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Two-stage cracking of metallic bi-layers on polymer substrates under tension

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ABSTRACT

This study demonstrates two-stage cracking of an Inconel silver film system on a flexible Teflon substrate under uniaxial tension. *In situ* fragmentation experiments revealed that primary fracture of the brittle 30 nm Inconel overcoat induced brittle, secondary fracture of the normally ductile 150 nm silver base layer. Good correlation exists between the ratios of primary and secondary saturation crack spacing and individual layer thicknesses. Two-stage cracking was confirmed by cross-sectional analysis and explained by different steady state energy release rates of the individual layers. The results further illustrate how brittle layers are detrimental to the mechanical behaviour of polymer-supported thin film multilayer structures.

Thin metal films on flexible polymer substrates facilitate a variety of innovative technologies, including flexible displays [1,2] or satellite insulation [3,4] by providing flexibility, weight-savings and a unique combination of material properties. The complex functionality of individual applications often requires a multilayer design [5–7] and the combination of different metal layers can lead to improved adhesion, corrosion resistance or thermal stability. Brittle interlayers [6,8] or overcoats are commonly used to improve adhesion or corrosion resistance of ductile metal layers. However, the failure behaviour of ductile and brittle thin films on polymers is fundamentally different under tension. Brittle films fracture at low strains while ductile films typically thin locally (necking) and multilayers tend to take the failure behaviour of the most brittle layer when strained, bent or twisted [9,10]. *In situ* fragmentation testing is a common method to investigate the fracture behaviour of thin films on compliant substrates [11–14] where the uniaxial tensile test is combined with a surface imaging technique, such as optical microscopy (OM), confocal laser scanning microscopy (CLSM), or scanning electron microscopy (SEM) to observe failure of the film. More advanced techniques also include X-ray diffraction (XRD) and 4-point-probe resistance measurements to monitor the evolution of film lattice stress and electrical resistance, respectively, as a function of applied strain [15]. Characteristic values derived from this analysis (fracture strain, ε_f, saturation cracks spacing, λ_{sat}) can be used to calculate interfacial properties or fracture toughness of films [16–19]. This study investigates the fragmentation behaviour of a brittle Inconel and ductile Ag bi-layer system on fluorinated ethylene propylene (FEP, Teflon®) used as a second surface mirror for thermal insulation of satellites [3,4]. It will be shown that when the system comprises of a brittle overcoat film on a ductile layer, two-stage cracking is observed, which demands further investigation of brittle/ductile multilayer architectures.

Evaporated Ag films (nominal thickness 150 nm) on a 50 μm FEP substrate were supplied by Sheldahl Brand. To prevent the Ag from oxidizing a 275 Å overcoat of Inconel was applied. The Ag layer has a nanocrystalline microstructure. The coherent domain size was measured with X-ray diffraction as 55 ± 15 nm, using the simple Scherrer-Equation and the advanced Williamson-Hall method [20,21]. Five diffraction peaks were used to estimate the grain size with both methods. For the Inconel overcoat the grain size is on the order of the film thickness. Rectangular samples were cut with a scalpel to the dimensions of 5 × 40 mm for *in situ* fragmentation experiments, which were performed with an Anton Paar TS600 straining stage under an optical microscope (Olympus BX51) and a CLSM (Olympus LEXT 4100 OLS). In order to study the fracture of the Inconel and Ag layers independently, *in situ* OM straining experiments were performed looking onto the Inconel surface and focusing through the transparent FEP substrate onto the Ag layer. Samples were strained to maximum strains between 14 and 15% at a rate of 2 μm/s using pauses at pre-determined strains to allow for imaging of the film surface. Crack spacing analysis was performed with the OM and CLSM images with Image-J [22] and Gwyddion [23].

Synchrotron radiation (KMC-2 beamline [24], BESSY II, Berlin) was used to measure the fracture stress of the Ag film *in situ* using XRD and the $\sin^2\psi$ method [25]. The longitudinal Ag lattice strain (parallel to the tensile direction) was measured *in situ* during continuous straining (Anton Paar TS600, $\varepsilon_{max} = 12\%$, displacement rates of 1 μm/s and 2 μm/s) in reflection geometry. The (111) reflections of the Ag

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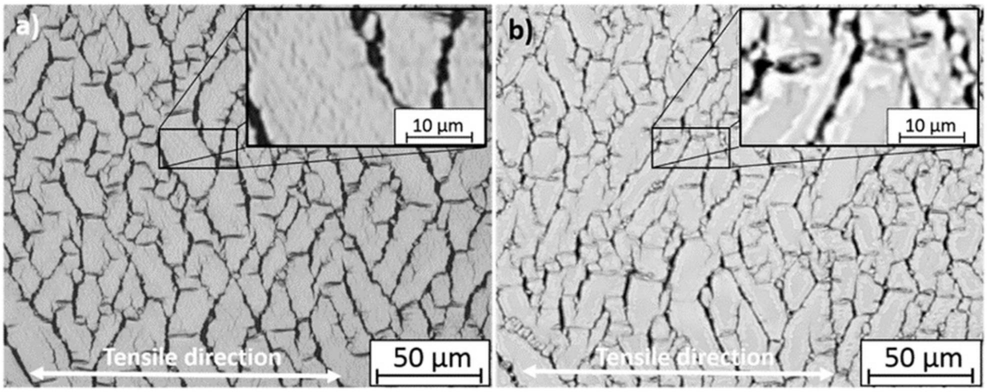

Fig. 1. *In situ* optical fragmentation analysis at $\varepsilon = 15\%$ revealing a two-stage cracking process. a) In the Inconel overcoat fine primary cracks (inset) and pronounced secondary cracks through both layers are visible. b) Only secondary cracks are visible in the Ag layer.

layer were recorded with a Bruker VÅNTEC 2000 detector using 5 different ψ angles between 0 and 50 degrees, with an exposure time of 5 s and a beam wavelength of 0.177 nm. A Pearson fit was applied to determine peak positions and peak widths. Film stresses were calculated using X-ray elastic constants (XECs) ($1/2\ S_2$) [26] for untextured (111) Ag reflections. XECs were calculated from single-crystal elastic constants assuming the Hill model with the software ElastiX [27]. The Inconel overcoat was too thin to receive a sufficient signal for $\sin^2\psi$ analysis and the Inconel fracture stress was estimated from *in situ* CLSM experiments using Hooke's law ($\sigma = \varepsilon E$, with the elastic modulus of the Inconel $E_{inc} = 166$ GPa [28]).

In situ optical fragmentation analysis from the Inconel side and the Ag side (through the transparent FEP substrate) revealed a two-stage cracking process of the bi-layer perpendicular to the straining direction (Fig. 1). On the Inconel surface a fine pattern of primary cracks (inset of Fig. 1a) is present between the secondary cracks, which form through the Ag film. From the Ag side only secondary cracks are visible, indicating that the fine primary cracks are constrained to the Inconel overcoat. The average distance between secondary cracks measured at 15% strain is identical for the Inconel and Ag sides indicating that secondary cracks have formed through both layers. The small features parallel to straining direction visible in Fig. 1a and b are tensile induced delaminations of the film at the Ag-FEP interface, which form around 4% strain. Buckling is compression-induced and a consequence of lateral contraction of the metal film on the polymer substrate during tensile straining. The tensile induced delamination phenomena has been described in [29,30].

To fully investigate the evolution of primary and secondary cracking as a function of strain, *in situ* CSLM experiments were performed. Fig. 2a shows a representative CLSM laser intensity image of the Inconel layer at 14% strain, with primary Inconel and secondary Ag cracks, indicated with black and white arrows, respectively. To measure the crack spacing of the primary and secondary cracks, surface profiles were extracted from the CLSM images. Primary cracks can be identified as small drops in the extracted profile (inset Fig. 2a) and the large drops correspond to secondary cracks. For each straining step, three profiles were extracted to calculate a mean value and standard deviation of the primary and secondary crack spacing. Fig. 2b summarizes the evolution of the primary and secondary crack spacing as a function of applied strain for all *in situ* OM and CLSM experiments.

Primary cracks were first observed at around 0.25% strain and the saturation regime was reached at about 4–6% strain (inset Fig. 2b). The saturation spacing of the primary Inconel cracks was measured as 2.3 ± 0.2 µm (dotted line, inset Fig. 2b). Around 1% strain the second stage of cracking initiated, where individual primary cracks transformed into secondary cracks through the Ag layer. The fracture strain of the Ag film is much lower than what was observed for 100 nm evaporated Ag and inkjet printed Ag, having fracture strains in the range of 5% [31]. With increased strain the density of the secondary cracks increased as more primary cracks propagate through the Ag layer until a saturation state is reached at around 8–10% strain. The evolution of secondary cracks from the OM and CLSM experiments compare well to one another (Fig. 2b) and show the same trend of decreasing crack spacing with increasing strain. The secondary saturation crack spacing (11.9 ± 1.9 µm), indicated with a dotted line, is 5 times larger than the saturation spacing of the primary Inconel cracks (2.3 µm) (ratio 5.2:1) and could be related to the film thickness ratio of 5:1.

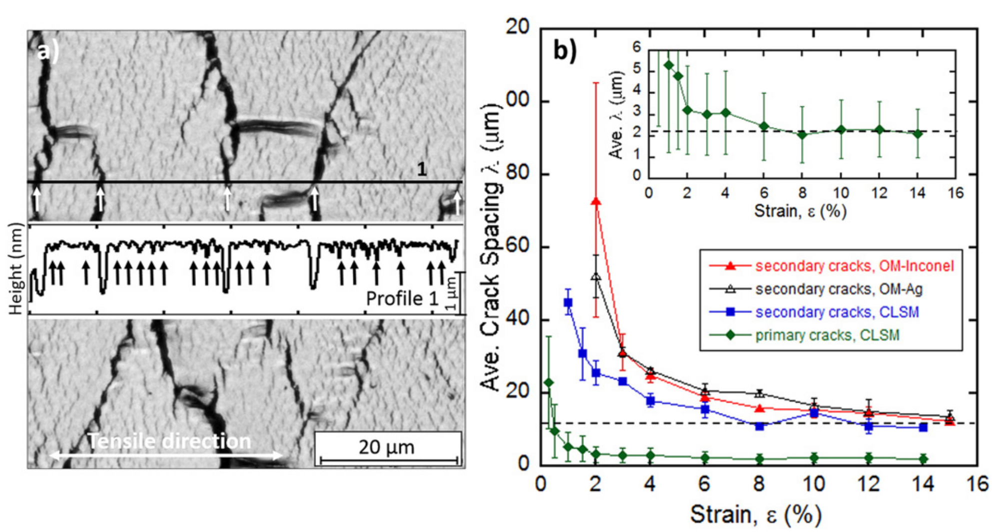

Fig. 2. a) CLSM laser intensity image ($\varepsilon = 14\%$) and extracted height profile. Primary and secondary cracks are indicated with black and white arrows, respectively. b) Crack spacing as function of strain for primary (*in situ* CLSM, inset) and secondary cracks.