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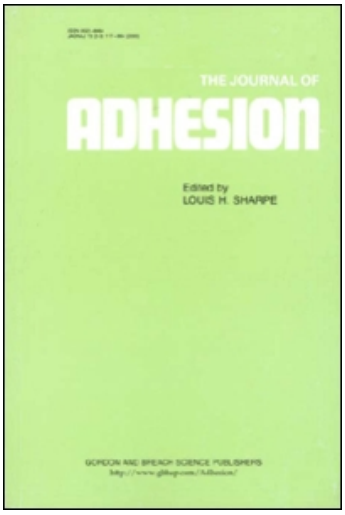

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QUANTITATIVE EVALUATION OF ADHESION STRENGTH OF INK DEPOSITED ON PLASTIC FILMS WITH A NANO INDENTER AND A SCANNING PROBE MICROSCOPE

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As our lifestyles change, the convenience food industry booms. Many convenience food products use flexible, boilable, and microwavable plastic packaging. Almost every package is printed with ink, promoting what is inside the package, listing ingredients and nutrition data, giving preparation instructions, etc. The adhesion of ink on plastic films merits increasing attention to ensure quality packaging. However, this property has not been systematically studied and lacks a scientific method to measure the adhesion strength quantitatively. We are developing a technique of using a Nano-Indenter and a Scanning Probe Microscope to evaluate the adhesion strength of ink deposited on plastic films. It shows promise, and the

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measurement will be helpful in better understanding the adhesion mechanism, thus giving direction as to how to improve the adhesion.

Keywords: Adhesion strength; Adhesion measurement; Interface failure; Ink adhesion; Adhesion on plastic film; Nanoscratch; Scanning probe microscope

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INTRODUCTION

As society modernizes, our lifestyles are constantly changing along with our eating habits. The microwave oven is becoming a standard feature in our homes. That fact alone has changed the nature of food packaging from metal to plastics. In addition, more and more households now have two adults working outside the home. That has attracted more and more manufacturers to convenience foods—and many of these new convenience food products have gravitated toward "boil-in-bag" or microwaveable, flexible packaging. And, aside from food products, countless other manufacturers have found flexible plastic packaging economically efficient and advantageous with respect to marketing. Virtually every package is printed with ink. The adhesion of ink to plastic films is, therefore, drawing more and more attention to ensure quality packaging [1], and the need for quantitative assessment of the adhesion is quite apparent. A cornucopia of methods to measure adhesion is available, ranging from very primitive to very sophisticated [2–6]. Some of these methods are qualitative in nature and no numerical values can be obtained, while the others may provide quantitative measurements. However, no quantitative measurement method of the adhesion strength of ink deposited on films of plastics has been documented.

Evaluation of adhesion strength of ink on plastic films is very challenging. Unlike some coatings and substrates that are crystals or inorganic materials, inks and plastic films are polymeric materials with complicated molecular structures. Typical plastic films used in packaging include polypropylene, polyethylene, polyester, and polyamide. These materials are all nonporous and cannot rely on any absorptive mechanisms to aid adhesion or drying. The surface energy of plastic films usually is low, ink does not wet the films well, and as a result the adhesion strength is often minimal. The jelly-like ink is in the liquid phase during printing, and becomes a film after drying. The thickness of the ink is only about 500 nm or less, and the thickness of the plastic film is no more than 50 microns. In addition, the film is typically not rigid, but soft and flexible. Laying such a thin, soft and

flexible film on a steel disk evenly so we can carry out the scratching test was the first challenge we faced, which would not be encountered in handling solid bulk substrate. It is very difficult to carry out a precision evaluation of the adhesion at such a small scale on the flexible material.

Fortunately nano-technology has been daily expanding its applications in recent years. The nanoscratch technique has been used by several groups in the quantitative study of adhesion of different types of coatings [7–10]. As a trial, we have used a Nano-Indenter (MTS systems Corp., Oak Ridge, TN, USA) combined with a Scanning Probe Microscope (SPM), NanoScope IIIa (Veeco Metrology, Santa Barbara, CA, USA), to carry out a quantitative measurement of adhesion strength of ink on plastic films. The experimental procedure is described below.

EXPERIMENTAL PROCEDURE

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In the test, 10 mm × 10 mm samples of laboratory prints of ink on plastic film were prepared. A small amount of epoxy was used to affix one edge of the film to a steel disk, and the remaining film was allowed to lie freely on the disk. We used a Nano-Indenter to perform the test. Our Nano-Indenter XP is one newly developed by MTS. It performs both nanoindentation and nanoscratch tests with a single head. The head is equipped with an electromagnetic force actuator, which can generate a normal force up to 650 mN with a resolution of 0.05 μN in the indentation tests as well as in scratch tests. In the scratch tests, the indenter first does a prescan under a light nondestructive force, *e.g.*, 10 μN, to measure and record the contour of the surface. It then scratches the surface with a selected mode, followed by a postscan to find the residual depth of the "ditch". The vertical movement of the tip is measured by a capacitance displacement gauge; it can measure a range up to 2 mm with a resolution of 0.02 nm. The real-time penetration depth and the residual depth are the vertical movement of the tip during scratching and postscan minus the contour in prescan, respectively. In the test, the Indenter, operated in the scratch mode, scraped the ink for a distance of 100 μm, 200 μm, or 500 μm under an increasing normal load. The range of the increasing load could be from 0–3 mN, 0–5 mN, 0–10 mN, etc., depending on the adhesion strength of the tested samples. The scraping speed is about 5 μm/s. The direction of scraping is perpendicular to the glued edge and toward the free side. A conical-shaped diamond tip of 90° with a radius of 1 μm at its apex was used in the Indenter for the scraping. As the normal force applied to the tip was increased, the penetration depth of the tip that moved laterally along the surface of the ink into

the ink was increased, and an increasing shear stress, generated by the scraping tip, propagated into the subsurface of the ink with its maximum point located underneath the scraping tip, according to tribology theory [11]. When the propagating stress reached the interface between the ink and plastic film and exceeded the strength of the adhesion, it destroyed the interface and peeled the ink off the plastic film. Attention was paid to the critical force at which the interface began to fail and the ink began to peel off. The critical force was used to characterize the strength of adhesion. The scratched ink and exposed plastic film were examined by the Scanning Probe Microscope operated in the contact mode, as well as in the lateral force mode to verify the interface failure.

To illustrate the test, Figure 1a is a two dimensional (2D) gray-scaled topographic image of a segment of scratch made under a force from 2.9 mN to 3.1 mN at the surface of a sample. The applied normal force increased linearly with the scraping distance. Figure 1b is the profile along the bottom of the ditch. As can be clearly seen, the interface began to fail at the middle, after the scraping tip passed the middle point of the segment and the normal force exceeded 3.0 mN; thus, the ink was peeled off and the tip reached the substrate immediately. Figure 1c is a 3D version of the image, and Figure 1d is a lateral force image, taken simultaneously with the topographic image (Figure 1a), which further confirmed the exposure of the substrate. The lateral force image is a map of the lateral force the scanning tip encountered at every point. The lateral force is mainly influenced by the frictional force between the tip and the sample surface in a flat region, but it is also affected by the features at the surface. When the tip scans upward over a feature it experiences a larger lateral force. Conversely, when the tip scans downward it experiences a smaller lateral force. In Figure 1d, the brighter area indicates the higher lateral force and the darker area indicates the lower lateral force, and the tip scanned from the left to the right. Before the ink was peeled off, the shape of the ditch was approximately the shape of a triangle, similar to the cross section of the conical-shaped tip. The scanning tip moved downward and upward over the ditch, encountering less and more lateral force, compared with the force encountered at the flat surface. After the ink was peeled off and the substrate was exposed, the scanning tip experienced a frictional force at the bottom of the ditch between the tip and plastic film, which was completely different from the frictional force between the tip and the ink outside the scratch, as clearly seen in the lateral force image. It further confirmed that the adhesion, as well as interface, failed at Point A. Thus, the critical force for interface failure was estimated to be 3.0 mN.

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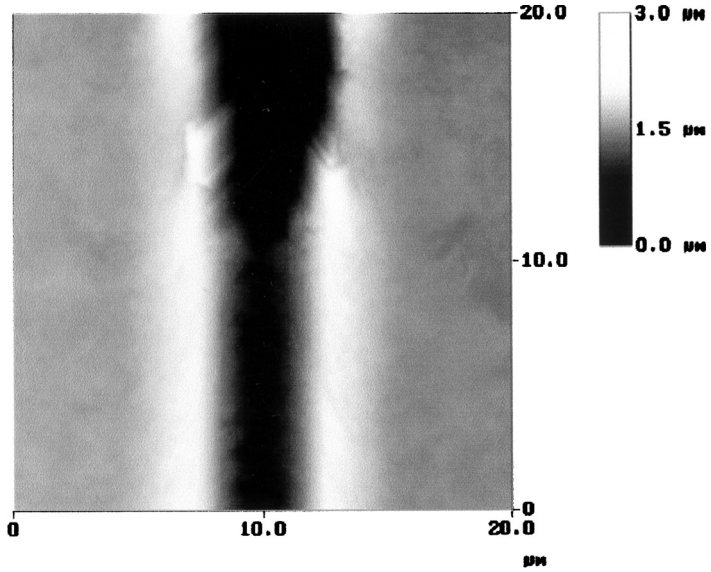
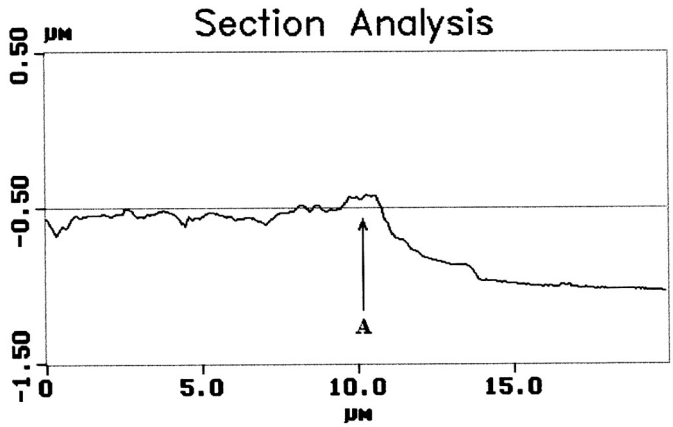


(a)



(b)

FIGURE 1 (a) 2D gray-scaled topographic image of a scratch segment made under an increasing force from 2.9 mN to 3.1 mN at the surface of a sample. (b) Profile along the bottom of the ditch. (c) 3D view of the topographic image. (d) Lateral force image taken simultaneously with the topographic image. (*Continued.*)

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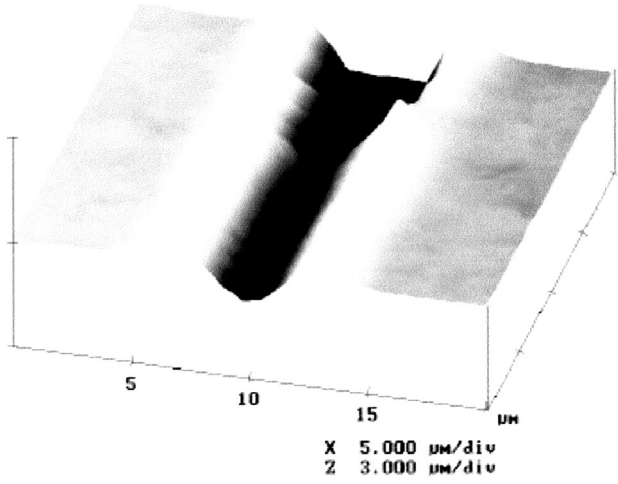
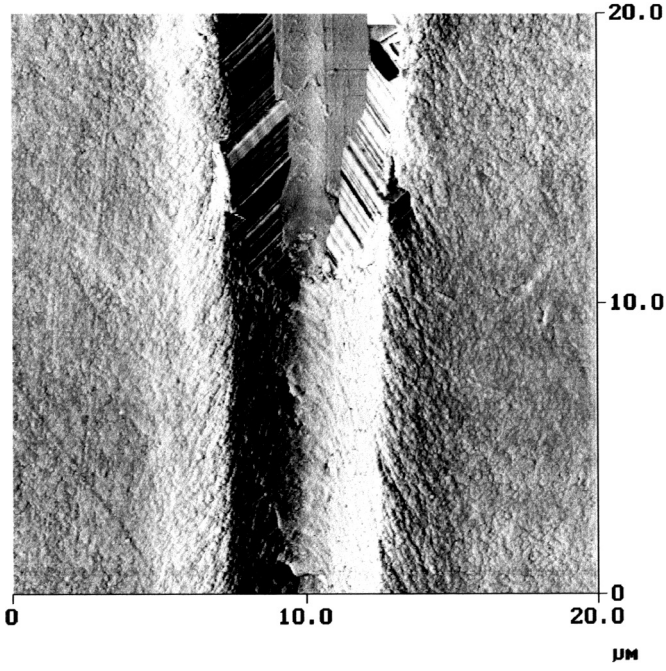


(c)



(d)

FIGURE 1 (Continued).

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TEST RESULTS AND DISCUSSION

Using the above technique, we have measured the adhesion strength of several dozen samples, different inks deposited on different plastic films. As an example, the test results of four samples, two different inks, labeled as A and B, deposited on two different plastic films, oriented polypropylene and polyester, are presented here. The test was performed at room temperature and in ambient conditions.

Ink A is a phthalocyanine blue flexo ink based on nitrocellulose and polyurethane, while Ink B is a phthalocyanine blue flexo ink based on nitrocellulose and plasticizer. The polypropylene film is T-523-3 corona-treated oriented polypropylene (AET Films, New Castle, DE, USA), and the polyester film is DuPont 48 LBT corona-treated polyester (DuPont Teijin Films, Hopewell, VA, USA). These purchased polymeric films were manufactured by an extrusion process into a thickness of about 12 μm.

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The prints were made in the laboratory using a Saueressig precision gravure proofer and were air dried.

Our Sample 1 is Ink A deposited on polypropylene; Sample 2 is Ink B deposited on polypropylene, Sample 3 is Ink A deposited on polyester, and Sample 4 is Ink B deposited on polyester. We used the technique described above to evaluate the critical force for adhesion failure with an increasing load from zero to 5 mN for Samples 2, 3, and 4, and with an increasing load from zero to 10 mN for Sample 1, since Sample 1 did not show apparent failure up to 5 mN. The scraping distance is 200 μm, and the scraping speed was 5 μm/s. We have made 10 measurements for each sample. The measured critical forces are plotted in Figure 2. As one can see, the results are scattered around with relatively large deviations and showed no "definite" value, which is not due to measurement error, but indicates the fact that the adhesion strength varies along the interface.

We have double-checked the technique and experimental procedure and confirmed that the scattered values of measured adhesion strength in the present test were not caused by measurement error. Using the same technique to measure adhesion strength of a plasmaenhanced chemical vapor film deposited on a siloxane-coated polycarbonate, we had a deviation as small as 1%.

The fluctuation of the adhesion strength may be attributed to the spatial variation of the chemical end groups on the surface of the plastic film, such as crosslink density, and the nonuniformly distributed pigments and polymers at the bottom of the ink film which could lead to an inhomogeneous interface, resulting in larger deviations of

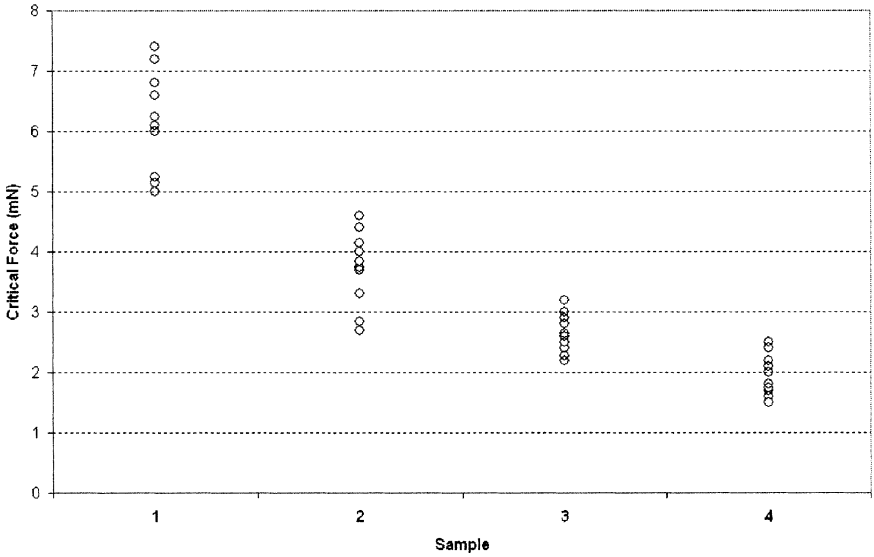

FIGURE 2 Measured critical forces for adhesion failure of samples 1, 2, 3, and 4 in the 10 tests. The averaged critical forces of samples 1, 2, 3, and 4 were 6.1 mN, 3.7 mN, 2.6 mN, and 2.0 mN, respectively, with deviations of 15%, 17%, 13%, and 14%, correspondingly.

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measured adhesion strength along the interface. The adhesion is stronger in some regions and weaker in others.

Although the fluctuation was relatively large and the measured values of the four samples overlapped to some extent, they were still distinguishable in their adhesion strength. The average critical forces for adhesion failure of Samples 1, 2, 3, and 4 were 6.1 mN, 3.7 mN, 2.6 mN, and 2.0 mN, respectively, with a deviation of 15%, 17%, 13%, and 14%, correspondingly. Irrespective of the close similarity of Ink A and Ink B, we could distinguish their subtle difference in adhesion strength by taking the average value of the measured critical forces from a large number of measurements. Thus, we think the resolution of our method is reasonably good.

While the critical force is a useful characteristic for adhesion strength, we found the configuration of interface failure also plays an important role. Different combinations of ink and plastic film showed different configurations of interface failure in the adhesion tests.

To illustrate this, three different configurations of interface failure were shown in a, b, and c of Figure 3. In a, adhesion failed at Point A; the ink began to be peeled off from Point A and continued being peeled

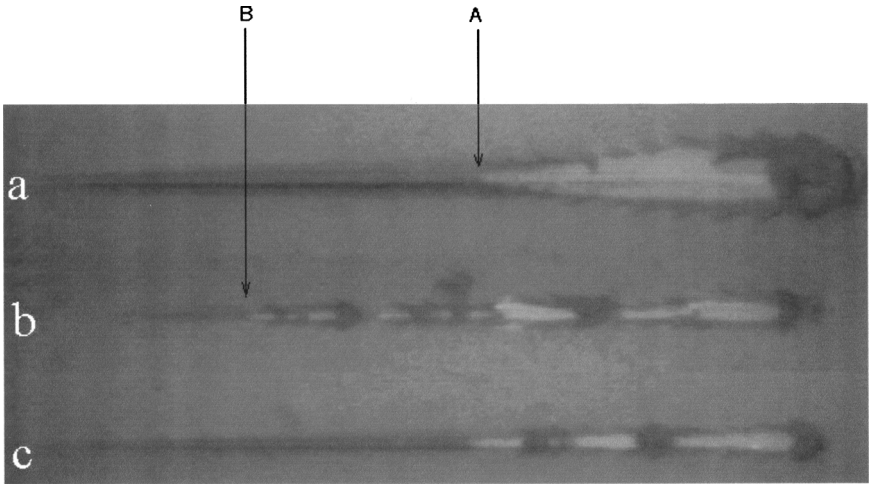

FIGURE 3 Different configurations of interface failure.

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up to the end of the scraping, having a ditch with increasing width. In contrast, in b, adhesion failed and ink was peeled at Point B. As a consequence, the accumulated energy in the interface was released and the stress was reduced, which prevented the ink from being further peeled off until the accumulated stress prevailed against the adhesion strength again and the ink was peeled off for the second time. In this case, the configuration of the failure usually consisted of a series of discrete damaged spots of peeled ink. The length of the damaged section of peeled ink became longer and longer and the interval between two consecutive damaged sections became shorter and shorter, as the normal load increased. In Figure 3c was similar to b; however, the critical force was larger and the damage was less severe.

Comparing Cases a and b in Figure 3, although the critical force for adhesion failure of ink a, F_A, is larger than the critical force for adhesion failure of ink b, F_B, it does not necessarily mean ink a will behave better than ink b all the time. If the normal force in the scraping is larger than F_B but smaller than F_A, ink b will fail but ink a will not, and we can definitely claim a is better than b. However, if the normal force is larger than F_A, both will fail, but the configuration of failure of ink a is much more severe than ink b, and we may think b behaves better than a. This indicates that using only the critical force for adhesion failure to evaluate the adhesion is not enough; the configuration of the failure complements the critical force in characterization of adhesion.

Different configurations of interface failure may be attributable to the different bonding mechanisms between different inks and different plastic films. To understand this an intensive and systematic study is needed.

Another factor that should to be mentioned is that, based on the tests we carried out on several dozen samples, different inks deposited on different plastic films, the adhesion degrades gradually over a time scale of several weeks to several months. The mechanism of the degradation is not clear now. The continuous loss of solvent/plasticizer might be one of the causes that merits further study.

SUMMARY

Adhesion of ink deposited on plastic film merits an investigation due to its increasing applications. Up to now, no scientific quantitative measurement method for the adhesion of ink on plastic films has been documented. It is a challenging task, regarding the jelly-like ink of 500 nm thickness deposited on flexible plastic films about 50 μm thick. We are trying to establish a reliable and scientific method to objectively evaluate the adhesion strength, by quantitatively measuring the critical force for adhesion failure using a newly developed NanoIndenter and a scanning probe microscope. Although the effect of the experimental conditions, such as scratching speed, loading rate, etc., on the measured critical forces needs further systematic study, we believe this new technique has the potential to be incomparably superior to the conventional tape adhesion test used widely now. In the conventional test, a commercially available grade of adhesive tape is affixed by hand to the ink surface, and is then removed by peeling the tape off the surface. The degree of adhesion is qualitatively characterized by the amount of ink remaining on the plastic film. Variations on this test include the differences in force applied during application of the tape, the rate of tape peel, and the angle of tape peel. In addition, the tape adhesion test does not discriminate between adhesive and cohesive forces. In contrast, the test conditions of critical force measurement can be well controlled, set the same for all the tested samples for an objective comparison, and provide a scientific quantitative evaluation of the adhesion strength. Combined use of a scanning probe microscope to examine the configuration of interface failure provides useful information in studying the adhesion mechanism. Thus, it helps us to understand better the mechanism of adhesion and improve the performance of printing inks on plastic films. Investigation of the adhesion mechanism of different inks on

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different plastic films, and exploration of the improvement using this measurement method will be reported in future papers.

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