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Multiple Cracking of Metallized Polyimide Kapton Films

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Abstract

Mechanical behaviour of metallized polyimide Kapton films was studied. The adhesion of metallized films to Kapton films is approximately hundred-folds higher than that of coatings made by usual sputter deposition of a metal on a polymer substrate. Multiple cracking of a metallized film was observed. Cracks of two different scale levels are observed. The cracks of the first type (microcracks) are short with 1-2 μm length. The cracks of the second type are much longer and have length of several hundreds microns. The cracks of this type appear at higher strains as compared with the shorter ones. In thinner 7-μm-thick samples long cracks were not observed.

Introduction

Polymers coated with a thin, nanometers in thickness, rigid film are used for medical and packaging applications. If a comparatively brittle material is used for coating, at elongation it breaks on a number of long tapes oriented perpendicularly to the stretching axis. The width of these tapes (fragments) reduces with an increase in strain. Fragmentation of a coating was studied for Ni/polyethyleneterephtalate (PET) [1], quartz/PET [2,3], Au/PET, Pt/PET and Pt/rubber [4,5] composites.

Wheeler and Osaki described three stages of the coating fragmentation in Ni/PET composite [1]. The first stage, random cracking of the coating, began when the applied strain was sufficient to produce cracks in the coating film. On this stage of fragmentation, the coating cracked at random locations along the sample length. Eventually, the film fragments began to divide in their midpoints. Finally, debonding of the largest fragments was observed.

The width of fragments was used to estimate adhesion strength between the coating and the substrate [1-3,6] using a modified Kelly-Tyson equation [7]. To characterize adhesion of the coating, Bazhenov with co-workers have developed an alternative method [8,9] based on an energetical analysis. With this method adhesion is described by fracture toughness, *i.e.* energy dissipated to debond the coating. Debonding behavior at stretching depends on the thickness of a coating. A thin coating did not debond from the substrate. However, if the thickness of the coating is higher than a critical value, it debonds at comparatively low strain as described by Wheeler and Osaki [1]. For Pt/PET the critical coating thickness is ≈ 30 nm. The critical thickness was used to determine the mode II fracture (debonding) toughness G_{IIc} [8].

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Experimental

Test pieces were cut from metallized polyimide Kapton films of different thickness. Samples, 5 mm in width and 20 mm in length, were elongated with a testing machine, allowing to see a stretching of a sample under the microscope. A metallized surface of a sample was regularly photographed during elongation and a digital image of a sample was displayed on a monitor. Tensile stress was determined by a division of a tensile force on a cross-sectional area of a sample.

Results

Figure 1 shows a typical stress-strain curve for a metallized film. The diagram is typical for plastic materials. Initially the material is deformed elastically, and at strains higher than 5% the yielding of the material begins. Fracture deformation of samples was 30 to 70% depending on a substrate. The thicker was the substrate the higher was the failure elongation.

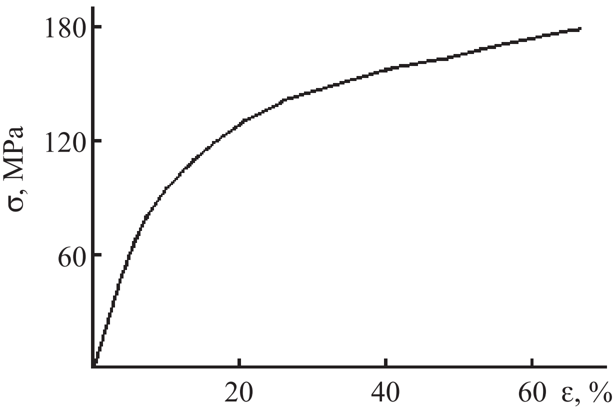

Fig. 1. Typical stress-strain curve for a metallized Ag-Co/Kapton film. The film thickness is 50 microns.

Optical microscopy

Figure 2 shows an optical micrograph of a metallized Ag-Co surface of a 50 μm-thick Kapton substrate after tensile failure. A number of rather long cracks in the metal coating are observed. Obviously, this means that fracture strain of a metal coating is lower than that of a Kapton substrate. Near the cracks a number of small bubbles are observed. Evidently, the bubbles are caused by the onset of a local debonding of the coating.

The surface microcracks in samples based on 25 μm-thick Kapton substrate were observed, while debonding of the coating film was not noticed. In samples based on 8 micron-thick Kapton film up to failure neither cracks nor debonding of the coating was observed.

Figure 3 shows linear density of the cracks in the coating plotted against the tensile strain. The onset of a coating cracking is observed at lower strains in samples with a thicker substrate. For the material based on the 75 μm-thick substrate cracking of the coating starts at approximately 10%, while with the 7 μm-thick substrate any cracks with an optical microscope up to failure strain of 30% was not observed.

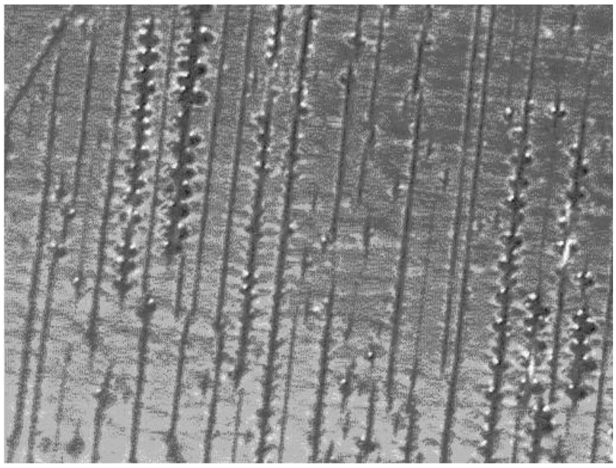

Fig. 2. An optical micrograph of Ag-Co metallized surface of a 50 micron-thick Kapton substrate after tensile failure.

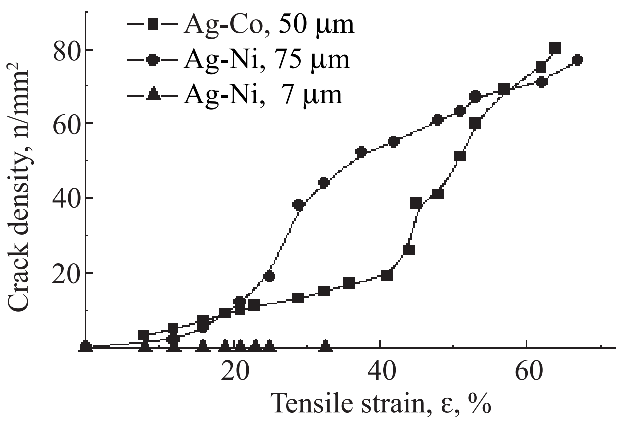

Fig. 3. Linear density of the cracks plotted against tensile strain.

Study of the metallized surface of Kapton in optical microscope allows making two preliminary conclusions:

1. Cracking of the coating in samples with thicker substrates is initiated at lower strains;
2. Cracking of the coating is followed by its local debonding near the cracks.

Scanning electron microscopy

Figure 4 shows an SEM image of cracks in a coating film after tensile failure at 60% strain. Long cracks from 20 to 200 μm in length are oriented perpendicularly to the elongation direction. These cracks are analogous to that observed in an optical microscope (Fig. 2). In addition, in the scanning electron microscope the cracks of the second type approximately hundred-folds shorter than that shown in Fig. 4 are observed. These cracks (Fig. 5) are 1-2 μm in length and are curved while the longer cracks are rather straight. A similar short cracks were found in samples based on 7 μm-thick substrate where with an optical microscope no cracks are observed. Thus, there are cracks of two different scale levels. The cracks of the first type (microcracks) are short and curved. They appear in all studied materials. The cracks of the second type (mesocracks) are rather straight and much longer, several hundred of microns in length. The cracks of this type appear at higher strains as compared with the shorter ones. In the 7-μm-thick samples they were not observed.

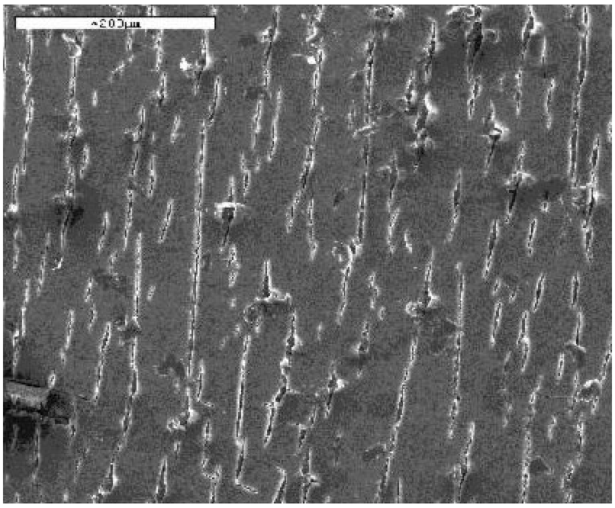

Fig. 4. SEM image of cracks in an Ag-Ni metallized coating of a 75 micron-thick Kapton after tensile failure.

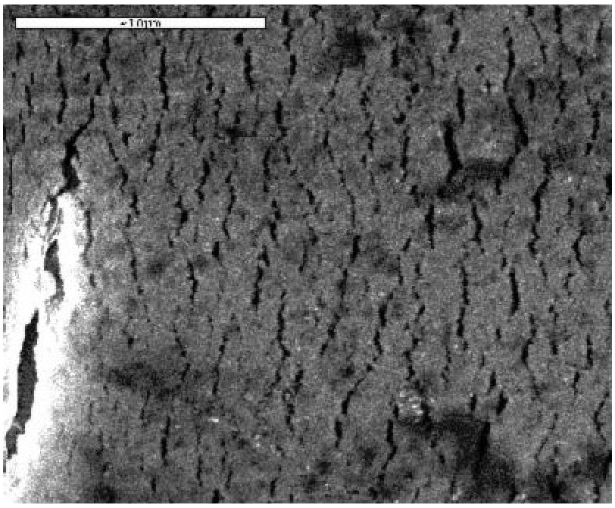

Fig. 5. Higher magnification SEM image of cracks in an Ag-Ni metallized 75 micron-thick Kapton.

Discussion

Further analysis shows that adhesion of a metallized coating to the Kapton film is extremely high. Accoding to [8], the debonding of a coating film is characterized by a critical thickness value. A coating debonds from the substrate if its thickness exceeds the critical magnitude. In contrast, thin coatings do not debond from the substrate. For the usual method of metal sputter deposition the critical thickness of a coating for platinum/PET composite is 30 nm [8]. The thickness of a coating film in the 75 μm-thick samples was 5 to 8 microns. And only in this composite an onset of debonding process was noticed. In other composites no debonding at strains up to 60% has been observed. This means that adhesion of a metal film to Kapton film is extremely high.

Below the debonding of a coating is analyzed. Figure 6 shows a schematic drawing of a coating debonding from a substrate. We assume that the crack grows along the surface between the coating and the substrate. The following derivation is based on an energy balance analysis similar to that used by Griffith. Elastic energy released by bebonding out coating is given by [9]:

$$dU_{+} = \frac{\sigma^2}{2E}\, dV = \frac{\sigma^2}{2E}\, wh\, dL \qquad (1)$$

where $\sigma^2/2E$ is the density of elastic energy in the coating, σ is the tensile stress in the coating, E is Young's modulus of the coating, dV is the increment in volume of the debonding coating, dL is an increment in the debonding length, h is the thickness of the coating and w is the width of the coating. The energy spent to debond the coating from the substrate is:

$$dU_{-} = G\, w\, dL \qquad (2)$$

where G is (mixed mode) fracture toughness of debonding. The coating debonds from the substrate if the released elastic energy dU_{+} is higher than the dissipated energy dU_{-}:

$$\sigma > \sqrt{\frac{2GE}{h}} \qquad (3)$$

Equation (3) is similar to the Griffith criterion,

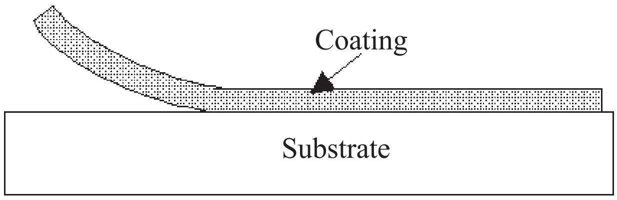

Fig. 6. Schematic drawing of a coating debonding from a substrate.

and the thickness of debonding coating in equation (3) is analogous to the crack length in the Griffith criterion. The debonding process is caused by a 90°-turn of a crack in the coating and its growth along the substrate/coating surface. According to equation (3), a coating debonds if the stress in it exceeds some critical value. However, in ductile metals this value cannot be higher than the yield stress of the coating. The critical value decreases with an increase in the coating thickness. This equation explains why the thin films do not debond from the substrate. The debonding fracture toughness is found from eq. (3):

$$G = \frac{\sigma^2 h^*}{2E} \quad (4)$$

where σ is the tensile stress in the coating and h^* is the critical coating thickness. We do not know the magnitude of the stress in the coating. However, we can roughly estimate it. The lower estimation of the stress is 150 MPa, the yield stress of the substrate. The yield stress of Ag and Ni is 140-180 and 400 MPa respectively. The Young's modulus of Ag is 70 GPa. Substituting σ = 160 MPa, E = 70 GPa and h^*=7 μm, we get $G \approx 1.3$ J/m^2. This value is approximately hundred-folds higher than the debonding value for metal films made by usual sputter deposition.

Conclusions

The adhesion of metallized films to Kapton films is much higher than that of coatings made by usual sputter deposition of a metal on a polymer substrate.

Multiple cracking of a metallized film was observed. Cracks of two different scale levels are observed. The cracks of the first type (microcracks) are short, 1-2 microns in length. The cracks of the second type are much longer, several hundred micron in length. The cracks of this type appear at higher strains as compared with the shorter ones. In thinner 7-μm-thick samples long cracks were not observed.

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