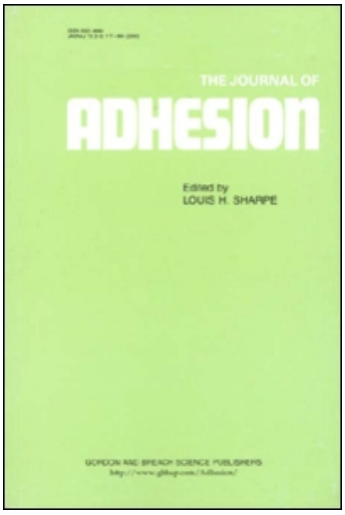

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A Method for Measuring the Adhesion Strength of Powder Coatings

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A method for testing and quantifying the adhesion strength of particulate coatings was developed. A gas impingement test was developed to measure adhesion forces of electrophoretically-deposited layers of phosphor particles for use as information display screens. A high pressure jet of nitrogen gas is directed perpendicularly to a particulate deposit. The particles are removed from the substrate in a ring pattern which can be correlated to the skin friction on the substrate, giving a measurement of adhesion strength in the range of 100 to 450 Pa.

Keywords: Adhesion strength; powder coatings; gas jet impingement; electrophoretic deposition; phosphor

INTRODUCTION

It is important to understand and, if possible, to improve the adhesion of phosphors deposited by electrophoretic deposition (EPD), as this is a major limitation of this display technology. EPD is a technique in which charged particles suspended in a non-aqueous solution are deposited onto a substrate under the influence of an electric field. The system of interest in this study consists of 3 μm diameter ZnS:Ag phosphor particles suspended in isopropyl alcohol containing 10^{-3} M $Mg(NO_3)_2$. The $Mg(NO_3)_2$ charges the particles positively causing them to migrate to the cathode under an applied electric field. At the

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cathode, $Mg(NO_3)^+$ reacts with water to form $Mg(OH)_2$ and with isopropyl alcohol to form $Mg(C_3H_7O)_2$. These hydroxides and alkoxides are the binder material [1].

The strength of particulate films is determined by both adhesion, the interfacial strength of a material to a substrate, and cohesion, the interfacial strength between two similar particles [2]. In this study, adhesion will refer to a macroscopic property of a deposit on a substrate which will include both adhesion and cohesion.

There are many physical and chemical forces which are responsible for particle adhesion. Van der Waals, electrostatic, and magnetic forces, as well as covalent chemical bonding, hydrogen bonding, adsorption and chemisorption all can play a role in particle adhesion. In addition to atomic or molecular forces, macroscopic forces due to a physical adhesive or capillaries can also greatly affect adhesion.

Capillary forces are particularly important when a material is hydrophilic, such as $Mg(OH)_2$. The capillary force (F_c in N) between two particles is given by [3]:

$$F_c = 4\pi r\gamma \tag{1}$$

where r (m) is the particle diameter and γ (mJ/m^2) is the liquid surface tension. There is evidence that liquid can remain in the particle lattice even after baking the material above the liquid's boiling point. Also, if there is crystallization upon evaporation, bridges may form between the particles. Both of these phenomena can increase the adhesion of the particles by an order of magnitude [3].

There are a number of adhesion measurement techniques for thin film ($< 1\ \mu m$), thick film ($> 1\ \mu m$) and bulk coatings ($> 25\ \mu m$). The following are some of the methods used for determining adhesion strength of thick films or bulk coatings [2]:

1. Pull-off The thin film, if thick enough, is pulled directly from the substrate. The force required to pull the film from the substrate is the adhesion strength. (Adhesion force: $\sim 10^7$ Pa)
2. Ultracentrifugal The coating is placed in an ultracentrifuge in air facing away from the center. The force (F in N) acting on the film or particle is [4]:

$$F = V\rho\omega^2 l \tag{2}$$

where $V(cm^3)$ is the volume of the deposit, $\rho(g/cm^3)$ is the density of the film or particle, $\omega(rad/s)$ is the angular velocity of the centrifuge, and l (cm) is the radius of rotation. (Adhesion force: $\sim 10^{-8} - 10^{-5}$ N for ~ 3 μm diameter particles, $\sim 2 g/cm^3$)

3. Ultrasonic The film is placed in front of an ultrasonic horn. The force (F in N) acting on the film is [4]:

$$F = m(4\pi^2 \nu^2 y) \tag{3}$$

where m(kg) is the mass of the film, $\nu(s^{-2})$ is the frequency of the sonic force, and y(m) is the distance from the horn. (Adhesion force: $\sim 10^{-7}$ N)

4. Adhesive Tape A piece of adhesive tape is attached to top side of the film and then pulled, perpendicularly away from the substrate. (Adhesion force: ~ 50 N)
5. Tangential Shear In this method a tangential shear is applied to the film either by passing a fluid over the film or attaching a grip to the top of the film and mechanically shearing the deposit. (Adhesion force: ~ 10 N)
6. Tension Test The film is pulled apart until it fractures. The force applied at the fracture point is a measure of adhesion. (Adhesion force range: $\sim 10 - 10^2$ N)
7. Knife or Scribe Test A knife or other sharp device is placed on the substrate. The force required to scrape away the film is a measure of adhesion. (Adhesion strength range: $10^7 - 10^9$ Pa)

All of these methods are useful when measuring adhesion of thick films [2]. Unfortunately, they are inappropriate for testing the adhesion strength of powder coatings. The adhesion to be measured is either too strong, as is the case of ultrasonic and ultracentrifugal methods where no particles are dislodged, or too weak as is the case of a pull-test or adhesive tape test where all particles are removed from the substrate.

Adhesion measurement methods have been designed specifically for powder coatings [5]. The techniques include horizontal and vertical tensile strength tests, as well as shear strength tests. Unfortunately, these methods are designed for packed powder beds, which are self-supporting, or use a split cell design. The force to split the cell is the

measure of the adhesion strength. Depending on the material being tested, the powder–powder adhesion may fail first, or the powder–substrate adhesion may fail first.

There are qualitative adhesion tests for phosphor coatings. The deposits are tested by placing them under a nitrogen gas jet or a water jet and increasing the impingement force until particles are removed [6, 7]. The weight % of particles remaining on the substrate or the pressure at which particles are clearly removed is used as an adhesion measurement.

In the manufacturing of screens for advanced displays, layers of small luminescent particles may be deposited by electrophoretic deposition (EPD). Electrophoretic deposition of phosphor particles is currently used, particularly for fine particles in the range of 1 to 10 μm in diameter, in the manufacturing of high resolution screens. A set of processing variables that enhance the adhesion strength of EPD phosphor coatings have been determined. Post-deposition baking at 425°C for 1 hour; addition of water or glycerin and the use of $Y(NO_3)_3$ instead of $Mg(NO_3)_2$ in the deposition bath, as well as the particle size distribution, can all enhance the adhesion strength of EPD phosphor deposits. The strongest coatings were deposited from a bath with 2% glycerin added. They still failed the tape test completely. An ultrasonic horn, with a maximum force of 10^{-7} N, was unable to dislodge any particles. An adhesion test which falls between these two limits was needed. This paper describes a nitrogen gas jet impingement method to measure, both qualitatively and quantitatively, the adhesion strength of particulate phosphor coatings.

EXPERIMENTAL

The adhesion strength was tested by applying a jet of N_2 gas perpendicularly to the coating. The testing apparatus is shown schematically in Figure 1. A high-purity nitrogen tank is connected to a pressure gauge on the apparatus. A timer starts and stops the flow of nitrogen after a set time. One of two nozzles with either a 1 or 2 mm diameter opening was connected to the exit tube.

The apparatus and procedure to deposit phosphors electrophoretically are described elsewhere [8]. The samples tested correspond to

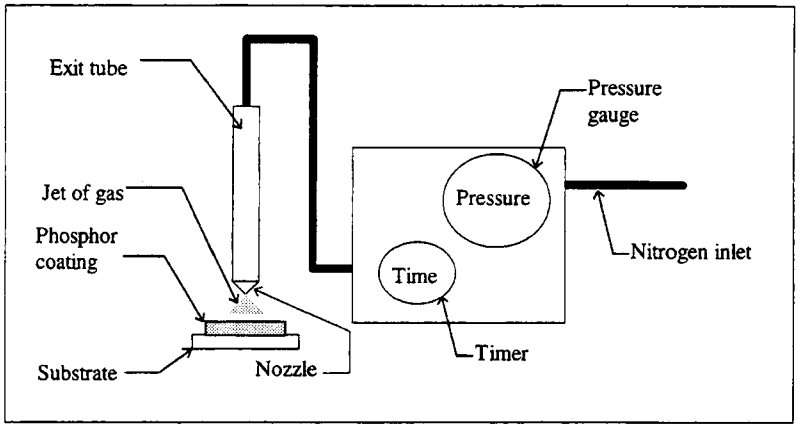

FIGURE 1 Adhesion testing apparatus.

those parameters which enhanced the adhesion strength as previously stated. Typical deposition conditions deposited 2 mg/cm^2 (15 μm thickness, 3 μm average diameter) of phosphors. The phosphor-coated screens were categorized as either weak or strong. These conditions correspond to settings of the pressure gauge, the nozzle diameter and the amount of phosphor removed during the adhesion test. The qualitative method was used to measure adhesion strength for weak screens, while the quantitative method was used for strong screens. The apparatus shown in Figure 1 was used for both cases, with slightly different operating parameters. The two pressure settings used were 40 psi and 30 psi (275.8 and 206.8 kPa) for the 1 mm and 2 mm nozzles, respectively. If the 2 mm nozzle setting removed more than 80% of the phosphor from the substrate, the 1 mm nozzle setting was used. The deposit was placed 0.5 cm below the tip of the gas nozzle. The timer was started for a 10-second period. During this time, the nitrogen flow was increased for 4 seconds to a steady-state flow, where it remained for 4 seconds and then decreased to no flow for 2 seconds. The nitrogen gas flow was ramped to reduce the effect of the transient behavior of the jet. Quantification of the adhesion strength is dependent on the steady-state flow of the jet and will be discussed later.

RESULTS AND DISCUSSION

Qualitative Adhesion Measurement

In order to study the effects of different EPD processing parameters [1] on the adhesion strength of phosphor deposits, a standard means of comparison needed to be developed. The first method used to compare adhesion strengths was a qualitative gas impingement method. The weight percentage of particles remaining on the substrate after testing was the measure of adhesion. The experimental conditions used for this method were a 40 psi pressure setting and the 1 mm diameter nozzle. This method is accurate to ± 5 wt.%. This method is useful for comparing the adhesion strength of deposits using the same apparatus and set-up. However, the results cannot be compared with results from other adhesion testing methods.

Quantitative Adhesion Measurements

A quantitative method was necessary to enable comparison of deposits tested with the nitrogen gas impingement test and deposits tested by other means. It is desirable to measure the adhesion strength in units of force, as this would allow comparisons of adhesion data from the nitrogen gas impingement tester with standard methods used for testing other deposits, particularly the tape test.

In order to determine the adhesion strength using the same nitrogen gas tester apparatus as used for the qualitative method, the shear stress on the particles needed to be estimated. If the flow of nitrogen against the substrate is approximated by wall flow, the skin friction (τ_w in N/cm^2) along the surface is approximated by [9]:

$$\tau_w = 0.0225\rho U^2 \left(\frac{2\nu}{U\delta}\right)^{1/4} \tag{4}$$

where ρ(g/cm^2) is the gas density, U(cm/s) is the gas velocity upstream, δ(cm) is the thickness of the boundary layer and ν(cm^2/s) is the gas kinematic viscosity. Eq. (4) comes from the analytical solution to flow in a pipe which has been shown to have similar boundary layers as the planar wall jet, and then was adjusted to agree

with the Blasius law [9]. The boundary layer thickness is proportional to $d^{4/5}$, which gives [9]:

$$\tau_w = 0.0225\rho U^2 \left(\frac{2\nu}{Ud^{4/5}}\right)^{1/4} \tag{5}$$

as a estimate of the shear stress, where d is the distance from the center of the substrate and measures the radius of the ring of material removed after adhesion testing. A schematic of the disk after adhesion testing for measurement of d is shown in Figure 2. The gas velocity, U, is calculated from the measured flow rate (Q, in cm^3/s) divided by the nozzle area (A, in cm^2). Table I shows the values of the parameters used in this study. A particle at the outermost region of the ring of material removed will experience a shear force equal to its adhesion force. Therefore, measurement of this shear force is a measurement of the adhesion strength of the deposit.

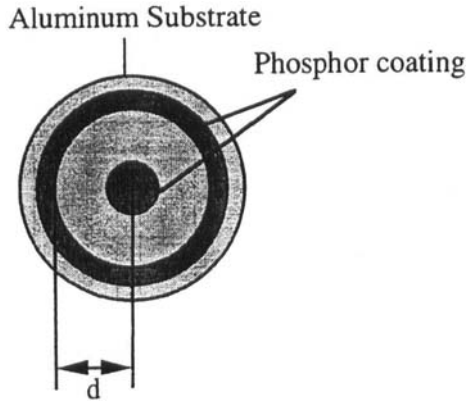

FIGURE 2 Determination of ring radius of particles removed by gas jet impingement.

TABLE I Parameters used for quantitative adhesion testing calculations

Nozzle pressure/diameter	*Gas density* $\rho(g/cm^2)$	*Kinematic viscosity* $\nu(cm^2/s)$	*Gas velocity* $U(cm/s)$
30 psi (0.21 MPa)/2 mm	2.35×10^{-3}	0.077	20,800
40 psi (0.28 MPa)/1 mm	3.14×10^{-3}	0.057	29,900

There were several key assumptions made in order to use Eq. (5). First, the flow field is assumed to approach ideal wall flow from an impinging jet as shown in Figure 3. The axisymmetric flow leaves the nozzle and impinges on the wall. Along the center line, there is a stagnation point. Away from the center line, the fluid spreads out radially. With the exception of the boundary layer, the fluid is considered to be inviscid. The next assumption is that the boundary layer developed is similar to that developed by flow inside a pipe [9, 10] and that the particulate deposit does not alter the boundary layer. This allows use of the analytical solutions for pipe flow to be applied to the laminar wall jet. The final assumption is that the equations governing the laminar wall jet are the same as those governing a turbulent wall jet, which has been previously explained [9]. While this method is not entirely rigorous, it does provide a first estimate of the adhesion strength of the particulate film.

A deposit which has been tested is shown in Figure 4. The material in the center of the substrate remains due to the fact that there is a stagnation point in the flow along the axis of symmetry. The experimental setup for these measurements is the same as that for the qualitative method except that the outlet pressure was 30 psi and the nozzle diameter was 2 mm, leading to a higher gas velocity. The area of

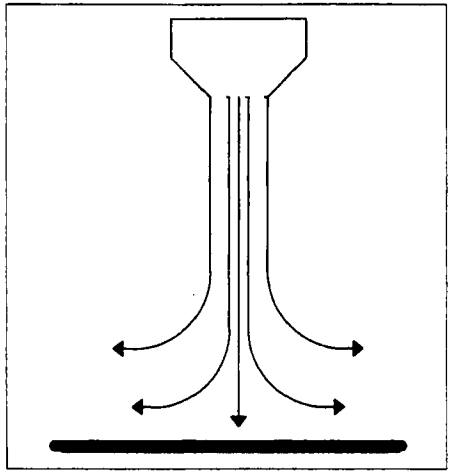

FIGURE 3 Ideal wall flow from a jet.

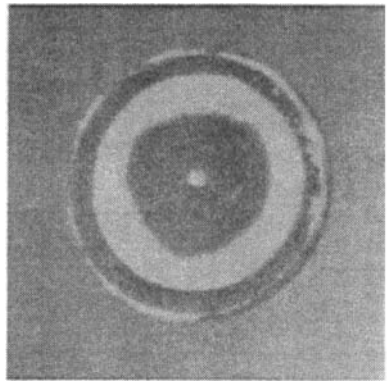

FIGURE 4 Particulate deposit after adhesion test (diameter 2.54 cm).

phosphor removed is not perfectly circular. In these cases, an average radius is calculated for the purposes of adhesion measurements by averaging the radii measured at four quarters of the area of removed particles.

Adhesion strengths for this method vary from a low value of 100 Pa using the 40 psi (0.28 MPa)/1 mm setting to a high value of 450 Pa using the 30 psi (0.21 MPa)/2 mm setting for d ranging from 1.6 to 0.1 cm. Experimental error is estimated at ± 80 Pa. The ranges of adhesion strength for each set of conditions is shown in Figure 5 as calculated from Eq. (5). The tape test can measure a force of 5 to 6 g ($1.5 \times 10^5 - 1.8 \times 10^5$ Pa) and the ultrasonic horn can measure a force of 3×10^{-4} Pa. The quantitative gas impingement method tests adhesion strength in an intermediate range of approximately 100 to 450 Pa. This range can be increased by changing the nozzle diameter and the applied pressure.

Comparison of Qualitative and Quantitative Adhesion

Both the qualitative and the quantitative adhesion testing methods remove particles from the center of the deposits. The qualitative is used for weaker deposits, which are completely removed with the conditions of the quantitative method.

While the qualitative adhesion testing method is useful in determining the adhesion strength of deposits formed by EPD, the quantitative

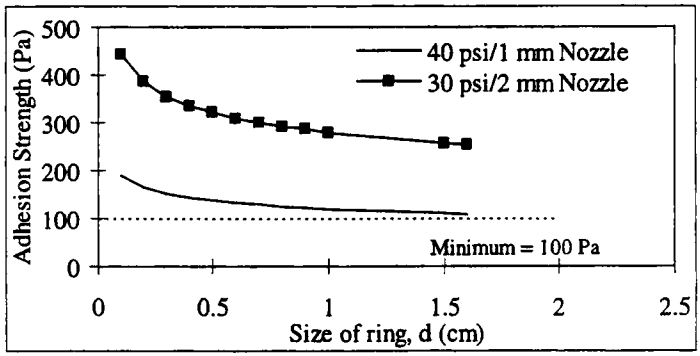

FIGURE 5 Range of adhesion strength measurements by jet impingement.

adhesion testing method was developed so that deposits with adhesion strengths in the range of 100 to 450 Pa could be analyzed more precisely. Figures 6 and 7 show the adhesion results of the quantitative and qualitative methods, respectively. From Figure 6, it is clear that the deposits from the bath with 3% added water are much weaker than deposits deposited with 2% glycerin in the bath. However, Figure 7 shows that the adhesion strength of the deposits from a bath with 3% added water are similar to the adhesion strength of those deposits from a 2% added glycerin bath. These differences can be attributed to the sensitivity of the two adhesion testing methods.

As the adhesion strength of the deposit increases, the qualitative test is unable to measure the strength as well as the quantitative test. This is illustrated in Figure 5. The region of steepest slope, *i.e.*, most

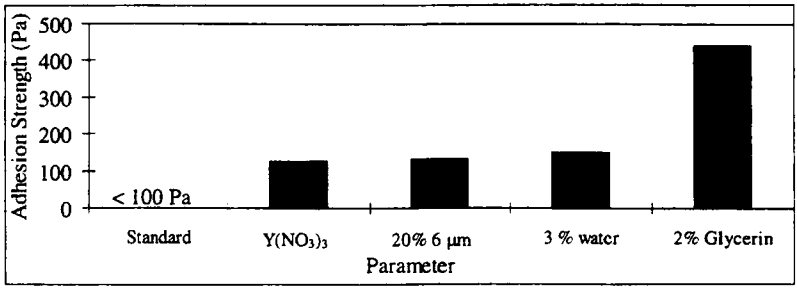

FIGURE 6 Quantitative adhesion data.

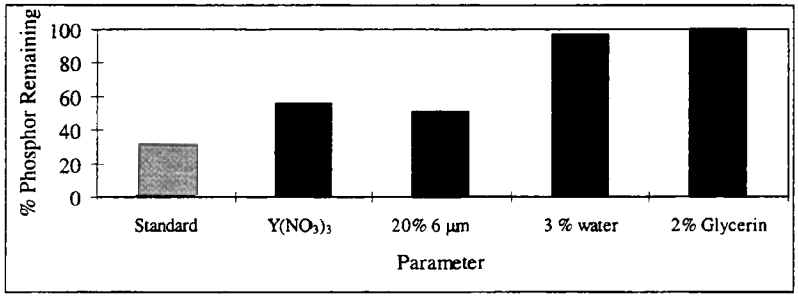

FIGURE 7 Qualitative adhesion data.

sensitivity, for the quantitative tests are at the higher adhesion strengths, 300–450 Pa for the 30 psi (0.21 MPa)/2 mm setting. At lower adhesion strengths the slope is flatter, *i.e.*, low sensitivity. The reverse is true of the qualitative test. At high adhesion strengths, there is very little difference in wt.% of particles remaining while, at lower adhesion strengths, the resolution of the measurements are adequate for comparison. When adhesion strengths become >60% for the qualitative test, the quantitative test is more appropriate to use.

CONCLUSIONS

A method for testing and quantifying the adhesion strength of EPD phosphor coatings was developed. Previous testing methods either were too weak to dislodge any particles, such as the ultrasonic horn method which measures forces of the order of 10^{-4} Pa, or were too strong and removed all particles, such as the tape test which measures forces of approximately 10^5 Pa. A gas-impingement test was developed which can measure adhesion forces in the range of 100 to 450 Pa.

While this method was developed specifically for phosphor deposits, it is applicable to any particulate deposit which has an adhesion strength in the appropriate range. This method can also give qualitative results by comparing the percent of particles remaining on the substrate after testing. This is a simple measurement, but it does not have the resolution of the quantitative method. For this reason, there is no direct correlation between the qualitative adhesion results and the quantitative results.

Acknowledgments

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