	-	-	10.9	0.2
			12.0	12.0	0.4	13.0	0.2	236	6.3	7.3	0.2
			16.1	12.0	0.4	8.0	0.4	-	-	6.8	0.5

* 12 is best/highest.
WVTR: water vapor transmission rate; RH: relative humidity; AVG: average; SD: standard deviation; SB: styrene butadiene.

III. Effect of clay type on barrier properties using styrene butadiene (SB) latex binder in 50/50 clay/latex coating systems.



1. Effect of kaolin type on oil and grease resistance (Kit test) at different coat weights.



2. Effect of kaolin type on water vapor transmission rate (WVTR) at different coat weights.

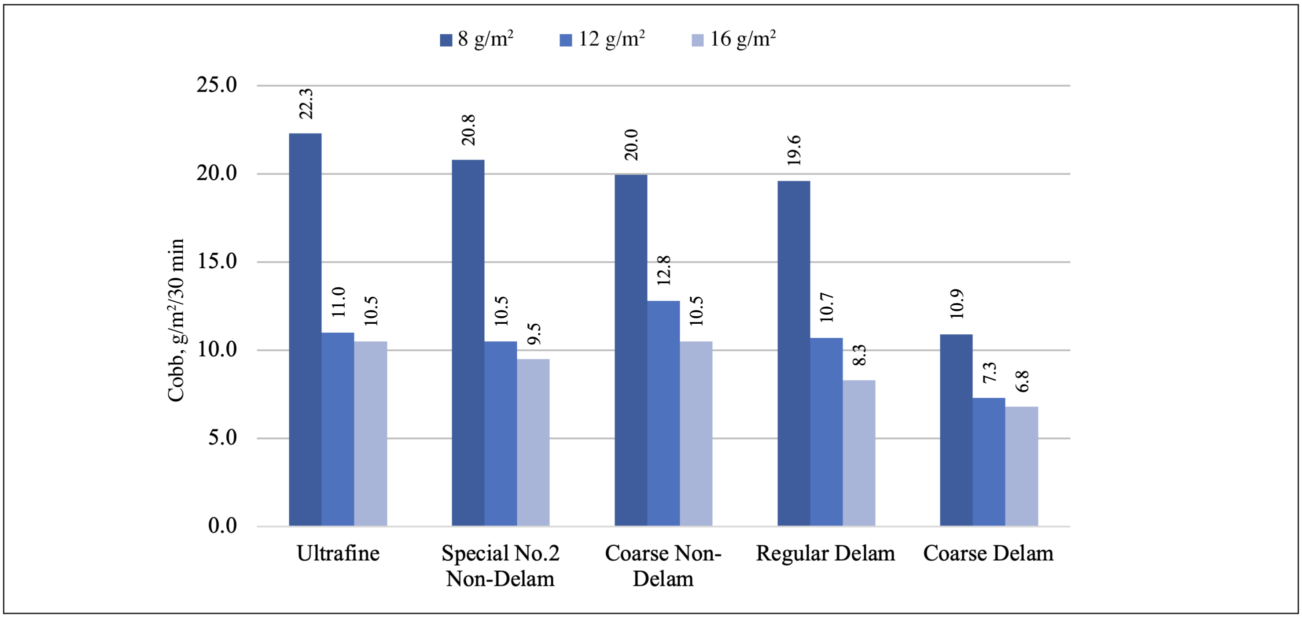
atures (at 414 kPa compression pressure for 5 s) by creating a coated surface-to-uncoated surface interface between its two platens. The interface joint formed is then heated from the back side of the coated sheet, with the other platen at ambient temperature. The bonded joints are then allowed to cool for 15 min before manually separating the bonds and evaluating the sealed area for percent fiber tear. Higher fiber tear values indicate better heat seal.

Blocking tests were performed coated-uncoated side with 896 kPa compression pressure at 65°C for 24 h, then separated after cooling in the standard TAPPI conditions for 30 min. The degree of blocking is expressed as a rating from 1 to 5, with 1 being the best with high blocking resistance. Either 5 (heat seal) or two sheets (blocking) were tested and the results were averaged.

RESULTS AND DISCUSSION

Effect of clay type

The kaolins listed in the materials section were compared to determine the effect of pigment particle size and shape on barrier performance. The shape factor data for different clays range from 9 to 88, as listed in Table I. The scanning electron microscope (SEM) images of ultrafine and coarse delaminated are shown on the cover page of this issue. The kaolin clays were evaluated in a 50/50 clay/latex coating formulation on a dry weight basis with an SB latex



3. Effect of kaolin type Cobb 30 min at different coat weights.

binder at three different coat weights of 8, 12, and 16 g/m² (**Table III**). The 50/50 clay-to-latex ratio has a pigment volume concentration (PVC) of 28.0%, which is lower than the critical pigment volume concentration (CPVC), which is estimated to be 48–59, depending on the clay type (Table II). Their performance was found to be very different in terms of different barrier properties (Cobb, Kit, and WVTR). In this study, the delaminated clays provided better Kit and WVTR results than ultrafine and non-delaminated clays. Such a trend is generally expected, but how the Kit and WVTR improve for the ultrafine and the non-delaminated clays at higher coat weights is intriguing, as demonstrated in **Figs. 1–3**.

The Kit test results achieved with different clays are given in Fig. 1. All clays except the coarse delaminated show inadequate oil and grease barrier quality at the lowest coat weight of 8 g/m² and Kit ratings of only ~3 to 4, whereas the coarse delaminated showed a decent Kit rating of 9. The Kit ratings for the regular delaminated were markedly improved, resulting in a Kit rating of 10 upon increasing coat weight to 12 g/m², while the performance of ultrafine clay was still inferior. The data at the higher coat weight (16 g/m²), on the other hand, paints a very different picture, with all clay products resulting in an acceptable Kit rating close to ~10 or higher.

The WVTR results (50% RH, 23°C) achieved with different clays are given in Fig. 2. The data obtained at the lowest coat weight of 8 g/m² indicate that coarse delaminated performed significantly better than regular delaminated, as well as the rest of the clays, but upon raising the coat weight to 12 and 16 g/m², the performance of regular delaminated improved significantly, with results almost matching that of coarse delaminated. Ultrafine and non-delaminated clays also show improved WVTR at higher coat weights, but to a lesser extent than regular delaminated clay. Nonetheless, the WVTR results improved continuously with coat weight for all clays.

The WVTR results obtained under tropical conditions at 12 g/m² coat weight can be found in Table III. As can be seen, the WVTR values increased with humidity and temperature (90% RH and 38°C) as expected when compared to the results at standard test conditions. However, more importantly, it should be noted that the relative differences in values with respect to clay type are still more or less the same.

The Cobb results as a function of clay type are given in Fig. 3. The coarse delaminated clay performed significantly better than the rest of the clays once again, especially at the lowest coat weight of 8 g/m². Unlike Kit and WVTR results, which improved continuously with an increase of coat weight up to 16 g/m², the improvement in Cobb was minimal after 12 g/m².

Effect of clay-to-latex ratio

The influence of kaolin clay amount (or clay-to-latex ratio) on barrier coating performance was studied. The SB latex and coarse delaminated clay of high shape factor were used for this set of experiments as a function of coat weight at 8, 12, and 16 g/m² (**Table IV**; Run Nos. C6–C12). Additionally, a 100% latex coating without clay at different coat weights was also included as a reference. The coatings containing clay generally showed an improvement over the 100% latex coated samples in WVTR (50% RH, 23°C), Kit ratings, and blocking properties. However, the water barrier (Cobb), WVTR at tropical conditions (90% RH, 38°C), and heat seal properties for the coatings with clay were deteriorated as compared to the 100% latex, as demonstrated in the figures referenced in the following sections.

Coating Run No.	Latex Type	Clay/ Latex Ratio, dry wt%	Coat Weight	3M Kit Test Rating, 1-12*		WVTR, g/m²/day				Cobb 30 Minutes, g/m²	
						@ 50% RH, 23°C		@ 90% RH, 38°C			
			g/m²	AVG	SD	AVG	SD	AVG	SD	AVG	SD
C6	SB Latex	0/100	8.3	1.3	0.6	53.8	2.8	-	-	7.4	2.0
			12.1	9.0	0.8	24.6	0.9	188	5.2	1.6	0.2
			16.1	9.0	-	17.3	0.7	134	5.6	1.5	0.1
C7		20/80	8.2	3.3	0.5	34.4	3.1	-	-	6.2	1.9
			12.0	10.0	-	18.9	0.4	-	-	2.8	0.3
			16.3	12.0	-	12.0	0.1	-	-	2.4	0.5
C8		35/65	8.0	6.0	0.5	28.6	1.0	-	-	7.0	1.2
			12.1	12.0	0.5	15.9	0.7	214	5.6	3.1	0.1
			16.1	12.2	-	11.2	0.3	-	-	3.4	0.2
C9		50/50	8.2	8.6	0.6	26.3	1.8	-	-	16.5	1.2
			12.0	12.0	-	13.0	0.5	235	3.7	7.3	1.5
			16.1	12.0	-	9.2	0.5	-	-	7.8	1.4
C10		65/35	8.0	6.0	0.5	23.1	2.8	-	-	21.3	2.5
			12.0	12.0	-	12.8	0.9	298	7.5	10.6	2.1
			16.1	12.0	-	8.8	0.2	-	-	9.4	0.0
C11		70/30	7.8	6.0	0.5	23.9	1.7	-	-	30.0	2.8
			11.9	12.0	-	11.8	0.4	-	-	13.1	0.4
			15.6	12.0	-	9.0	0.2	-	-	11.0	0.4
C12		85/15	7.9	5.0	1.0	161	16.7	-	-	39.3	3.1
			11.9	8.5	1.5	102	19.2	-	-	28.0	2.0
			16.0	11.0	1.3	84	1.8	-	-	18.5	1.8
C13	Acrylic	0/100	12.2	12	-	113	1.4	873	38.2	2.7	0.8
C14		50/50	12.2	12	-	46.2	1.3	403	28.3	3.2	0.3
C15	SA	0/100	12.0	12	-	159	0.7	1060	35.4	9.6	0.6
C16		50/50	11.9	12	-	70.5	0.7	570	40.3	43	3.8

* 12 is best/highest.

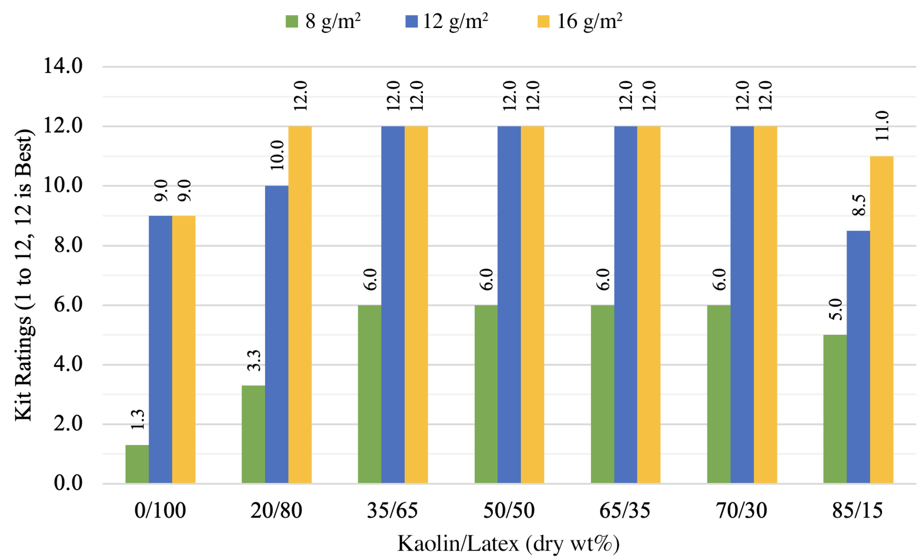
WVTR: water vapor transmission rate; RH: relative humidity; AVG: average; SD: standard deviation; SB: styrene butadiene; SA: styrene acrylate.

IV. Effect of clay/latex ratio on barrier properties for various coating systems containing coarse delaminated clay (K5) and different binders.

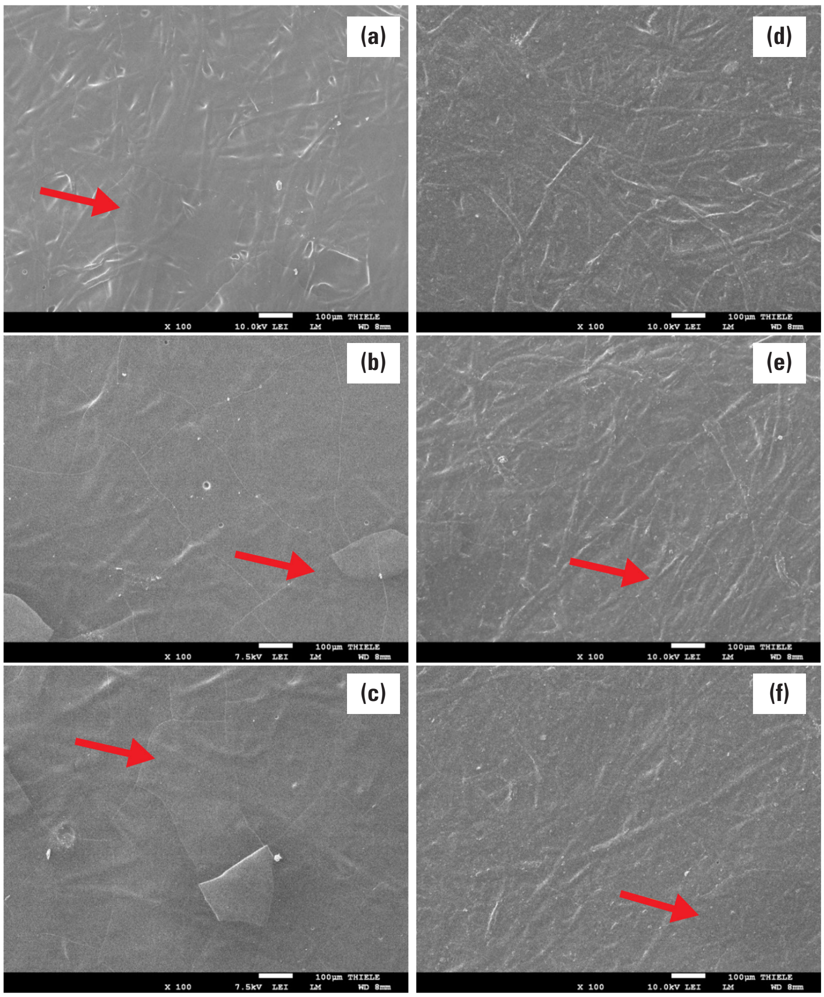
Effect of clay-to-latex ratio on oil and grease barrier

The 100% latex coating without any clay showed very poor Kit at the lowest coat weight of 8 g/m² (**Fig. 4**). It should be noted that we observed some degree of coating dissolution when Kit solutions were deposited on 100% latex coated surface. The poorer results at lower coat weight may be attributed to the observed coating (latex) film dissolution with kit solution, poorer film formation, or both. The Kit values improved with coat weight, yielding a Kit of about 9 at 12 g/m², and no further improvement at 16 g/m². Addition of a minor amount of clay (20/80 clay/latex) resulted in slightly better Kit ratings of 3, 10, and 12, at 8, 12, and 16 g/m², respectively. The coating with 35% clay, on the other hand, achieved a maximum Kit of 12 at 12 g/m², indicating further improvement of oil and grease barrier with 35% clay compared to 20% clay. Increases in clay up to 70% continued to maintain the same level of oil and grease barrier, as noted with 35% clay.

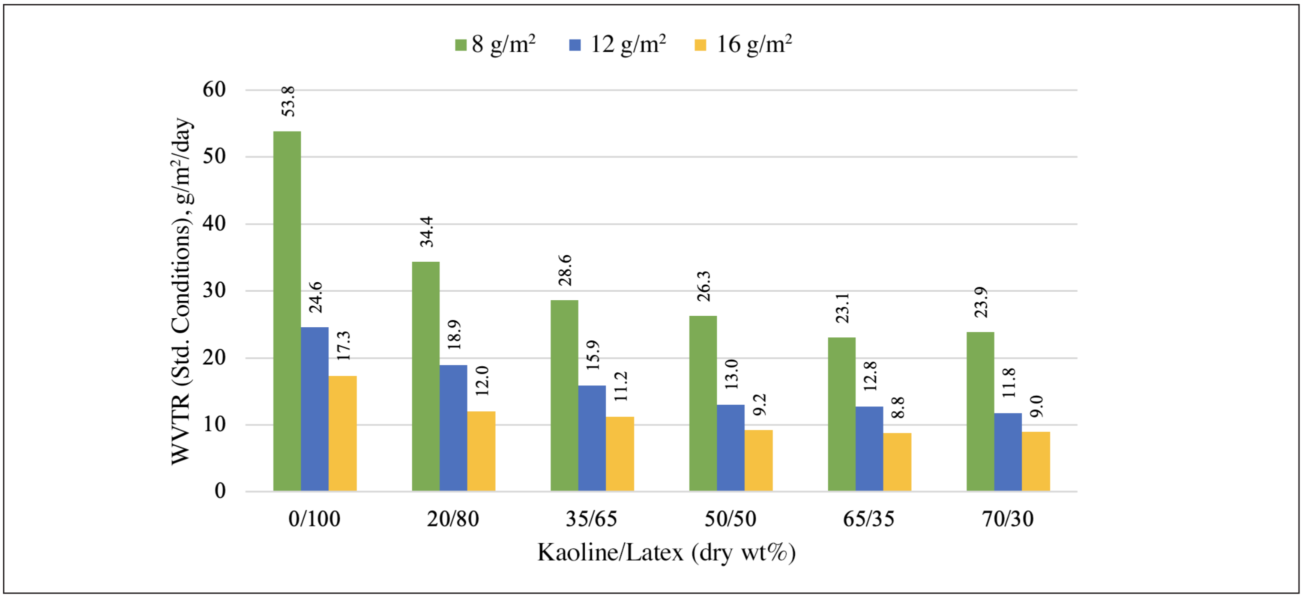
A higher amount of clay in the coating actually means higher coating density and consequently less coating volume per unit weight. So, coatings with more clay should theoretically result in a denser and reduced film thickness due to their lower coating volume. In fact, the SEM images of coated surfaces at equal coat weights corroborate this hypothesis, where the clay/latex (50/50) coated surface shows notably less fiber hiding than 100% latex coating, which is most likely due to its reduced coating volume and film thickness (**Fig. 5**). Even



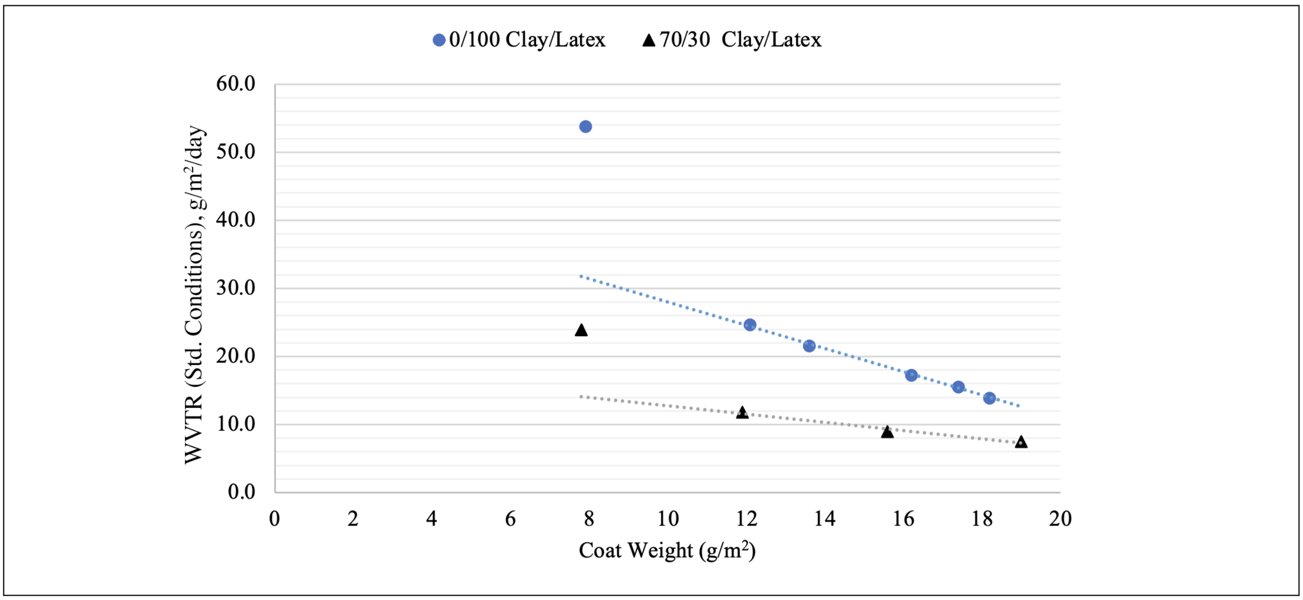
4. Oil and grease barrier vs. different clay loadings at different coat weights.



5. Scanning electron microscopy (SEM) images of coated surface at 100X magnification: (a) 100% SB latex coated at 8 g/m², (b) 12 g/m², and (c) 16 g/m²; and (d) coarse delaminated clay/SB latex (50/50) at 8 g/m², (e) 12 g/m², and (f) 16 g/m². Note: The arrows indicate coating cracks.



6. The WVTR vs. different clay loadings at different coat weights.



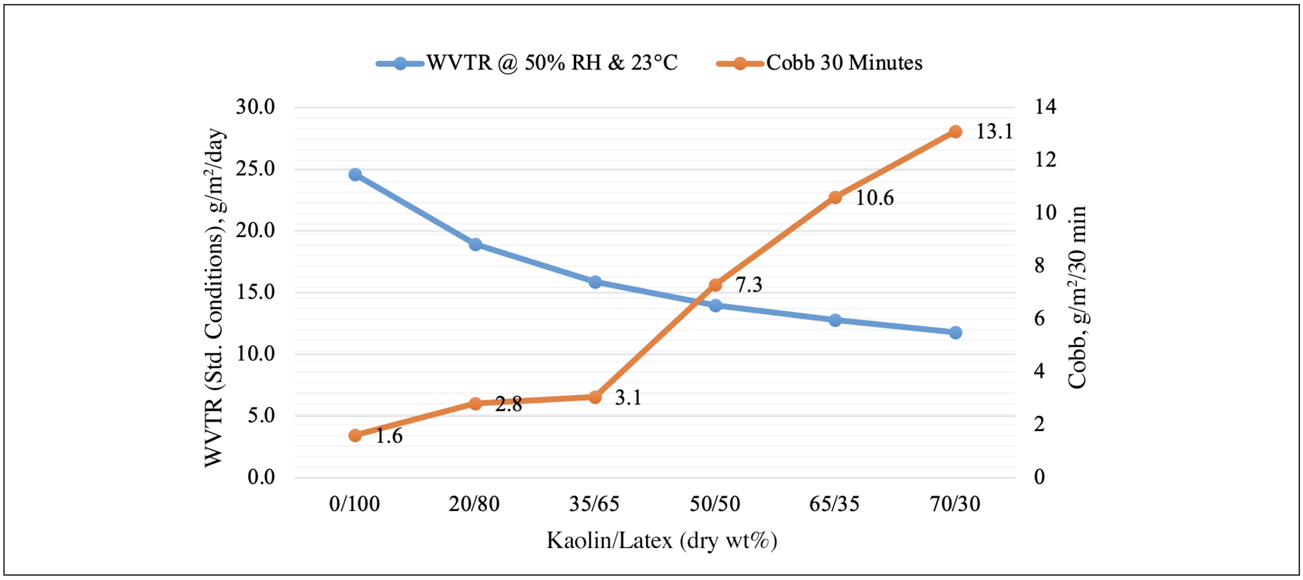
7. The WVTR data trend vs. coat weight.

then, it is interesting to see that the Kit results were maintained at high proportions of clay.

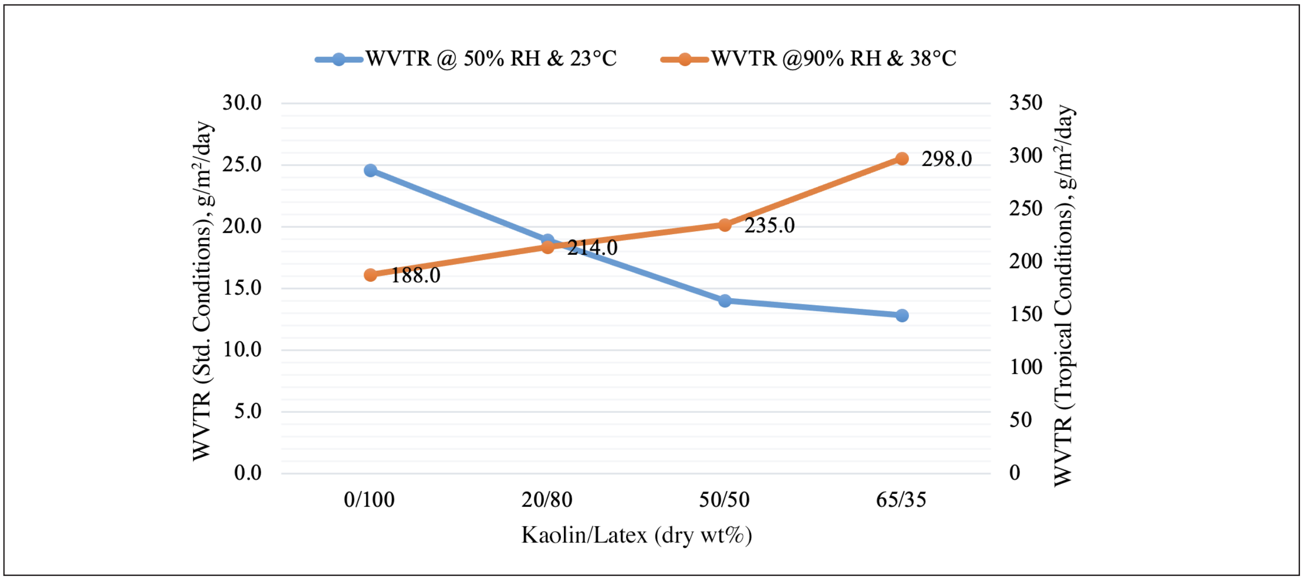
Coated surface scanning electron micrograph images and coverage

The scanning electron micrograph (SEM) images of coated surfaces at different coat weights for clay/latex (50/50) and 100% latex coating systems are shown in Fig. 5. As can be seen, coverage for both coating systems was improved notably with an increase of coat weight. As a result, there is more hiding of fibers at higher coat weights. More interestingly, at a given coat weight, including at 8 g/m², the 100% latex coating showed better fiber hiding than the clay/latex coating, most likely due to its higher coating volume.

As seen in Fig. 5, in addition to some pin holes, microcracks are visible in coating film, and they are present at a higher extent in the 100% latex coating than the clay/latex coating. Also, cracking increased with coat weight for both types of coatings. Cracking of barrier coating films is well documented in the literature [11], but the cracking observed was much larger in width than in the present study. Nevertheless, based on the improved Cobb and WVTR for the 100% latex coating (as discussed further in the following section), the degree of cracking seen in this study does not seem to influence the barrier performance.



8. Comparison of WVTR and Cobb 30 min vs. different clay loadings at 12 g/m².



9. Comparison of WVTR at standard and tropical conditions vs. different clay loadings at 12 g/m².

Effect of clay-to-latex ratio on water vapor transmission rate and water resistance

The WVTR results (50% RH, 23°C) for the coatings at different clay levels are given in **Fig. 6**. The results improved rapidly with coat weight, irrespective of the clay level. The difference in WVTR at 8 vs. 12 g/m² for each coating is much higher than the difference at 12 vs. 16 g/m². A significant improvement in WVTR at 8 g/m² with only a 20% clay compared to the 100% latex coating is worth noting, which can be attributed to the tortuosity created by the clay particles. More importantly, the results gradually improved with an increase in clay level up to 70% (70/30 clay/latex) as compared to the 100% latex coating without clay. The fact that the water vapor barrier results are promising at high proportions of clay (up to 70%) is very important, as it offers more cost savings. The WVTR results were extremely poor at 85% clay (Table IV, Run C12), due to its gain in coating porosity. These results are consistent with earlier work [4], where WVTR improved with clay up to CPVC and then started to deteriorate.

The WVTR data at different coat weights for 100% latex and 70/30 clay/latex coating are presented in **Fig. 7**. As can be seen, the results change linearly at coat weights of 12 g/m² and above, while the results at 8 g/m² do not fit the linear trend. A possible reason for this breakaway from linear relationship could be insufficient coating coverage or insufficient coating thickness at 8 g/m². In any case, it is obvious that a threshold coat weight must be crossed to sufficiently cover the substrate surface so that the coating becomes more effective to achieve the desired properties in any coating application. So, the threshold coat weight in this study may be somewhere between 8 to 12 g/m², depending on the substrate or if it was pre-coated.

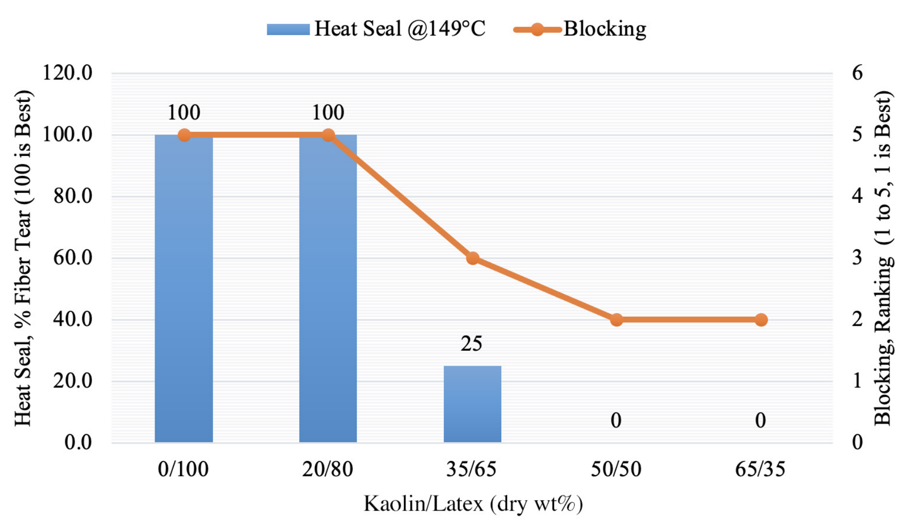
The WVTR (50% RH, 23°C) and Cobb results obtained at 12 g/m² coat weight for the coatings with different clay levels are presented in **Fig. 8**. As can be seen, the 100% latex coating without clay gave the lowest Cobb, while most of the coatings containing clay showed Cobb of ~10 g/m² or less. The Cobb tended to gradually deteriorate with an increase of clay, as opposed to WVTR, which was improved. A possible reason for the deterioration of Cobb may be the disruption of coating film integrity in the presence of liquid water with more clay loading. On the other hand, under relatively mild humidity and temperature (50% RH, 23°C), the tortuosity of coating with more clay may have been able to sufficiently control the water vapor permeation.

The WVTR data obtained at tropical conditions (90% RH, 38°C) at 12 g/m² coat weight for the coatings at different clay levels are presented in **Fig. 9**. The WVTR data at standard conditions (50% RH, 23°C) are also included for side-by-side comparison. As can be seen, the results at the high humidity and high temperature tropical conditions gradually deteriorated with an increase of clay. In essence, the trend observed at tropical conditions was analogous to the trend observed with Cobb results. It is probably be-

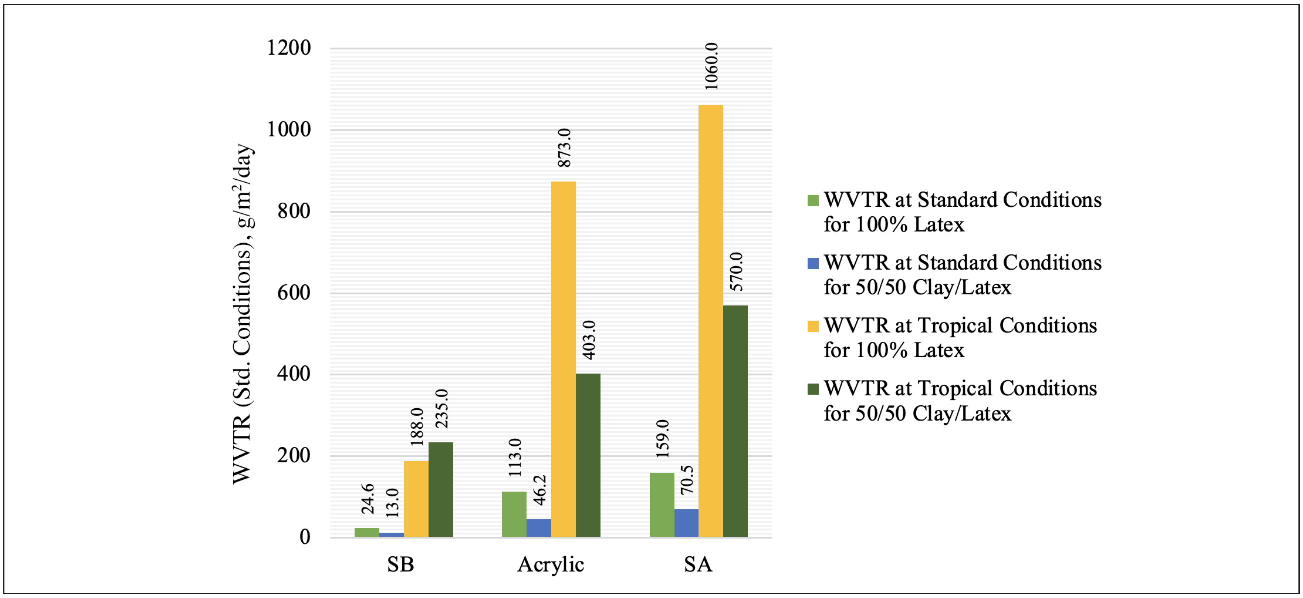
Coating Run No.	Latex Type	Clay/Latex Ratio, dry wt%	Coat Weight, g/m²	Heat Seal at Different Temperatures, % fiber tear*			Blocking Ranking, 1 to 5**
				149°C	177°C	204°C	
C6	SB Latex	0/100	12.1	100	100	100	5.0
C7		20/80	12.0	100	100	100	5.0
C8		35/65	12.1	25	45	86	2.5
C9		50/50	12.0	0	0	20	2.0
C10		65/35	12.0	0	0	15	2.0
C11		70/30	11.9	-	-	-	-
C12		85/15	11.9	-	-	-	-
C13	Acrylic	0/100	12.2	100	100	100	4.0
C14		50/50	12.2	-	60	80	2.0
C15	SA	0/100	12.0	100	100	100	5.0
C16		50/50	11.9	-	35	54	2.5

* Higher is better.
** 1 is best.
SB: styrene butadiene; SA: styrene acrylate.

V. Heat seal and blocking results for various coating systems containing coarse delaminated clay (K5) and different binders.



10. Heat seal and blocking vs. different clay loadings at 12 g/m².



11. The WVTR at standard and tropical conditions for different binder systems with and without clay (SB: styrene butadiene; SA: styrene acrylate).

cause the coatings with higher clay levels were not able to prevent the permeation of water vapor under the harsher tropical conditions that are very similar to the conditions under Cobb testing, where more latex binder seems to play a dominant role in controlling the permeation. The exact reason for the deterioration of WVTR at the tropical conditions is not known, although some loss of coating integrity at the high humidity and temperature conditions is a possibility.

Effect of clay-to-latex ratio on heat seal and blocking properties

The heat seal and blocking data obtained with different clay loadings, as well as the 100% latex coated samples, are given in **Table V** and **Fig. 10**. As can be seen, the 100% latex coating without clay resulted in an exceptional heat seal, resulting in 100% fiber tear at 300°F (148.9°C), indicating that the temperature may be reduced further. Addition of a minor amount of clay (20/80 clay/latex) has maintained the heat seal at the same temperature, but the results deteriorated in a dramatic fashion with any further increase of clay, especially at 50% clay and higher. A higher surface polymer mobility under test conditions is required for a good heat seal [16]. There may be a significant disruption in latex film continuity and surface latex at higher clay loadings, and hence a loss of heat sealability.

Earlier work [16] reported that blocking generally follows an opposite trend to heat seal and improves when there is less work of adhesion upon separation of surfaces in contact [15]. Figure 10 shows the blocking data obtained with different clay levels, as well as the 100% latex coated samples. As can be seen, the 100% latex coating resulted in much poorer blocking, and addition of a minor amount of clay (20/80 clay/latex) did not show any improvement, while there was a dramatic improvement in blocking at 35% clay level. In summary, blocking improved at higher clay levels, whereas heat seal deteriorated. The opposing trends of heat seal and blocking suggests that it would be challenging to maintain both in a single coating system and requires further investigation.

Effect of binder type on the barrier performance of kaolin clay

Three latex binders (SB latex, acrylic, and styrene acrylic) were compared in a constant clay/latex (50/50) ratio formulation at 12 g/m² using coarse delaminated clay (Table IV, Run Nos. C6, C9, C13–C16). The results with 100% latex without clay are also included for each binder system. The main goal was to identify the potential merits of different binders so that the overall coating system performance can be improved. As can be seen from Table IV, all three binders with and without clay performed well and similarly for oil and grease barrier, with Kit values around 12. The water barrier (Cobb) achieved with SB and acrylic binders with and without clay was much improved over the SA latex.

The WVTR results were significantly improved for the clay/latex (50/50) coatings with acrylic, as well as styrene acrylic, binders as compared to their 100% latex counterparts (without clay), and it was true irrespective of the humidity and temperature testing conditions, as shown in **Fig. 11**. On the other hand, as discussed earlier, the WVTR for the clay/latex (50/50) coating with SB latex improved as compared to the 100% latex when evaluated at standard conditions, but it deteriorated at tropical/jungle conditions. So, the clay addition appears to be detrimental to WVTR at high humidity and high temperature in the case of the SB latex binder system, but it was found to be beneficial in the acrylic and styrene acrylic binder-based coatings. Nevertheless, clay improved WVTR at the standard conditions for all binder systems studied. It also improved WVTR at the tropical conditions for acrylic and styrene acrylic, but deteriorated for SB binder. The improved WVTR for most of the systems described previously can be attributed to the coating tortuosity provided by kaolin clay. However, the reason for the deterioration of WVTR for SB latex at the tropical conditions is not known at this time and needs further investigation. As stated earlier, it is possible that integrity of the coating containing kaolin was somewhat compromised under tropical conditions.

The heat seal values (Table V) with each latex without clay was exceptional (100% fiber tear), while the clay/latex coatings with acrylic and styrene acrylic binders resulted in notably better heat seal (data at 177°C–204°C) as compared to the clay/latex coatings with SB latex.

In summary, a change in binder system can significantly alter the barrier properties and the role that a mineral pigment can play. The results indicate that a proper selection of binder systems for each barrier property would be required while considering the clay/latex coating systems. Further research is required to gain a fuller understanding of the mechanism(s) of the effect of binder type on WVTR with and without kaolin.

CONCLUSIONS

Barrier properties of coated samples prepared from coatings containing five different kaolin clays with various particle sizes, morphologies, and shape factors were studied as a function of coat weight. The high shape factor delaminated clays showed better barrier properties than the ultrafine and non-delaminated clays, especially at reduced coat weights. Interestingly, the results also showed that it would be possible to maintain acceptable barrier properties with ultrafine and non-delaminated clays, but this requires significantly higher coat weights.

ABOUT THE AUTHORS

There is an increasing demand for fiber-based packaging with barrier properties to replace single use plastic as part of sustainability efforts in the packaging industry. We conducted research on this topic to develop a comprehensive understanding of the influence of kaolin clays on various barrier properties in latex-based aqueous coatings.

Synthetic latex-based barrier coatings are still a dominant alternative to the traditional barrier coating system, as biopolymer-based barrier coatings are still in the initial stages of development. The use of mineral pigments such as kaolin to substitute part of the synthetic latex binder in aqueous barrier coatings is known to control water vapor transmission and blocking tendency. However, we feel that the information available about kaolin/latex coatings to date is still fragmented and needs continued development.

The drying and curing of coated sheets appear to play a major role in water resistance barrier property. As a result, there was initially significant noise in the results as the curing continued over time. The issue was resolved by subjecting the coated samples to constant levels of drying, as well as curing, prior to the evaluation.

Our work suggests that the barrier properties can be improved using a wide range of kaolin clays, although only the kaolins with high aspect ratio provide superior performance at reduced coat weight. So, proper selection of kaolins is especially important, because coatings that require high coat weights are not going to be cost competitive in the marketplace.

We found that heat seal and blocking properties follow opposing trends and to maintain both within a single coating systems seems to be a challenge.

Devisetti

Lempsink

Malla

Mineral pigments are known to provide blocking resistance. We found that a certain minimum level of kaolin loading was necessary to maintain blocking. However, heat seal property was compromised at the same minimum kaolin loading that was required for blocking. This topic needs further development to enhance the current understanding.

For mills, this work provides insights required to make proper selection of kaolins for barrier coating applications based on different types of latex binders and different target barrier properties.

As part of further research, we aim to further enhance water vapor transmission, water resistance, and heat seal properties of kaolin-containing coatings by incorporating functional layer top coats in the design of coating systems, while still maintaining cost effectiveness. We also plan to develop a coating system that provides acceptable blocking resistance and heat seal together. **TJ**

Devisetti is senior paper research engineer, Lempsink is manager of the Paper Research & Applications Lab, and Malla is director of Research & Development for Thiele Kaolin Company in Sandersville, GA, USA. Email Devisetti at siva.devisetti@thielekaolin.com.

Comparison of coatings at different clay-to-latex ratios as a function of coat weight showed that a significantly high proportion of high shape factor clay can be incorporated to replace SB latex and still achieve exceptional barrier properties. However, heat seal deteriorated at a much lower clay loading than water barrier (Cobb) and water vapor transmission rate (WVTR). Thus, heat seal seems to be the major limiting factor on how much clay can be incorporated.

The WVTR performance with SB latex binder improved with an increase of clay at standard humidity and temperature, but deteriorated at tropical test conditions. On the other hand, for acrylic and styrene acrylate binder coating systems, the results were improved with the addition of clay, irrespective of the testing ambience. The acrylic latex coating with clay showed better Cobb and heat seal than the clay containing coatings with SB latex and styrene acrylate, while the WVTR for the acrylic and styrene acrylate systems was much poorer

Thus, a change in binder system can significantly alter the barrier properties and the role that a mineral pigment can play. Further research is required to gain a fuller understanding of the effect of binder type and different clay-to-latex ratios on various barrier properties, including upon creasing. **TJ**

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