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Although exterior architectural coatings improve the appearance of a structure, they are ultimately intended to be protective, preserving the integrity of a substrate by isolating it from environmental exposure. As the regulatory landscape shifts, however, coating composition is becoming equal in importance to coating performance. Perfluoroalkyl substances (PFAS), defined in a 2022 Organization for Economic Co-operation and Development (OECD) report as "fluorinated substances that contain at least one fully-fluorinated methyl or methylene carbon atom (without any H/Cl/Br/I atom attached to it)," are an effective tool for improving flow, gloss, adhesion, and water- and stain-resistance of coatings, but their use has a negative impact on both human health and the environment. Eliminating PFAS, while maintaining coating performance, requires unique binder chemistry that is both flexible and durable. An all-acrylic waterborne latex for exterior architectural applications has been engineered without PFAS. This new, low-coalescent demand binder has improved surfactant leach resistance when compared to a similar PFAS-containing product, while maintaining other performance properties, such as block resistance and dirt pick-up resistance. Through creative polymer design, a more ecologically sound waterborne all-acrylic latex with improved exterior performance is possible.

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Improved Performance in a Waterborne All-Acrylic Latex Produced without PFAS

Introduction

Although liquid suspensions of pigment were first used more than 30,000 years ago in a decorative capacity, the utility of paint as protection was not fully realized until the dawn of the Industrial Revolution, when the first paint and varnish factories began mass-production of ready-made coatings.[1] While the automotive industry introduced the need for anti-corrosive coatings, special-purpose coatings were eventually developed for other products and industries, including farm equipment, children’s toys, furniture, and food production.[2] Historically, in addition to innovation, the coatings industry has recognized and responded to potential negative impacts of product chemistries on human health and the surrounding environment. For example, during the years leading up to World War II, as consumers began to fully comprehend the inherent health and environmental risks associated with a common paint ingredient, lead, paint manufacturers ultimately found a safer alternative, titanium dioxide. The use of lead pigments in consumer paints was limited in the United States by the 1950s and fully banned in 1978.[3]

As academic institutions, governmental agencies, corporations, and consumer groups began to acknowledge the extent of coatings’ impact on the environment in the early part of the 21st century, demand increased for eco-friendly solutions that both comply with environmental regulations and maintain product performance. Coating manufacturers first met these calls for change by either reducing or eliminating volatile organic compounds (VOCs) in solvent-based and water-based coating formulations, but recent regulatory developments and industry trends have led to more aggressive measures to reduce industrial environmental footprints.[4,5] To continue to expand this $26.1 billion industry, development of new, high-performance coatings and a commitment to sustainable chemistry and manufacturing practices is now paramount.[6]

In February 2021, the U.S. Environmental Protection Agency (EPA) began developing the Fifth Unregulated Contaminant Monitoring Rule (UCMR 5) to provide new data on 29 perfluoroalkyl substances (PFAS) and to clarify their impact on community drinking water. Since then, the EPA Council of PFAS has been created to better understand, and ultimately reduce, the potential risks of PFAS. EPA has also released preliminary Toxic Release Inventory (TRI) data, enhanced TRI reporting requirements, and begun development of a national testing strategy for PFAS. Additionally, in June 2022, EPA released four drinking water health advisories for PFAS; as a direct result, $1 billion in Infrastructure Law grant funding has become available to address PFAS contaminants in drinking water. More recently, in January 2023, EPA proposed a rule that would prevent starting or resuming the manufacture, processing, or use of an estimated 300 PFAS that have not been made or used for many years, known as "inactive PFAS," without a complete EPA review and risk determination.[7]

Earlier generations of acrylic latexes were created with the intention of meeting the consumer expectation that a fully formulated coating would contain no more than 50 g/L volatile organic content (VOC). Reduced VOC targets were readily met by latexes having lower glass transition (T_g) temperatures and minimum film-forming temperatures (MFFT), but their use resulted in softer films. Softer films were deficient in several performance areas, including surface feel, dirt pick-up resistance, block resistance, and stain removal. Surfactant leaching was also observed. Surfactant leaching is defined as the unsightly staining caused by coating components migrating through the film and streaking down the coated surface. This defect is unique to lower-VOC exterior coatings, particularly when a deep tint coating is applied in low-temperature and high-humidity environments.

Improved surfactant leach resistance was identified as a product improvement target. Built on an exterior acrylic platform recognized for its durability and color acceptance, a new polymer was designed specifically to resist surfactant leaching. While surfactant leach resistance was successfully achieved through polymer design, certain performance properties were compromised. Block resistance, in particular, was found to be deficient. Performance improvement was realized, in part, through the incorporation of a specific class of PFAS, fluoropolymers, into the polymer recipe. Fluoropolymers have non-wetting and non-sticking properties, and are a well-established way to introduce block resistance.[8] The release of the *PFAS Strategic Roadmap* by EPA in October 2021, however, indicated a dramatic shift in the regulatory landscape.[9] In response, an improved exterior all-acrylic latex produced was developed to exceed the performance of its PFAS-containing predecessor, without the incorporation of PFAS.

Background

PFAS are widely used synthetic substances. Since the discovery of polytetrafluoroethylene (PTFE) in 1938, PFAS have been used extensively in a myriad of applications, including paints, non-stick cookware, fast food wrappers and packaging, pizza boxes, firefighting foam, nail polish, and dental floss. This is due to their resistance to heat and chemical agents, and their anti-weathering, anti-UV (ultraviolet) fading, and surface modification properties.[10] In coatings, paints, and varnishes, fluorinated substances have traditionally been added to provide resistance to corrosion, weathering, blocking, abrasion and scratching, UV exposure, and to improve substrate wetting and overall durability. The highly stable carbon-fluorine bond and the unique physicochemical properties of PFAS make these substances valuable ingredients for products that require strength, resilience, and longevity. Since 2002, global manufacturers have been replacing "long-chain (LC)" PFAS, their salts, and their potential precursors, with chemicals containing shorter perfluoroalkyl chains or with non-perfluoroalkyl products.[11] This trend has been driven by concerns related to the impact certain LC PFAS may have on health and on the environment. Current peer-reviewed scientific studies have shown that exposure to certain type and concentrations of PFAS may lead to adverse human health outcomes,[12] including:

- decreased fertility in reproductive-age women or increased high blood pressure in pregnant women;
- developmental effects in children, including low birth weight, accelerated puberty, bone variations, or behavioral changes;
- increased risk of some cancers, including prostate, kidney, and testicular cancers;
- reduced ability of the body's immune system to fight infections, including reduced vaccine response;
- interference with the body's natural hormones; and
- increased cholesterol levels and/or risk of obesity.

The most recent definition of PFAS, published in a 2022 Organization for Economic Co-operation and Development (OECD) report, is "fluorinated substances that contain at least one fully-fluorinated methyl or methylene carbon atom (without any attached H/Cl/Br/I atoms)."[13] This broad definition, which is under consideration by EU member states and U.S. federal, state, and city governments, is intended to restrict PFAS with high persistence, high mobility, and tendency to bioaccumulate in the environment.[14]

In response to changing market dynamics, a new all-acrylic waterborne latex produced without PFAS has been developed and tested as a possible high-performance alternative to more conventional all-acrylic latexes. This new binder was built on a successful high-solids, cost-effective acrylic platform, which delivered a low-VOC capable, exterior acrylic with superior surfactant leach resistance. In addition to this functionally important and aesthetically desirable property, the new exterior acrylic was also designed with improved block resistance and dirt pick-up resistance.

Experimental Procedures

Paint Preparation: A water-based exterior matte deep base paint (volume solids = 36.4%; Pigment Volume Concentration (PVC) = 33.3%) and a water-based exterior semigloss white base paint (volume solids = 37.1%; Pigment Volume Concentration (PVC) = 26.3%) were prepared using standard laboratory techniques. Refer to **Table 1** and **Table 2** for details. The exterior matte deep base paints were tinted with COLORTREND® 888 universal machine colorants (Chromaflo Technologies) in either red, blue, or green.

Paint Viscosity: Krebs Unit (KU) viscosity was measured with a Brookfield KU-1 Viscometer, using the method described by ASTM D562.[15] High-shear (ICI) viscosity was measured with a Brookfield ICI Viscometer CAP2000+ (Brookfield AMETECH) using the method described by ASTM D4287.[16]

Paint pH: The pH of all paints was measured with an Oakton series 11, 100, or 110 pH Meter and Oakton #35811-71 or 35811-72 electrode (Fisher Scientific), using the method described by ASTM D5324.[17]

Paint Density: The density (in pounds per gallon) of all paints was quantified, using the method described by ASTM D1475.[18]

Low Temperature Coalescence (LTC) of Paint Films: The film formation of all paints (10-mil wet films at 40°F) was determined, using the method described by ASTM D7306.[19]

Contrast Ratio/Y% Reflectance of Paint Films: Contrast ratio and Y% reflectance

TABLE 1
Exterior Matte Deep Base Paint Formula Utilized for Surfactant Leach Resistance Studies

Ingredient	Weight (lbs)	Volume (gallons)
Grind Base		
Water	220.0	26.4
NATROSOL™ 330 PLUS	4.0	0.3
BYK® 019	3.0	0.4
COADIS™ BR-40	6.0	0.7
Carbowet® 109	2.0	0.2
ATTAGEL® 50	2.0	0.1
MINEX® 3	239.1	11.0
Ammonium hydroxide (28% aq.)	2.5	0.3
ACTICIDE® BW-20	1.1	0.1
BYK® 025	2.0	0.2
Thindown		
50% solids acrylic	392.8	44.6
TEXANOL™ Ester Alcohol	10.4	1.3
COAPUR™ 2020W	6.0	0.7
COAPUR™ 817W	1.0	0.1
Water	28.5	2.4
TOTAL	920.4	88.8
Property	**Value**	
Wt. solids (%)	47.9	
Vol. solids (%)	36.4	
PVC (%)	33.3	
VOC (g/L)	<50	

TABLE 2
Exterior Semigloss Paint Formula Utilized for Other Paint Performance Studies

Ingredient	Weight (lbs)	Volume (gallons)
Grind Base		
Water	85.0	10.2
NATROSOL™ 330 PLUS	1.0	0.1
Ti-Pure™ R-746 (slurry)	325.0	16.8
BYK® 024	2.0	0.2
COADIS™ 123K	8.0	0.9
Ammonium hydroxide (28% aq.)	1.5	0.2
BYK® 349	2.5	0.3
MINEX® 7	10.0	0.5
COAPUR™ 2020W	25.0	2.9
Thindown		
DREWPLUS™ T-4507	2.0	0.3
ACTICIDE® MBS	2.0	0.2
POLYPHASE® 663	18.0	2.0
COAPUR™ 817W	6.0	0.6
50% solids acrylic	525.0	59.6
TEXANOL™ Ester Alcohol	15.6	1.8
LOXANOL® CA 5310	4.3	0.6
Water	30.6	3.7
TOTAL	1063.5	100.9
Property	**Value**	
Wt. solids (%)	50.1	
Vol. solids (%)	37.1	
PVC (%)	26.3	
VOC (g/L)	<50	

for all paint films was measured with a Colorimeter (BYK-Gardner), using the method described by ASTM D2805.[20]

Gloss of Paint Films: The specular gloss of all paint films for glossmeter geometries of 20°, 60°, and 85° was measured with a micro-Tri-gloss Meter (BYK-Gardner), using the method described by ASTM D523.[21]

Paint Tint Strength and Rub-Up Resistance: Five grams of Phthalo blue colorant were weighed into the bottom of a half-pint can, after which 250 grams of paint base were added. The colorant was then thoroughly dispersed by shaking the half-pint can for at least 5 minutes on a paint shaker (Red Devil). Once the colorant had been fully incorporated, a 6-mil Bird bar drawdown was applied to a 1B Leneta chart. To assess color development, the drawdown was allowed to dry for 1 minute, and then the paint was manipulated using light finger pressure; 20 cycles of rub-ups were completed on both the sealed and unsealed sections of the chart. The 1B Leneta chart was then allowed to further dry at room temperature and humidity for 24 hours. After the 24-hour dry time, the Y% brightness values for the sealed and unsealed sections of the drawdown and the rub-up areas were read, using a Colorimeter (BYK-Gardner). The percent tint strength was then calculated by the Kubelka-Munk formula:

$$\%TS_{KM} = \frac{[1\text{-}Y\%_{BAT})^2/(2Y\%_{BAT})]}{[1\text{-}Y\%_{Control})^2/(2Y\%_{Control})]}$$

where $Y\%_{BAT}$ & $Y\%_{Control}$ are brightness values of batch and control, in fractional form [$0< Y\%_{BAT} <1$], and where $\%TS_{KM}$ is Tint Strength of Batch where control is equal to 100%. Typically, the deviation from 100% indicates that TiO_2 should either be added [if >100%] or removed [if<100%] to match the color of the control paint.

Block Resistance of Paint Films: Paint films (3-mil wet) were dried under controlled environmental conditions (77 °F/50% relative humidity) for 1, 3, and 7 days. Films for each dry time were cut into 1.5” x 1.5” squares. Two squares, film sides touching, were compressed with a 1000 g weight under controlled environmental conditions (either 77 °F/50% relative humidity for 24 hours or 120 °F for 30 minutes). After the allotted time, the strips were separated and rated as described by ASTM D4946.[22]

Dirt Pick-Up Resistance Evaluation

A 6-mil wet film of each coating was applied to a treated 3" x 8" aluminum panel and allowed to cure at least 12 hours at ambient temperature and humidity. Coated panels then were exposed to QUV-A radiation for 100 hours. Iron oxide slurry (125 grams of BAYFERROX® 600 pigment (Lanxess) in 250 grams of water, dispersed to a fineness of grind of 5) was brushed onto the lower half of each panel, allowed to dry, and then removed with a wet sponge (1 ¼" x 1 ¼" x 3 ¼" sponge, measured dry) using a scrub machine (BYK-Gardner) for 30 cycles. This procedure was repeated for a total of three cycles. Y reflectance values were taken initially and after each of the three cycles, using a colorimeter (BYK-Gardner). The %Y change was reported, using the following equation:

%Y Change = 100 * $(Y_0 - Y_F) / Y_0$

where

Y_0 = Initial Y reflectance value

Y_F = Final Y reflectance value

Surfactant Leach Resistance Evaluation ("water droplet" method)

Using a method derived from ASTM D7190,[23] a 3-mil (dry thickness) coating was applied to a Leneta 3B chart or Leneta Scrub panel using a 6-mil Bird bar and allowed to dry for either 4 hours, 1 day, or 3 days. At each time interval, three pools of water, consisting of three drops each, were applied to the film. The three water pools were allowed to remain on the film for 10 minutes. After 10 minutes, the panel was tipped vertically, allowing the water pools to run down the surface of the film. Samples were then allowed to fully dry in a vertical position. When dry, each section was rated on a scale of 0 to 5 (where 0 = severe surfactant leaching, and 5 = no sign of surfactant leaching).

Surfactant Leach Resistance Evaluation ("water extraction" method)

Leneta Scrub panels were cut into 4" x 11.25" rectangles and weighed prior to paint application. Panels were then coated, using a 4-inch, 20-mil gap square bar, and allowed to dry at ambient temperature and humidity for 3 days. Dried, painted panels were weighed, and then the painted surface of the panel was completely submerged in water. After a 1-hour soak time, panels were removed from the water and hung to dry. Once the panel had dried completely, it was weighed again. Extracted leachate was calculated using the following equation:

Extracted Leachate = [Painted Panel (dry, before extraction) – Painted Panel (dry, after extraction) / Painted Panel (dry, before extraction) – Uncoated Panel]

Results and Discussion

Surfactant leach resistance in matte deep base

The surfactant leach resistance of the acrylic latexes was assessed in lab-generated exterior matte deep base formulation, using two different methods, as shown in **Table 3**.

TABLE 3
Acrylic Latexes Evaluated in Exterior Matte and Semigloss Paint Studies

Description	Total Solids (%)	MFFT (°C)	PFAS Content
Conventional acrylic	57	16	No
PFAS-containing acrylic	52	16	Yes
Acrylic produced without PFAS	49	10	No

The "water droplet" surfactant leaching method is a visual assessment of paint film appearance, after the film has been streaked with water and allowed to dry. The "water extraction" surfactant leaching method quantifies materials that migrate through the paint film during an extended water soak. In both methods, test coatings were tinted with 12 ounces of either red, blue, or green colorant. Tinted coatings were evaluated using both the "water droplet" and the "water extraction" method. The untinted white coating was also included as a candidate for assessment in the "water extraction" test.

A low-VOC matte deep base formulation was used exclusively to evaluate surfactant leaching resistance. This paint was highly filled and designed to be tinted with 12 ounces of colorant. While ideal for differentiating between the surfactant leach resistance capabilities of acrylic latexes, this formulation was not well suited for other kinds of testing. As a result, a semigloss white base was also developed to assess other performance properties.

While both methods examine aspects of leaching, results do not necessarily correlate: not all compounds that migrate through a paint film leave behind visible residue. The "water droplet" method, however, allows for a quick assessment to determine the extent of discoloration caused by leaching. The "water extraction" method provides a way to quantify the ability of a film to trap substances and limit their migration to the surface.

Film appearance of the standard acrylic, the PFAS-containing exterior acrylic, and the new exterior acrylic produced without PFAS in tinted matte paints was evaluated using the "water droplet" method after 4 hours, 24 hours, and 5 days of drying time. A rating of zero indicated heavy accumulation of leached residue, easily visible once the surface of the paint film had completely dried, and a rating of five indicated that the film was devoid of any discoloration or change in appearance (i.e., shine or streaks). Refer to **Figures 1, 2,** and **3** for results. Refer to **Figure 4** for a photograph of panels with high and low ratings.

Although all three colorants contained species that migrated readily through the paint film, in most instances, the blue colorant appeared to be the colorant most likely to leave visible residue on the film surface. Film appearance for paints formulated with any of the three acrylic emulsions improved with increasing dry times. The film of the paint formulated with the acrylic containing PFAS almost uniformly accumulated more residue than either of the other two paint films. Film appearance of the PFAS-containing acrylic paint may be more closely linked to the presence of PFAS-containing surfactant than to the ability of the latex binder to inhibit material migration. Detergent-like surfactants, such as sodium lauryl sulfate (SLS), that can migrate through a film, may effectively wet a surface, allowing leachate to spread rather than streak. A highly hydrophobic perfluorinated surfactant, though, may encourage a longer dwell time of a water droplet containing concentrated species, which ultimately can stain the paint surface. As dry times increased, less residual surfactant appeared to accumulate on the surface of the new acrylic produced without PFAS films, regardless of colorant.

Species migrating through tinted and untinted paint films formulated with the conventional acrylic, the PFAS-containing acrylic, and the new acrylic produced without PFAS were quantified after a one-hour, ambient temperature water soak. The surfactant leaching resistance of the conventional acrylic and the new acrylic

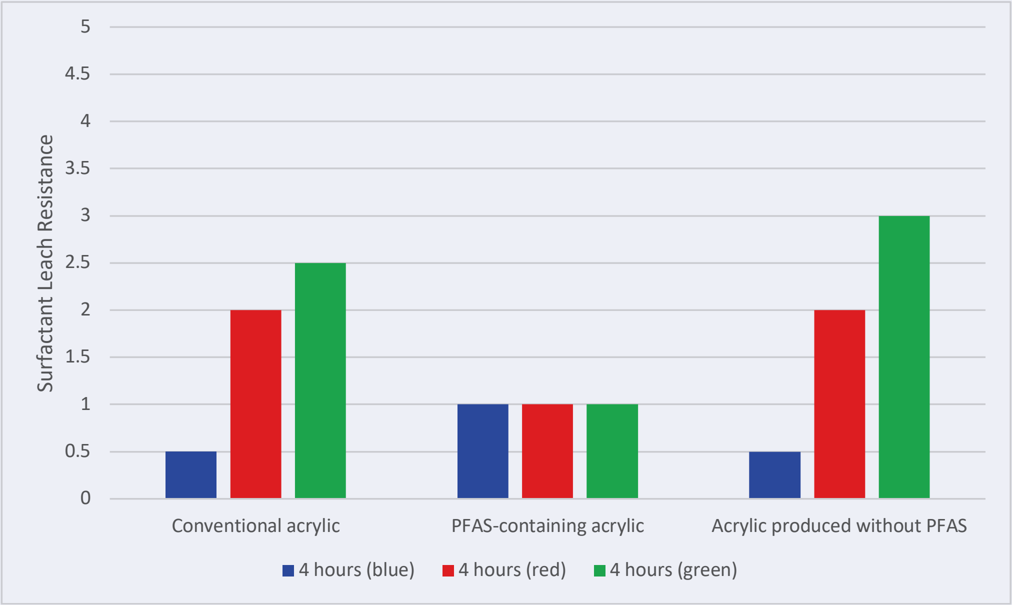


FIGURE 1
Results from surfactant leaching resistance "water droplet" test in tinted matte paints formulated with conventional and developmental acrylics after 4 hours of dry time.

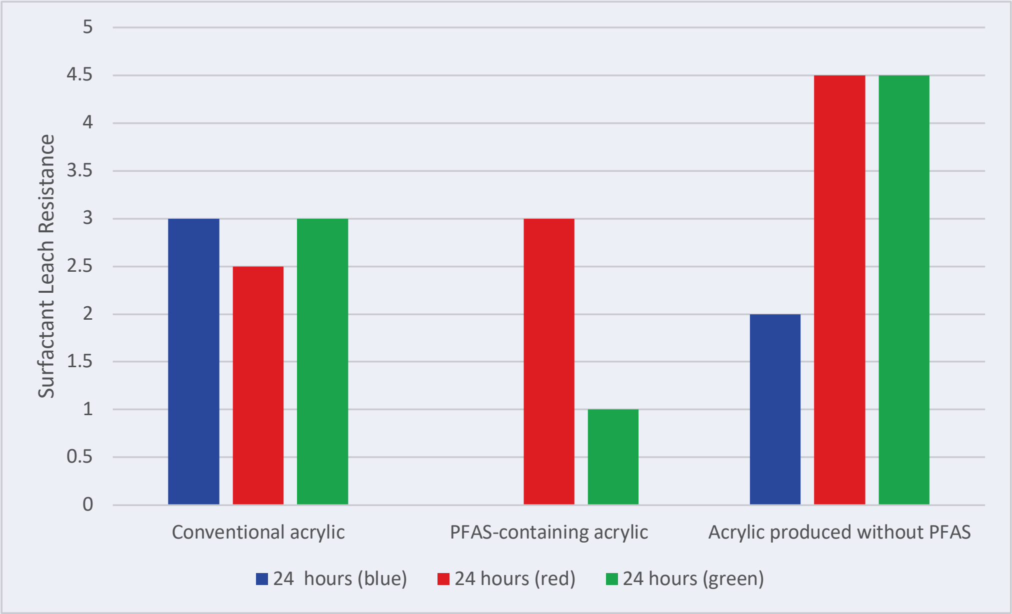


FIGURE 2
Results from surfactant leaching resistance "water droplet" test in tinted matte paints formulated with conventional and developmental acrylics after 24 hours of dry time.

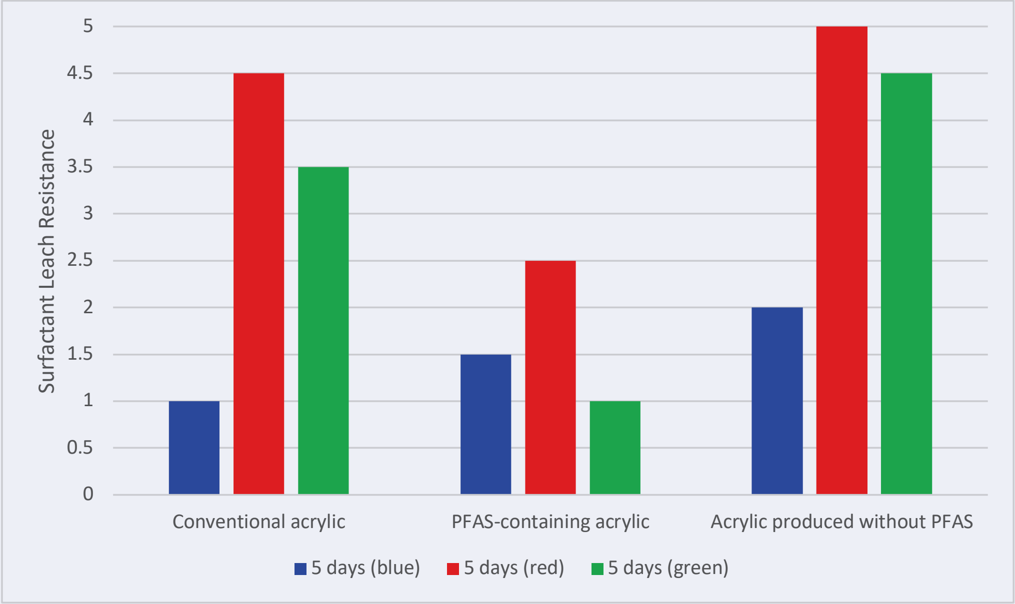


FIGURE 3
Results from surfactant leaching resistance "water droplet" test in tinted matte paints formulated with conventional and developmental acrylics after 5 days of dry time.

FIGURE 4
Photograph of green-tinted matte deep base coatings, after the completion of the "water droplet" test. Left panel: rating of 5 (no surfactant leaching). Center panel: rating of 3 (moderate surfactant leaching). Right panel: rating of 1 (excessive surfactant leaching).

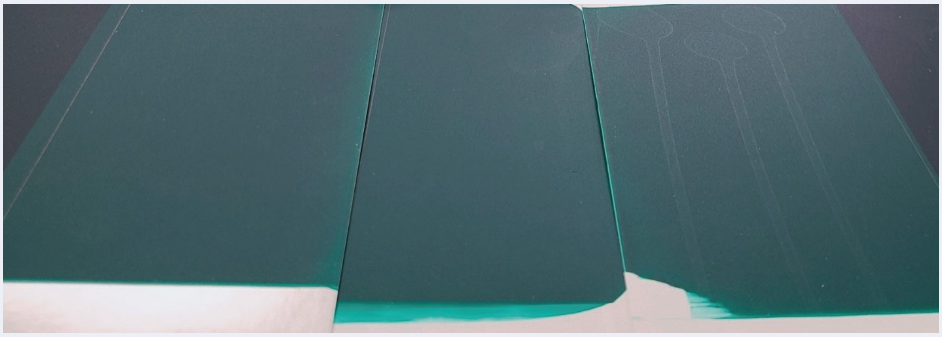


FIGURE 5
Results from surfactant leaching resistance "water extraction" test in untinted and tinted matte paints formulated with conventional and developmental acrylics after a one-hour soak time. Performance compared to PFAS-containing acrylic; a lower number indicates better performance.

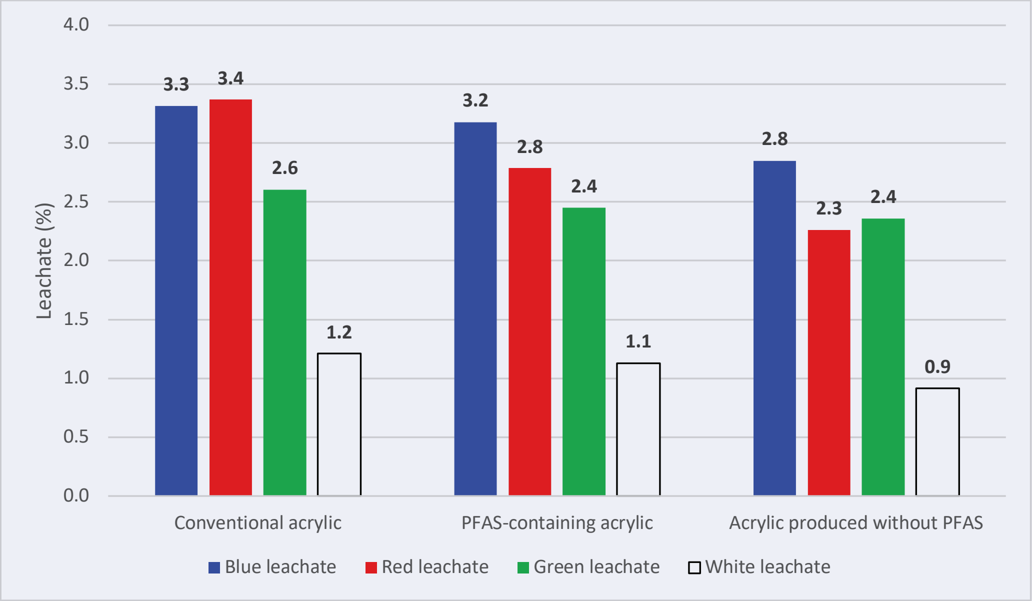
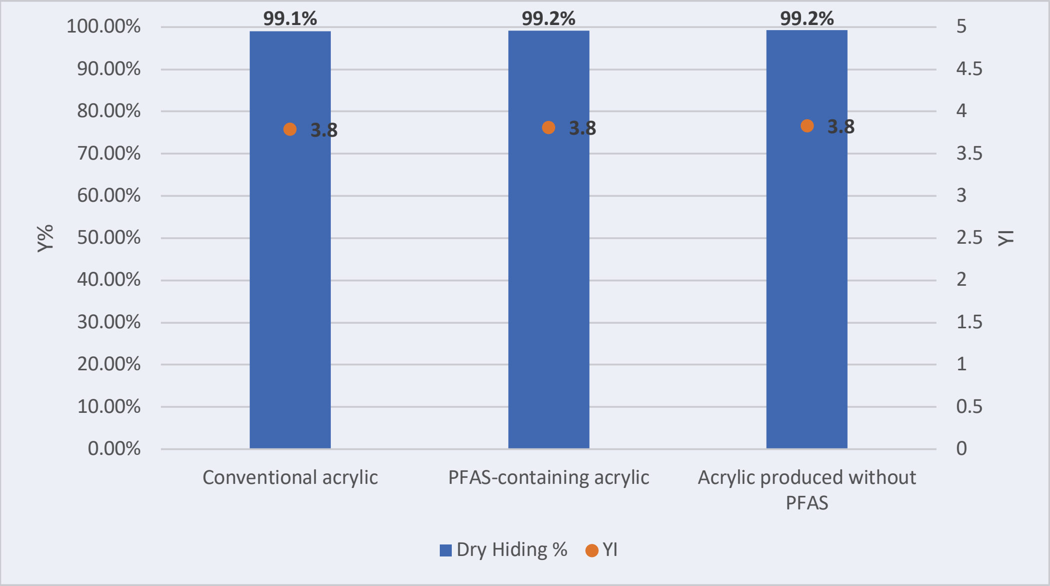


FIGURE 6
Dry hiding and yellowness of conventional and developmental acrylics in an exterior semigloss formulation.



produced without PFAS was compared to the performance of the PFAS-containing acrylic. Refer to **Figure 5**.

The new acrylic produced without PFAS showed improved surfactant leaching resistance in both untinted and tinted matte paints, when compared to the performance of conventional and PFAS-containing acrylics.

Performance in exterior semigloss white base

To further assess and compare the performance of the acrylic binders, all three products were formulated into an exterior semigloss paint and evaluated for optical properties, gloss development, color acceptance, block resistance, and dirt pick-up resistance.

Dry hiding, yellowness, and gloss development of the conventional acrylic, the PFAS-containing acrylic, and the new acrylic produced without PFAS were similar in the exterior semigloss formulation. All three semigloss formulas exhibited low yellowness and high hiding power. While gloss development for the three formulas tracked closely, 20-, 60-, and 85-degree gloss values for the PFAS-containing acrylic were slightly lower than the gloss values of the other two formulas. Gloss values for the conventional acrylic and the new acrylic produced without PFAS, however, were closely aligned. Refer to **Figures 6 and 7**.

Color acceptance testing, as evidenced in **Figure 8**, is often comparative, with a standard tinted paint used as a control. In this study, tint strength and "rub-up" resistance of the conventional acrylic and the new acrylic produced without PFAS were compared with values measured for the PFAS-containing acrylic (control paint). The paint formulated with the new acrylic produced without PFAS had a tint strength close to that of the control paint. The conventional acrylic had slightly lower tint strength than that of the control. Color stability for all three acrylic formulations was within expected test parameters. For tint strength comparison, refer to **Figure 8** for details.

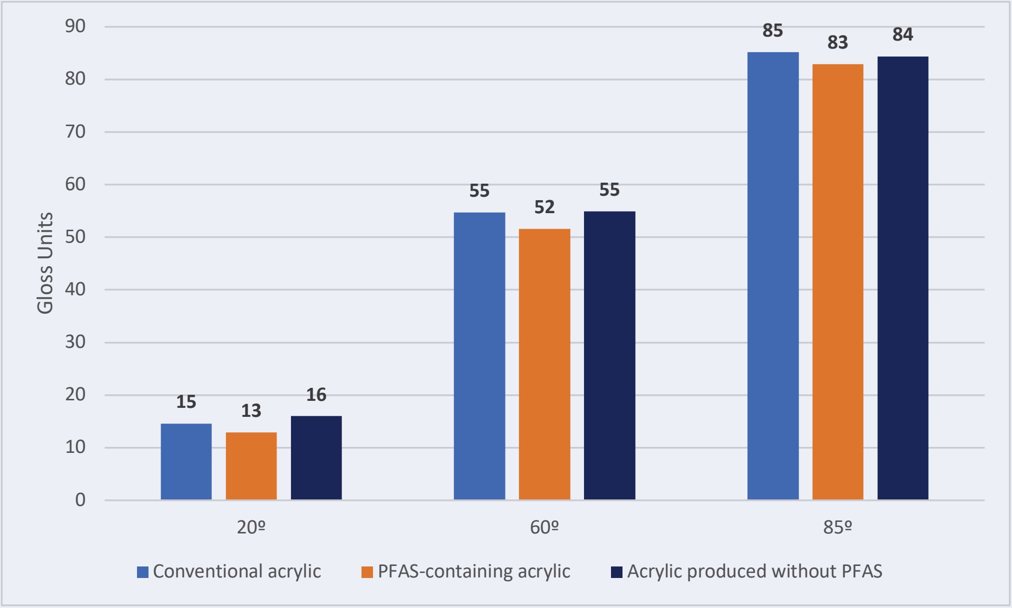


FIGURE 7
Gloss development of conventional and developmental acrylics in an exterior semigloss formulation.

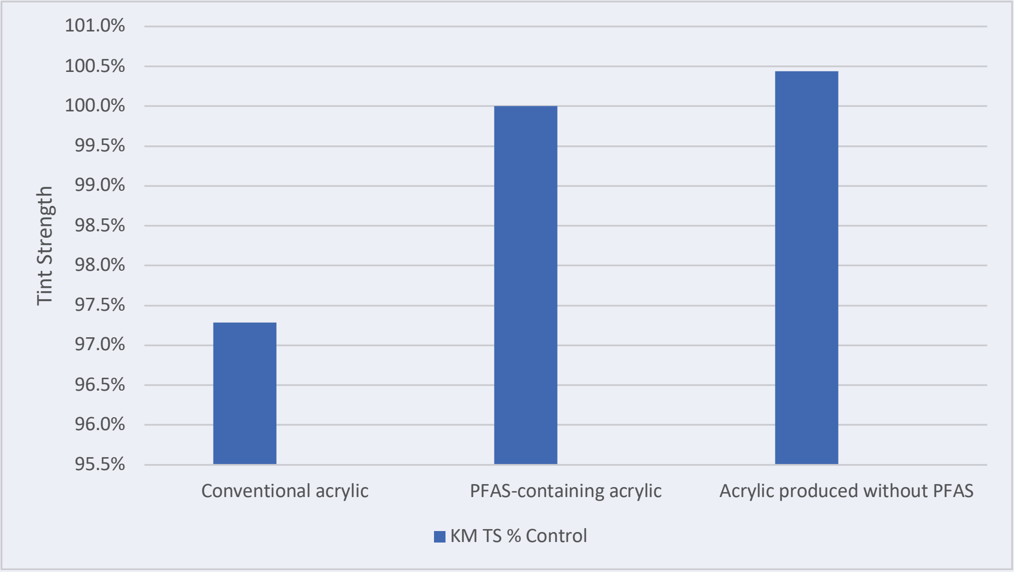


FIGURE 8
Color acceptance (tint strength) of conventional and developmental acrylics in an exterior semigloss formulation.

FIGURE 9
Ambient temperature block resistance of conventional and developmental acrylics in an exterior semigloss formulation.

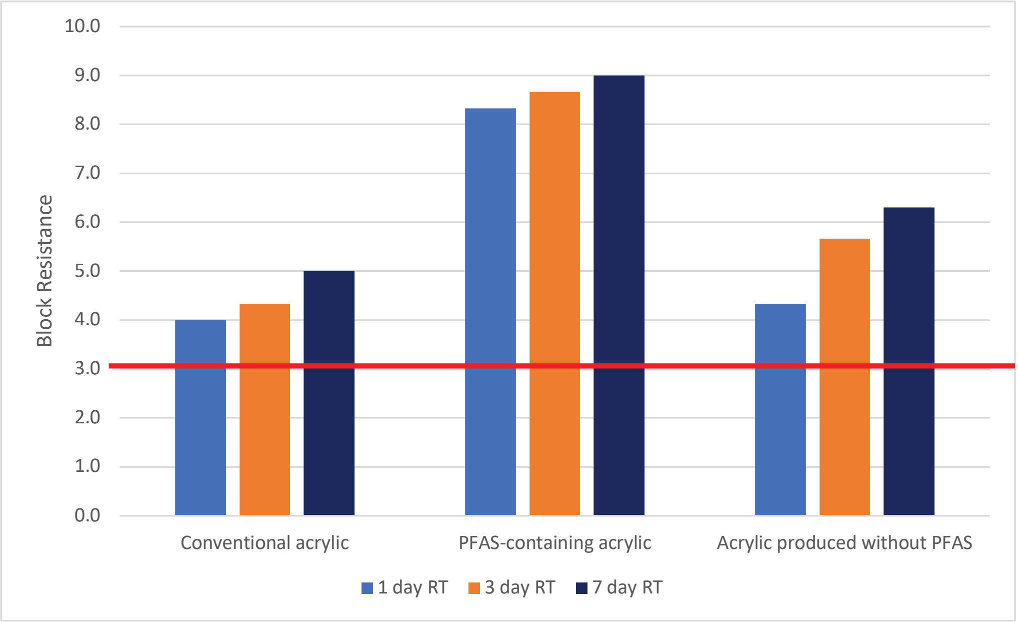
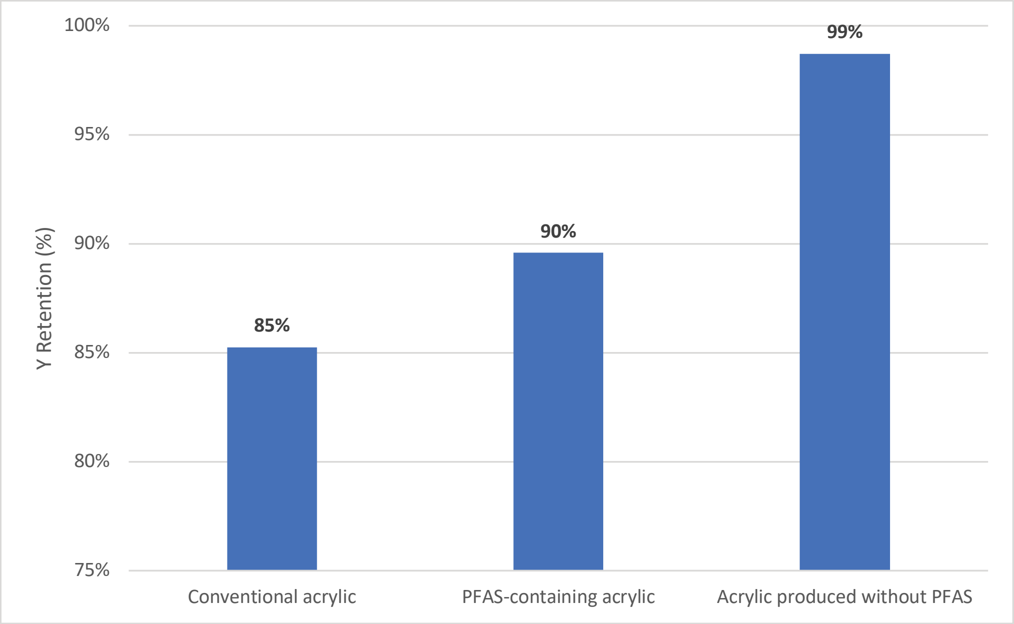


FIGURE 10
Dirt pick-up resistance of conventional and developmental acrylics in an exterior semigloss formulation.



Block resistance for this set of acrylic latexes was measured at ambient (72 °F) temperature. In this test, a rating of zero indicated that the two surfaces were stuck together and could not be separated, while a rating of 10 indicated that the surfaces did not adhere to each other, and simply fell apart. A rating of 3 or below indicated that the film ruptured when the painted surfaces were pulled apart. The use of PFAS is a long-standing practice for introducing block resistance to a waterborne latex, as evidenced by the block resistance of the PFAS-containing acrylic, when compared to the performance of the other two acrylics, both formulated without PFAS. The conventional acrylic and new acrylic produced without PFAS still achieved acceptable ambient temperature block resistance without the use of PFAS. Refer to **Figure 9**.

Dirt pick-up resistance of paint films was measured by comparing the color and condition of an aged surface treated with an environmental dirt solution to the appearance of the original, freshly applied film. A coating judged to have acceptable dirt pick-up resistance maintains color and gloss, despite repeated exposure to environmental soil. Although all three latexes evaluated exhibited exceptional dirt pick-up resistance in the exterior semigloss formulation, the PFAS-containing acrylic had improved dirt pick-up resistance over that of the conventional acrylic. The new acrylic produced without PFAS had the best overall performance. Refer to **Figure 10**.

Conclusion

As demonstrated by two different test methods, an acrylic produced without PFAS can meet the surfactant leach resistance of a PFAS-containing acrylic developed specifically for this purpose. With one exception, other performance properties for the acrylic produced without PFAS, such as hiding power, gloss development, and dirt pick-up resistance, remained at least equivalent to those of older acrylic technologies. Although block resistance, a paint-performance property attributed to PFAS, was not as pronounced in the new acrylic produced without PFAS, performance was not completely absent. Paint performance may therefore be preserved through creative polymer design, and further modulated through formulation techniques and the incorporation of additives, effectively demonstrating that a more environmentally sound acrylic can be a viable alternative to conventional all-acrylic latexes. ❊

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