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Debonding of particles in thin films

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ABSTRACT

Thin composite films consisting of a matrix with embedded particles are currently being developed both as hard, wear resistant coatings and as functional surfaces. The effect of stiff particles in the film are studied for systems where the film is under residual tensile stresses. The particles, when they are fully bonded to the matrix, increase the stiffness of the composite film. In cases where the particles debond from the matrix material, the stiffness of the composite film decreases. The conditions under which the debonding process is stable are studied. For systems properly designed, a controlled debonding process of the particles can thus be used to reduce the stress levels in composite film lowering the risk for delamination of the composite film from the substrate as well as the risk of through cracks in the film. The work includes finite element based unit cell calculations of interface debonding between spherical particles and the film, and the release of residual stresses following this. The three dimensional unit cell calculations assume a periodic distribution of particles in the plane parallel to the substrate interface with equi-biaxial tension and periodicity with zero overall stress perpendicular to the substrate interface.

1. Introduction

Thin films containing particles are used in a variety of applications including hard coatings with embedded nano-particles and paint systems with micro glass beads (Gunnarsson and Gudmundsson, 2007; Kibsgaard, 2010). These particles may influence the stiffness and fracture toughness of the film. In addition to changing the mechanical properties of the film, the particles may change the physical properties e.g., by applying coatings to the particles which is the case for the paint system in Gunnarsson and Gudmundsson (2007) and Kibsgaard (2010) where the glass beads have been coated by TiO_2 to make the paint system chemically active, by utilising the photo-catolytic properties of anatase.

Thin film fracture modes include tensile delamination at the film-substrate interface (Jensen et al., 1990) and compressive delamination modes driven by film buckling (Hutchinson and Suo, 1992; Moon et al., 2002). Other types of delamination failure modes include the initiation, propagation and buckling of edge cracks and corner cracks (Jensen and Thouless, 1995; Veluri and Jensen, 2013). For failure by interface delamination between the film and the substrate, the formulation of proper, mixed mode interface fracture criteria is essential (Hutchinson and Suo, 1992). Thin film fracture modes further include tensile film cracking such as channelling cracks (Thouless, 1990; Beuth, 1992). For a review of different failure modes in layered materials and methods for analysing the competition between these modes, see Hutchinson and Suo (1992). The present work deals with tensile fracture modes in the form of particle-film debonding.

The purpose of this paper is to describe the effects of debonding of particles in a film which is composed of an elastic matrix containing micro glass beads. Such particles, both hollow and solid, are used in various matrices such as polypropylene, epoxy, alkyd and concrete. The particles might be manufactured under controlled conditions or obtained from the burning of coal in the form of fly ash. Debonding of nano-particles in bulk materials was studied recently in Salviato et al. (2013) for hydrostatic tension.

When particles are introduced in the thin films, the resulting composite film may experience increased resistance to crack growth as well as an overall increase in Young's modulus. Many explanations have been given for the increase in fracture resistance such as crack front pinning, particle cracking and inter-particle cracking (Garg and Mai, 1988). These studies are also focussed on bulk material behaviour. This paper shows that localised debonding in the film-glass particle interface can be a vital contributor to the overall crack resistance of composite thin films, by lowering the overall stress levels in the cases where the films remain bonded to the substrate.

For the system considered, it is important that residual strains arising during deposition of the film do not lead to through-thickness cracks thus exposing the substrate. This can be achieved by ensuring that the fracture toughness of the film is sufficiently

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high that micro-cracks in the film will not grow. One way of avoiding exposure of the substrate in cases of cracks developing in the film has been to introduce a thin, soft interlayer between the film and the substrate acting as a crack arrestor (Suresh et al., 1993). Another, safe way of ensuring that micro-cracks do not propagate through the film is to release the residual stresses by controlled particle debonding and possibly also a limited amount of inter-particle cracking as will be shown in the present paper.

A SEM of a paint system motivating the present study is shown in Fig. 1 (Kibsgaard, 2010; Gunnarsson and Gudmundsson, 2007). The TiO_2-coated glass particles at the film surface are seen, as well as some limited amount of trans-particle cracking of the alkyd matrix. The mechanism focussed on is the effects on the stress state and overall mechanical properties of the particle reinforced film of debonding between the particles and the film for cases where the film remains bonded to the substrate.

Micro glass particles have been introduced to other bulk materials with increased fracture toughness and Young's modulus as a result (Green et al., 1979; Moloney and Kausch, 1983; Moloney et al., 1987; Mallick and Broutman, 1975; Spanoudakis and Young, 1984; Amdouni et al., 1992; Dibenedetto and Wambach, 1972). Different explanations have been suggested for the increased fracture toughness of these particle reinforced composites such as: trans-particle fracture, crack tip blunting, debonding of particles, crack deflection around particles, crack front pinning and more (Garg and Mai, 1988). In the present paper, a detailed study of the particle debonding mechanism in thin films is analysed by letting a crack grow in the interface between the glass particles and the film.

2. Homogeneous films, tensile fracture

In thin film systems, residual stresses may arise at manufacturing due to curing and thermal expansion mismatch between film, particles and substrate, but also external loads may induce stresses in the film causing fracture.

In this Section, the notion *delamination* is used for interface fracture at the film substrate interface while in the remaining Sections of the paper; the notion *debonding* is used for interface fracture at the particle/film interface.

Films in residual compression typically fail by buckling driven delamination caused by delamination at the film/substrate interface, see e.g., Hutchinson and Suo (1992) for circular and straight-sided delamination, and Jensen (1993) and Moon et al. (2002) for the more commonly observed "telephone cord" delamination mode. The fracture modes involve the interaction of fracture at the film/substrate interface and buckling due to compression of the film in the delaminated region.

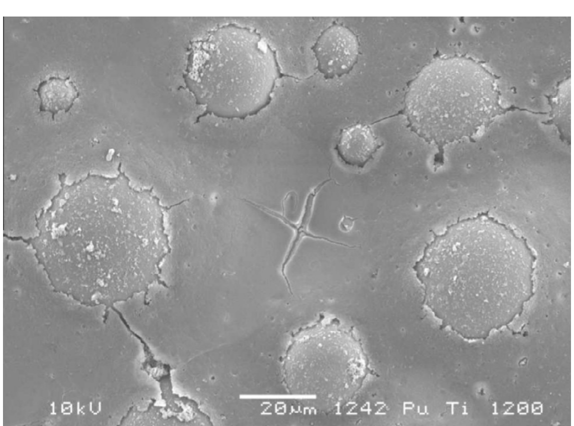

Fig. 1. A SEM picture of film with TiO_2-coated glass particles indicating debonding between particles and film as well as some limited amount of trans-particle cracking.

The focus here is on films which are in residual tension. For sufficiently high interface fracture toughness the prevailing fracture mode of such systems is crack propagation by channelling. The requirements for the interface fracture toughness compared to the fracture toughness of the film for channelling to be more likely than interface delamination have been studied in He and Hutchinson (1989). Roughly speaking the film will fail by channelling rather than delamination at the film/substrate interface if the fracture toughness of the film, G_c, is less than half the mode adjusted interface fracture toughness (He and Hutchinson, 1989). The films in Kibsgaard (2010) in general adhere well and delamination at the film/substrate interface is not observed.

The residual stresses, σ_0, in the film due to a curing strain, ϵ_0, is in-plane and equi-biaxial under the assumption that the film and substrate are linear elastic and isotropic, and is given by

$$\sigma_0 = \frac{E_f}{1 - \nu_f}\epsilon_0 \quad (1)$$

while other stress components are zero. In the following the film is assumed linear elastic and isotropic with a Young's moduli, E_f, and Poisson's ratio, ν_f.

Conditions for crack channelling in thin homogeneous films were studied in Thouless (1990) and Beuth (1992) for cases of single and multiple channelling cracks and for cases of elastic mismatch between the film and the substrate. For the case of no elastic mismatch, channelling is possible for (Thouless, 1990)

$$G_c < c\epsilon_0^2 h \frac{E_f(1+\nu_f)}{1-\nu_f} \quad (2)$$

where h is the thickness of the film and $c \cong 2$. If the residual strain level exceeds the minimum requirement of Eq. (2) for channelling of a single crack, propagation of multiple channel cracks will take place, Thouless (1990). By (2) it is seen that the risk of channel cracks is reduced by increasing the fracture toughness of the film G_c, by reducing the residual strains in the film ϵ_0, by reducing the film thickness h and by reducing the film modulus E_f. The usual recommendation to avoid channel cracking in layered materials is to reduce layer thickness, since the residual strain is most commonly dictated by the manufacturing conditions. In the next Sections, we investigate the possibility of reducing the last term in (2); the stiffness of the film by allowing embedded particles to undergo debonding.

3. Particle reinforcement and effect of debonding

To gain insight in the phenomenon of debonding of particles in the film with the subsequent release of residual stresses, simplified models based on existing solutions are set up in this Section. The qualitative behaviour of the models is held up against more complete three dimensional finite element calculations for representative volume elements in later Sections of the paper.

The effective equi-biaxial in-plane residual stresses, σ_e, in the reinforced film are similar to the residual stress in Eq. (1) and given by

$$\sigma_e = \frac{E_e}{1 - \nu_e}\epsilon_e \quad (3)$$

where the effective Young's modulus of the particle reinforced film is denoted E_e and the effective Poisson ratio is ν_e and the residual strain in the particle reinforced film is the residual strain of the un-reinforced film multiplied by the matrix volume fraction, i.e.,

$$\epsilon_e = \epsilon_0(1 - f) \quad (4)$$

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