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Adhesion study of high reflectivity water-based coatings

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ABSTRACT

A waterborne polyurethane coating with reflectivity higher than 95% in the visible wavelength range of 380–750 nm has been formulated. The impact of pigment concentration and carboxyl functional dispersing agent percentage on the adhesion of this waterborne polyurethane-based coating to an aluminum substrate is characterized at various pigment weight fractions by peel strength using modified ASTM D 1876-93. The formulated coating was also examined by SEM, UV/vis spectro-photometer with integrating sphere, and ATR-FTIR. Results indicate that the reflectivity of the coating increases with pigment volume concentration while the peel strength decreases. At high pigment-to-binder ratios a change in failure mode from adhesive to cohesive was observed. Further experiments indicate that adhesion increased at high pigment-to-binder ratios due to the presence of carboxyl functional groups of the dispersing agent. The increase in adhesion was identified for the first time and attributed to the interaction of the carboxyl functional groups in the dispersing agent with the aluminum surface.

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1. Introduction

Organic-inorganic hybrid materials are well established in the coatings industry as they combine the properties of organic polymers and inorganic pigments. High reflectivity coatings contribute to energy savings [1,2] in multiple applications such as fluorescent lighting panels, high reflectivity roof coatings (cool roofs), and cool coatings. Industrially available titania is one of the most important white inorganic pigments used by the paint industry due to its high refractive index, UV resistance, and high hiding power. Recent EPA regulations in the coatings industry [3] under the Clean Air Act are restricting the use of volatile organic compounds (VOCs). This has spurred the development of waterbased paints. Water borne polyurethanes are especially suitable for paints, coatings, and adhesives industries due to their inherent advantages of low VOC, fast drying, outstanding flexibility, impact resistance, abrasion resistance, non-flammability and easy adhesion to a variety of substrates [4].

High reflectivity elastomeric waterborne coatings are used in cool roof systems [5] and a number of products are commercially available [6]. However, the reflectance of these coatings is only up to 90%. Some of the alternate technologies to achieve high reflectivity coatings are powder coating of metallic particles [7,8], hollow latex particles [9], chemically modifying the

inorganic pigment surface [10], and high refractive index glass beads [11,12].

For a number of applications, the coating is required to adhere strongly to the substrate. For coatings on metal substrates, good adhesion to the base metal is required in addition to other functional properties of the coating. Mirabedini et al. [13] studied the impact of various surface treatments of aluminum on the adhesion of epoxy/polyester hybrid polymer coatings to aluminum using the pull-off and the tape tests. In a recent study [14] they also studied the effect of TiO₂ on the mechanical and adhesion properties of silicone elastomer coatings to aluminum alloy sheets. The pull-off test results showed an increase in the adhesion strength with increasing TiO₂ up to 10 wt%, beyond which, the trend was reversed. Starostina et al. [15] found that the treatment of polyethylene with primary aromatic amines increased its adhesion to steel due to the interaction of the primary aromatic amines with steel. Yan et al. [16] studied the effect of pre-heat treatment on the adhesion of bilayer polymer coatings of ethylene methacrylic acid copolymer to steel and found that the adhesion improved with increasing pre-heat treatment temperature. Suryanarayana and Mittal [17] studied the effect of pH of silane solution on the adhesion of polyimide to a silica substrate.

In this work we have investigated the effect of TiO_2 concentration (pigment-to-binder ratio) and dispersing agent percentage on the peel strength of high reflectivity (~95%) waterborne polyurethane-based coatings to an aluminum substrate. The coating morphology was studied by electron microscopy.

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Reflectivity and peel-test measurements were conducted to understand the impact of various changes in the formulation on these properties.

A procedure available to quantify coating adhesion is ASTM D 3359 [18], which could be used to measure peel strength. However, NIST [19] has shown that ASTM D 3359 has a few limitations and is primarily a qualitative method. They have reported that two coatings having the same rating as per ASTM D 3359 failed by two different failure modes, cohesive and adhesive, and that the measured peel strength values can differ by an order of magnitude. We have introduced herein a quantitative procedure with good repeatability for measuring the peel strength of coatings by modifying ASTM D 1876-93 [20] which was developed for adhesives.

The primary findings of this work are that the adhesion decreased with increasing pigment-to-binder ratio, while the adhesion increased as the carboxyl functional dispersing agent level was increased at high pigment-to-binder ratio. This reason for this increase in adhesion was identified for the first time and attributed to the interaction of the carboxyl functional in the dispersing agent with the aluminum surface.

1.1. Pigment volume concentration (PVC)

Formulations of waterborne coatings strongly depend on the desired application properties and therefore on the pigment volume concentration (PVC) as defined below [21–24].

PVC of paint is calculated by measuring the volumetric percentage of pigments in total solids present in the coating and excludes all volatiles and solvents from the calculation:

$$PVC = \frac{Volume of Pigments}{(Volume of Pigments + Volume of Binders)} \times 100$$
 (1)

1.2. Pigment-to-binder ratio (P/B)

Pigment-to-binder (P/B) ratio can also be expressed in terms of weight fraction as follows:

$$P/B = \frac{\text{Weight of Pigments}}{(\text{Weight of Pigments} + \text{Weight of Binders})}$$
(2)

2. Experimental methods and materials

2.1. Coating formulation

The coating formulation is a water-based polyurethane paint system which is cross-linked prior to coating the substrate. The preparation of the waterborne paint formulation was divided into two stages as (i) there were many different constituents and (ii) to ensure good pigment dispersion by breaking down pigment agglomerates. The two stages are the *titania pre-grind stage*, and the *let down stage*. Details of the formulation are summarized in Table 1.

2.1.1. Titania pre-grind stage

Titania pre-grind was prepared by first adding the dispersing agent to distilled water, followed by the addition of a defoamer, and finally adding the TiO_2 pigment. This mixture was then mixed with Angled Tooth Blade or Cowles Style Propeller (2.54 cm or 6.35 cm diameter), under high shear at speeds of around 2000–2400 rpm for 10 min to ensure complete dispersion and de-agglomeration of titania particles. The defoamer is added to counteract the foam created due to mixing of TiO_2 pigment in water.

The dispersing agent used was a solution of a high molecular weight block copolymer with a large number of pigment affinic groups (anchor groups with high affinity for the pigment surface), which establish electrostatic repulsion between the particles as well as steric hindrance [25–27]. The weight percentage of the dispersing agent was determined by a ladder series test in which the amount of dispersing agent was gradually increased to obtain the minimum viscosity. The defoamer used was a mixture of foam-destroying polysiloxanes and hydrophobic solids in polyglycol. The pigment used was a fine dry powder of rutile titania, with a median particle size of 0.41 μ m.

2.1.2. Let down stage

In the let down stage, the rest of the components including the binder, coalescing agent, and wetting and leveling agent were added to the titania pre-grind as indicated in Table 2. The components were then mixed with a Cowle's blade at 1400 rpm for 10 min and the formulation was set aside for 24 h to obtain the full effect of the defoamer. Different samples (Table 3) were made by increasing the titania weight in the formulations in turn increasing the *P*/*B* ratio (Eq. (2)). It should be noted that with increasing *P*/*B* ratio, the amount of dispersing agent was increased proportionally so as to maintain a fixed wt% of titania (Table 2). Complete composition and details of the formulation, being proprietary, could not be revealed.

The binder used was a waterborne aliphatic high solids anionic polyurethane (PU) dispersion with a maximum of 2.5 micronsized anionic particles. In waterborne coatings, a coalescing agent is required to aid in good film formation. We used an ester-based coalescing agent, which temporarily lowers the glass transition temperature (T_g) providing mobility to the polymer chains. The softened polymer can then flow and polymer particles can coalesce with each other. It also enables slow drying. To enhance

Table 1

Some key properties of the chemical components used in the formulations as described in Table 2.

Component	Type ^a	Solvent	Key properties
Polyurethane binder	Aliphatic anionic PU dispersion	Water	High solids ^a pH: 7.0–9.0
Dispersing agent	Solution of a high molecular weight block copolymer with a large number of pigment affinic groups	Water	SC ^b : 40 wt% AN ^c : 10
Polysiloxane defoamer	Mixture of polysiloxanes and hydrophobic solids	Polyglycol	SC ^b : $>$ 98 wt%
Coalescing agent	Ester-based	Dispersible in water	SC ^b : 100 wt%
Wetting and leveling additive	Acrylic modified polyester	DPM	SC ^b : 50 wt%
Cross-linker	Aliphatic polyisocyanate based	Dispersible in water	SC^{b} : > 98 wt%

^a Proprietary formulation, hence more details could not be revealed.

^b Solid content.

^c Acid number (mg KOH/g).

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