Effect of Water Diffusion on Adhesion Strength

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Abstract:

The adhesion strength between Polyethylene (PE) film and Aluminum surface by using the adhesive material (Cyanoacrylate) has been studied. Aluminum (Al) was used as a substrate, and polyethylene (PE) was used as a film adhered to the Al surface. Standard specimens were prepared to use in the peeling test in dry condition, other specimens were immersed in water for 12 days at room temperature. the results for the specimens in the dry condition had shown that high value in the peel force and the peel energy, the peel force was $0.38*10^3$ N/m and the peel energy was $0.605*10^3$ N/m, peeling the film from Al surface leaves a residual of the adhesive material on both adherend, the failure for this specimen were combination of adhesive and cohesive failure. For the specimens that were immersed in water for 12 days, the results had shown that the effect of water on the specimen (Al /PE), where the water diffusion coefficient on the adhesive joint was $1.09*10^{-14}$ m^2/s and the peel force was $0.062*10^3$ N/m and the peel energy was $0.124*10^3$ N/m, PE film was peeled from Al surface without any resistance.

Key words: water absorption, peel force

Introduction:

Adhesive bonding technology has been widely applied in modern structures in industries such as automotive, aerospace and in microelectronic device. The core idea of this technology is to bond two similar or dissimilar structure members with a thin continuous interface layer which can provide far better stress transfer across the interface than those traditional point-wise joining technologies such as spot welding, riveting and bolting. [1]. For elastic adhesive joints, in which the deformation of the adherends is dominated by elasticity during the fracture process, the conditions for fracture can be predicted using linear-elastic fracture mechanics (L.E.F.M) [2]. To evaluate the deformation and fracture energy, tensile stress-strain curve and peel tests were carried out. In the tensile stress-strain curve the specimen is elongated until it breaks. The stress is recorded as a function of extension. Energy is absorbed within the sample by viscoelastic deformation of the extension (deformation of the polymer chains), and finally by the creation of new surface areas [3]. In the peel test, the practical strength of an adhesive bond, termed the critical energy release rate during debonding G_c, is orders of magnitude higher than the thermodynamic work of adhesion, of the interface, because of energy dissipation associated with the bulk deformations of the adhesive and the adherends that occur during the debonding process in a peel test Energy Release Rate for elastic membranes [4, 5]:

$$G_c = \frac{F_c}{b}(1-\cos\theta) - W_\lambda h \qquad (1)$$

Where

F_c is the load required for peeling the membrane from a rigid substrate (N);

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b is the width for membrane (mm);
θ is the peel angle;
W_λ is the strain energy density for the membrane (N/mm^2).
h is the thickness for the membrane (mm).

The first term represents the peel energy, which will take to be the energy (per unit area of peeled substrate surface) dissipated in the broad region of the peel front. The second was the work done in stretching the freed strip, by the force F, which will be the stain- energy density W_λ , and can be evaluated from the work done in an appropriate tensile test of the material of the freed strip [6].

Materials and Methods

Employed Materials
Aluminum (Al) was used as a substrate, and polyethylene (PE) was used as a film adhered to the Aluminum (Al) surface by using cyanoacrylate adhesive supplied by Quickstar Company.

Samples Cutting
The dimensions of the samples were cut accordance with the related international specification; ASTM standards (D903-1983) .Aluminum (Al) was cut to the length of 203.2 mm, width 25.4 mm and thickness 2 mm. The flexible adherend (PE film) shall be capable of being to bend through an angle of 180° when the film fixed to the Instron machine. PE film shall be not less than 304.8 mm in length; its width shall be 25.4 mm with thickness 0.09 mm.

Surface Preparation of Adherends
In order to obtain clean metal surface, it was necessary to remove any contaminated layer such as cutting work pieces, greasing and oxides. The metal was first edge freed burrs, then followed by mechanically polished process through (60) grade metallographic paper and then (1000) grade metallographic paper. Wiped the metal with soft cloth immersed in acetone after polishing.
PE films were cleaned with soft cloth immersed in acetone in order to remove any debris or grease from the surface.

Surface Roughness Test
Surface roughness test was used to determine the surface roughness for Aluminum surface, using the surface roughness tester (model TR200). The mean value of the surface roughness (R_a) for Al surface is 0.665 µm.

Adhesion Operation
The Cyanoacrylate adhesive was applied over the metal surface (Al) along the length of 152.4 mm. Surface joint were created with cyanoacrylate commercial one part adhesive, then PE film was put on the surface of the metal. The specimens must left for 24 hours before testing.

Tests for Water Absorption
Dry specimen (Al/PE) was weighted and denoted as the initial mass (M_1). Then we put dried specimen in large glass beaker full with water at room temperature. During the sorption tests, the specimen was periodically weighted each 24 hours and denoted as (M_2) for 12 days. Before weighting the specimen we must dry the excess water. The specimen's mass was measured using electronic analytical balance with an (accuracy 0.01 gm). After 12 days, the specimen was taken out and calculated the weight gain by using equation (2) and then tested with an instrument of peeling test, Instron (model 1122).

$$W_t.\% = \frac{M_2 - M_1}{M_1} \times 100.\% ----(2)$$

M_2: The specimens mass's after immersion in water at time t, (gm)

M_1: The specimens mass's before immersion in water, (gm).

Mechanical Tests

Tensile and peeling tests were performed at room temperature 25±20C of relative humidity 50±5%. For Tensile and peeling test, Instron universal testing machine (model 1122) of 5 KN, full scale load capacity was used. The test speed was fixed in all tests to 100 mm/min. The data was recorded on Instron chart, which its speed was fixed to 100 mm/min.

Tensile Test

This test was carried to find the deformation for PE film. PE film was positioned vertically with the aid of the grips of the testing machine, which was tightened evenly and firmly to prevent any slipping during the tension procedure. The load value (force) and the rate speed of crosshead (100 mm/min) were plotted to the length of (100 mm) of PE film.

Peeling Test

This test was carried to assess the strength of adhesion bonding between PE film and Al interface. A specimen was prepared, and marked the length of PE film which will be tested by 100 mm. Bend back the unbonded end of PE film and clamp in the fixed grip, and the rigid adherend (Al) fixed in the other grip, taking care that the test specimen is accurately positioned between the grips, as shown in figure 1 . Set the crosshead speed for Instron machine at 100 mm/min. Adjust the chart drive at a constant speed (100 mm/min). Strip the separating member from the specimen at angle of 180^0.

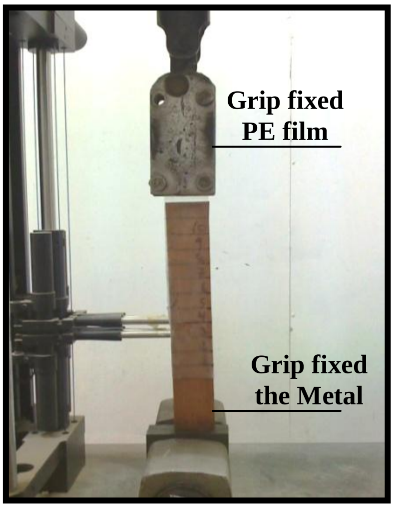

Fig (1): The specimen fixed between the grips

Results and discussion :

Stress – Strain Curve for PE Film

The stress-strain curve for PE film was shown at figure (2), for 100 mm in length. The curve almost predicts two region, elastic and plastic region. Initially, Hookean elastic behavior was observed until the specimen draws, and permanent deformation occurs, as the specimen draws, PE film chains were reorientation and recrystallization will form stronger material until the specimen was tearing. According to the experimental work, the specimen didn’t tear, because of high elongation for PE film.

The value for strain-energy density W_λ was extracted from the stress – strain curve, which was an important parameter to evaluate the values for critical energy release rate via Equation (1).

Strain-energy density W_λ is given by the area under a stress-strain curve [7]. The area under the curve was found by using trapezoidal method. The value for strain-energy density is found to be $W_\lambda=17.28\times10^6$ N/m^2 for the length of 100 mm, and W_λ is expressed per unit volume. It has to be multiplied by the thickness of peeled strip.

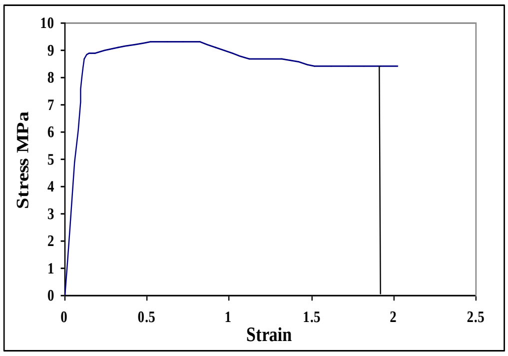

Fig (2): Strain – energy density was given by the area under a stress – strain curve

Peel Test

Peel test is used to characterize the adhesion of film on substrate. Figure (3), shows a typical set of load-peel distance curve for the specimen Al /PE. In this figure, PE films exhibit two distinct behaviors, stretch and peel. The works done on the specimen was the sum of two terms. The first term was the peel energy, which will be taken to be the energy per unit area of the peeled substrate surface. The second term was the work done in stretching the freed strip, which will be the strain-energy density W_λ.

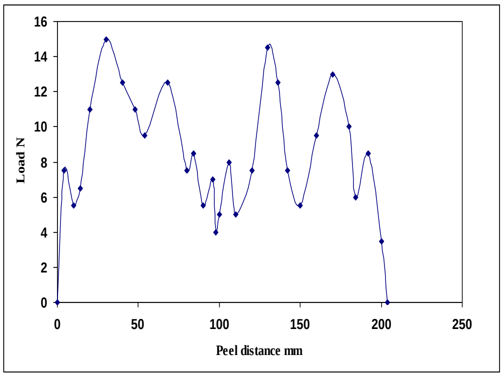

Fig (3): Load-peel distanced curve for Al / PE. Crosshead speed 100 mm/min

There were variations in the applied load, which means during the test, when the film was stretched, the load would be applied on the contact area, which lies between the film and substrate, and the load will be increased until cracks between the film and substrate will propagated, so that the load will be decreased to minimum value.

To evaluate the peel force (P_f) and the critical energy release rate (G_c) for the dry adherend (Al/PE), where the peel force (F_c/b) was taken as the load per unit width during peeling process, and the critical energy release rate (G_c) was taken by using equation (1). From figure (3), the highest average peel force attainable at the onset and over the entire peel length was 0.38 $*10^3$ N/m, and the average value for critical energy release rate (G_c) was 0.605 $*10^3$ N/m. From macroscopic view, the mode of failure for Al/PE were combination of cohesive and adhesive failure, so that some of adhesive material remains on Al surface and, the other of adhesive material pull out with PE film, that means, the adhesive material would diffused into PE film and make with the film strong bond [8]. Figure (4), shows some of the adhesive materials lies on Al surface.

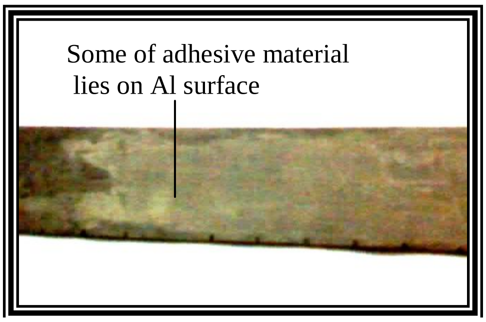

Fig (4): Some of adhesive material lies on Al surface (Adhesion failure) and, the other of adhesive material pull out with PE film (Cohesion failure)

Effect of Water Absorption on Specimen :

Water Diffusion Properties of Adhesive Joint

The mechanism and measurement of water diffusion, is very important to characterize the moisture transport in adhesive joint. Measurement of diffusion, obeys Fick's laws, where weight of water absorbed is increased linear with the square root time gradually and slowly to saturation state [9].

The moisture diffusion coefficient may be determined readily by exposing the dry specimen (after recording its dry weight) to a constant moisture environment, plotting its weight gain versus $t^{1/2}$ [10]. The diffusion coefficient is then calculated by using equation (3):

$$D = \pi\left(\frac{kh}{4m_s}\right)^2 \text{---------} (3)$$

Where

D = the diffusion coefficient, m^2/s

k = slope of linear portion of the plot of weight gain versus $t^{½}$.

h = the thickness of PE film and adhesive material, m

ms = the equilibrium moisture content taken at saturation.

Table (1): Water absorption data for Al/PE

Time days	***(Time)$^{0.5}$ (Hour)$^{0.5}$***	***weight Gain %***
1	4.89	0.12
2	6.92	0.181
3	8.48	0.301
4	9.79	0.482
7	12.96	0.603
8	13.85	0.603
9	14.69	0.663
10	15.49	0.663
11	16.24	0.663

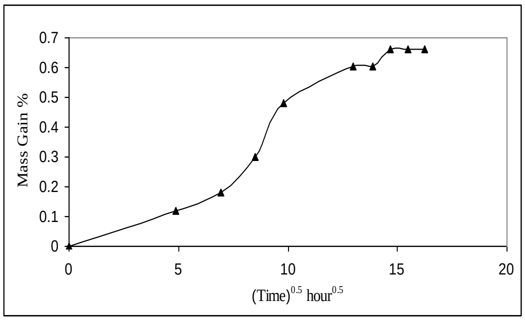

Fig (5): Relation between weight gain and square root time for the wet Al/PE

From equation (3) the diffusion coefficient for Al / PE is 1.09 *10 $^{-14}$ m^2/s.Weight gains for this specimen at the equilibrium moisture content taken at saturation was more than 0.6 %.

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Figure (6), shows a typical set of load – peel distance curve for specimen absorbed water (Al/ PE).

In figure (6), PE film exhibit one behavior in this test, peeling only, because the film was pull out fast without any resistance, that means, all of the force would be changed to energy for peel process.

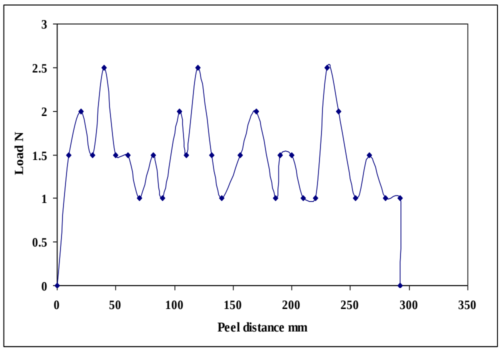

Fig (6): Load – peel distanced curve for wet Al/PE. Crosshead speed (100 mm/min)

From figure (6), the highest average peel force (P_f) attainable at the onset and over the entire peel distance was $0.062*10^3$ N/m, and the critical energy release rate (G_c) was taken by using the first term in equation (1) because the film was pull out fast without any stretching, the average value for critical energy release rate (G_c) was $0.124 *10^3$ N/m.

The energy for peeling was decreased, when the specimen absorbed water. To explain this result, the water diffused in the interface (adhesive region) between PE film and the metal. So that, local stresses would form at the film of PE, because of the swelling process would occurred at adhesive material, make decreased the adhesion force. The cracks would propagate between PE film and metal. Figure (7), a schematic view of the water entered the adhesive joint. Beside that, when failure occurs, it is the weak boundary layer that fails, although failure seems to occur at the adhesive – adherend interface. Polyethylene and metal oxides are examples of this effect.

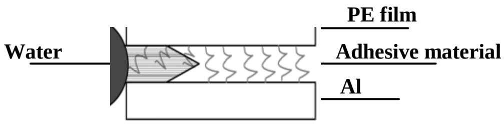

Fig (7): Schematic view of the water entered the adhesive joint

According to Comyn [11], water having entered the adhesive joint may cause strength loss by one or a combination of the following mechanisms:
a) Altering the properties of the adhesive in a reversible manner (e.g., plasticization) or an irreversible manner (e.g., hydrolysis, cracking, or crazing);
b) Attacking the adhesive/adherend interface either by displacing the adhesive or by hydrating the metal or the metal oxide surface of the adherend;
c) Inducing swelling stresses in adhesive joints.

Conclusions:

The main conclusions of this research can be summarized as follows:
1. For the adhesion of the specimen in the dry condition, it has been found that:
The factor that affected on the peel force and the critical energy release rate were attributed to the surface roughness for the metal.
2. For the adhesion of the specimen in the wet condition, it has been found that:
The value of the peel force and the critical energy release rate were decreased and was attributed to the local swelling due to the water absorption for Al/PE.

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تأثير امتصاص الماء على قوة التلاصق

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الخلاصة:
تم دراسة قوة التلاصق بين غشاء البولي اثيلين ومعدن الالومنيوم باستخدام مادة اللاصق (سيانواكريليت). وقد استخدم الألمنيوم كقاعدة أساسية , كما استخدم البولي اثيلين كغشاء يلصق على سطح المعدن.هيأت عينات قياسية لاستخدامها في فحص السلخ في الحالة الجافة وعينات اخرى غمرت في الماء لمدة 12 يوما بدرجة حرارة الغرفة. أظهرت النتائج القيمة العالية لقوة وطاقة السلخ للعينات الجافة حيث كـــانت قوة السلخ $0.38*10^3$ N/m وطاقة السلخ $0.605*10^3$ N/m .وعند سلخ غشاء البولي اثيلين من سطح الالومنيوم كانت هناك بقايا من مادة اللاصق على السطحين حيث فشل العينة هو فشل مزدوج (فشل التلاصق والتماسك). اما بالنسبة للعينات المغمورة بالماء لمدة 12 يوما اظهرت النتائج تأثير الماء على العينات حيث كان معامل انتشار الماء عند منطقة التلاصق $1.09*10^{-14}$ m^2/s اما قوة السلخ فكانت $0.062*10^3$ N/m وطاقة السلخ كانت $0.124*10^3$ N/m وسلخ الغشاء من سطح المعدن كان بدون أي مقاومة.