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Models for saturation damage state and interfacial shear strengths in multilayer coatings

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ABSTRACT

The present work investigates the saturation damage state of a two-layer coating on a substrate (layer 1/layer 2/substrate) under uniaxial tensile loading in order to derive expressions for the interfacial strength between layer 1 and layer 2, and between layer 2 and substrate. It is based on experimental data on specimens where layer 1 is an inorganic film, layer 2 is an organic coating and the substrate is a polymer. The analysis is relevant to the cases where layer 1 cracks first, followed by layer 2, in which cracks appear due to stress concentrations caused by the cracks in layer 1. It considers the cases where at least one interface is completely yielded with shear stress equal to the interfacial shear stress, and where the crack density in layer 1 is equal to or higher than the crack density in layer 2. The possible situations depend on the relative shear strengths between layers 1 and 2 and between layer 2 and the substrate. The interfacial shear strength between layer 1 and layer 2, and between layer 2 and substrate are derived for elastic and yielded stress transfer cases and found to frame experimental values obtained with single-layer coatings.

1. Introduction

Multilayer coatings on polymer substrates are developed for an increasing number of applications ranging from optical systems to flexible electronic devices and photovoltaic modules (Chalamala et al., 2004; Crawford, 2005). A representative case of such multilayer films consists of an inorganic layer (*e.g.*, oxide or nitride diffusion barrier) and an organic layer (*e.g.*, acrylate-based planarization coating, so-called hard-coat, HC) deposited on a polymer substrate (Leterrier et al., 2004). Hard-coats have been developed to buffer the influence of the polymer substrates on the generation of defects during deposition of the inorganic coating, resulting in improved surface quality and increased stiffness compared to the substrate alone (Shu et al., 2007). The functional and mechanical performance of the multilayer is controlled by the cohesive properties of each individual layer and the interfacial adhesion of adjacent layers.

The fragmentation test method, in which coating cracking is analyzed as a function of tensile strain has been extensively used to obtain these properties (Leterrier, 2003). The accuracy of this method is primarily related to the absence of third body interactions, such as indenter-coating friction in case of scratch and indentation tests, or adherent-coating traction in case of peel and pull-out tests. The topic of multiple cracking of coatings on high elongation substrates (and matrices in composite laminates) has motivated a considerable amount of work, for instance to obtain statistical strength parameters from crack spacing distributions (Hui et al., 1999; Leterrier et al., 1997a,b; Ochiai et al., 2007) and layer toughness (Kim and Nairn, 2000; Nairn, 2000). Prior analyses of experimental data, however, are limited to single coatings (Andersons et al., 2007; Handge, 2002; Handge et al., 2000; Howells et al., 2008; Hsueh and Yanaka, 2003; Leterrier et al., 2004; Tang et al., 2001; Yanaka et al., 1999). In this case, the interfacial shear strength (IFSS, representative of

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practical adhesion) can be derived from an interfacial stress transfer analysis. Solutions for elastic stress transfer at interfaces in multiphase materials were derived using shear-lag (Cox, 1952; Mendels et al., 1999; Nairn, 1997) and variational methods (Hashin and Shtrikman, 1963; Nairn, 1992). The case of yielded interfaces was considered since in practice the interfacial shear stress may reach the yield limit (Kelly and Tyson, 1965). Models for partially yielded interfaces were also derived to account for the elasto-plastic behavior of the matrix in the case of composites (Piggott, 1980) and of the substrate in the case of multilayers (McGuigan et al., 2003). For yielded and partially yielded interfaces IFSS is proportional to the density of tensile cracks at saturation of the fragmentation process (CD_{sat}, *i.e.*, when no more cracks appear as the tensile strain is increased), related to the so-called critical stress transfer length (Leterrier et al., 1997b).

For multilayer coatings the failure of adjacent layers is coupled (cracking of one of the layers influences cracking of the other layer and vice-versa), which invalidates prior stress transfer theories used to derive the IFSS. This paper considers two-layer coatings. In this case, when the first layer starts cracking, stress concentrations are induced in the second layer, thus lowering its strain to failure. Cracking of the second layer relaxes the axial stress in the first layer, which prevents further cracking of the first layer. In other words the stress partitions between the two layers, which changes the critical length to achieve failure stress in one or both layers. The problem is complicated due to the uncertainty about the stress state at the interface between the two layers.

The objective of this paper is to solve the stress transfer problem for a two-layer coating on a substrate under uniaxial tensile loading (in the order layer 1/layer 2/substrate) for specimens where layer 1 cracks first, and enable determination of the IFSS between layer 1 and layer 2, and between layer 2 and the polymer substrate. The theoretical derivation is applied to several inorganic layer/HC/polymer films representative of multilayer structures developed as substrates for flexible electronic devices.

2. Materials and experimental methods

2.1. Materials

Two main types of multilayer films were analyzed, including single-layer (layer 1/substrate) and two-layer coatings (layer 1/layer 2/substrate, which in some cases had the same layer 2 on the opposite side of the substrate). In the first type, layer 1 was a silicon nitride film (SIN) with three different thickness (300, 400 and 500 nm), layer 2 was a silica-acrylate hybrid hard-coat (HC, thickness 2 μm) and the substrate was a 100 μm thick, high temperature aromatic polyester film (ARY, Arylite, Ferrania Technologies), also coated on the opposite side with the same 2 μm thick HC layer. The top HC, on the inorganic film side, is labeled here as ‘HC1’, and the bottom HC, located on the other side of the substrate, is labeled ‘HC2’. The HC2 layer with same thickness and same thermal history as the HC1 layer will serve as a reference to quantify the influence of SIN on HC cracking, and to obtain the IFSS between HC and the substrate. In the second type of multilayer film, layer 1 was a 209 nm thick oxide film (OXI), layer 2 was a HC layer with four different thicknesses (1, 2, 3, 4.5 μm) and the substrate was a 125 μm thick polyethylene terephthalate film (PET, U34, Toray Company). HC/PET films without the OXI layer and with the same HC thicknesses as in the two-layer coatings were also produced as references. The elastic properties of the layers relevant for the analysis developed in the following are reported in Table 1. The Young’s modulus values were obtained using nanoindentation tests of the layers deposited on glass substrates and the Poisson’s ratio values were estimated. All investigated multilayer structures are listed in Table 2.

2.2. The fragmentation test

In the fragmentation test, the evolution of crack patterns in the brittle coating is monitored as a function of the uniaxial tensile load applied to the substrate (Leterrier et al., 1997b). Rectangular samples with gauge length 40 mm and width 10 mm were carefully cut from the foils using a razor blade. Tests were carried out at a nominal strain rate of $4.2 \times 10^{-4}\ s^{-1}$ in situ in an optical microscope (Olympus BX60), using a computer-controlled tensile frame equipped with contactless video extensometry to overcome compliance effects. The coating strain at failure was measured with accuracy better than 10^{-3}. The progressive cracking of the coating was analyzed in terms of crack density, equal to the number of tensile cracks per unit length multiplied by substrate elongation to correct, to a first approximation, for crack opening. As will be shown later, the contrast was high enough to discriminate cracks in layer 1 from cracks in layer 2. Two samples of each type of film were analyzed.

3. Experimental results

Fig. 1 shows the damage state in SIN and SIN/HC coatings on ARY and in OXI and OXI/HC coatings on PET at approximately 3% strain. In all cases tensile cracks perpendicular to the applied load are evident. The density of tensile cracks in the SIN and OXI layers is much higher when there is no HC layer. Cracks in the HC layer are also visible and were easily discriminated from cracks in the top layer 1 thanks to large differences in contrast, the HC cracks being much darker under reflected illumination mode. Interestingly, the HC layer systematically cracked at cracks previously formed in the SIN or OXI layers, due to stress concentrations at the tip of the crack at the SIN/HC or OXI/HC interface. This is evident in Fig. 2, which displays

Table 1
Elastic properties of layers.

Layer	Young’s modulus [GPa]	Poisson’s ratio
SIN	100	0.2
OXI	130	0.15
HC	6	0.35