Film thickness and architecture effects in biaxially strained polymer supported Al/Mo bilayers

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Abstract

Multilayers on polymer substrates are of interest for several technical applications. However, the true understanding of how one layer affects the other during mechanical loading is still unknown.

In order to address this lack of knowledge, single and bilayer thin films of Al (250, 125, and 75 nm) and Mo (50 nm) were sputter deposited onto polyimide (PI, 50 μm) and biaxially strained in-situ with X-ray diffraction. The technique allows for the simultaneous measurement of lattice strains of Al and Mo to correlate the observed mechanical behavior of both materials. Using the evaluated film stress and full width at half maximum evolutions, four domains of mechanical behavior are identified. The presence of the domains in either Al or Mo depends on the thickness of the Al films and, more importantly, the bilayer architecture (Al/Mo/PI vs. Mo/Al/PI). Domain boundaries in Al were found to correspond well with domain boundaries in the Mo, such as necking in Al with fracture in Mo or simultaneous fracture in both materials. The maximum stress achieved in the Mo layers during biaxial straining was also found to highly depend on the architecture and depend less on the Al film thickness. Results will demonstrate that the film architecture (layer order) is more important than the film thickness to enhance fracture resistance.

Keywords: Thin films; X-ray synchrotron radiation; Deformation; Fracture; In situ tension test

1. Introduction

Polymer supported metallic thin films have a wide range of applications, from multilayer insulation systems in space exploration, packaging (food or as antistatic foil) and electrodes in flexible displays or wearable electronics [1,2]. In the field of flexible and wearable electronics, the thin film material systems are mechanically evaluated with uniaxial, biaxial, twisting and bending tests [3–7]. Biaxial testing is very similar to the mechanical application load experienced by wearable applications. Likewise, it also resembles loading conditions caused by thermal cycling as a result of different thermal expansion coefficients between substrate and coating, for example in space applications. The enormous benefit of in-situ mechanical testing with XRD is the possibility for combined, yet material-differentiated investigation of deformation mechanisms in multilayer systems, which enables deeper insights and understanding of the interplay of various materials, layer thicknesses, and film architectures during testing.

In-situ experiments with XRD on polymer supported metal films have evolved from uniaxial loading to measure the material behavior of single films or bilayers [3,8–13] and multilayers [14–16], to biaxial straining [4,17–20]. From these results, deformation, fracture, and adhesion behavior of single ductile and brittle metal films on polymer substrates, which contain only one interface, is well understood under uniaxial testing [3,11–13,21–26]. Insight into how brittle adhesion interlayers or protection layers (containing two interfaces) induce brittle fracture in ductile films is also known for several bilayer systems [3,11,27–30]. There, the thickness of the ductile layer was found to be a deciding factor when through thickness cracks form. With multilayers, also known as nanolaminates, film thickness effects [14,15,31–34] and layer order [15] have been studied with in-situ XRD. Major findings were load sharing phenomena in ductile layers [14] and an improved adhesion in Cu/Nb nanolaminates when Cu is the first layer [15]. But, thickness variations remained below 100 nm and usually a 1:1 ratio of ductile-to-brittle film thickness was investigated. Layer order is hardly examined since significant changes in crack spacing were not observed in these material systems with many interfaces [15,35]. However, there is a need to reduce these multilayer architectures down to bilayers in order to study on a fundamental level the effect of thin film architecture (layer order) and to vary the ductile layer thickness to determine how different thin film architectures behave based on the position of the different interfaces in a systematic way. The hypothesis is that one of the interfaces in the bilayer or multilayer geometry, i.e. the ductile-polymer, brittle-polymer, or ductile-brittle (and vice versa) on polymer, control the mechanical behavior of the whole system.

The importance of the layer order in multilayer systems for crack initiation, propagation and electro-mechanical failure has already been demonstrated for Al/Mo bilayers [20] and thickness effects have been studied in other bilayer systems [3,8,26,28]. In this work, the role of Al layer thickness and layer order in Al/Mo bilayer thin film systems will be discussed based on a systematic thickness variation and in-situ XRD biaxial tensile experiments, whereby the Al film is thicker than the Mo film. Al/Mo multilayer films are a commonly used material system for electrodes in thin film transistor displays, whereby Al functions as a conductive layer and Mo simultaneously can act as an adhesion, diffusion barrier and protective layer. It will be demonstrated how the thickness of the Al layer impacts deformation and fracture mechanisms in different geometric configurations (single Al films compared to Mo/Al and Al/Mo bilayers) and how, based on that knowledge, multilayer architectures can be optimized for improved mechanical performance and flexibility without compromising functional properties.

2. Methods and Materials

Bilayers of Al and Mo, as well as single films of Al, were deposited onto 50 µm thick Upilex-S polyimide (PI) using direct current (DC) magnetron sputtering. The PI substrates were pre-cut into cruciform shapes of 200 mm diameter, 16 mm arm width and 5 mm radius where the arms cross. Films were deposited using an industrial scale magnetron sputter system (FHR.Line.600-V) equipped with a planar Al target (600 × 125 mm, 99.9995% purity, provided by FHR, Germany) and a rotary Mo target (∅ 125 × 600 mm, 99.97% purity, provided by Plansee, Austria) using an Ar flow rate of 300 sccm, corresponding to a working pressure of 5.3×10^{-3} mbar and a base pressure was equal to or less than 5×10^{-8} mbar for all depositions. For the Al and Mo targets a DC power of 3.5 kW and 4.0 kW was applied, respectively, yielding deposition rates of 3.6 nm/s for Al and 5.3 nm/s for Mo. Applying these conditions, the used FHR sputter deposition system enables a film thickness homogeneity of less than 5 % [36] for the PI cruciform substrates. Three thin film architectures, Al/PI, Mo/Al/PI, and Al/Mo/PI, were deposited. For Mo layers, the film thickness was kept constant at 50 nm, the Al layers were deposited with thicknesses of 75, 125 and 250 nm. After deposition, the films were partly etched away from the substrate to avoid (non equi-biaxial) edge effects [18], leaving circles with a diameter of approximately 15 mm in the center of the cruciform for testing. The Mo layers were removed with 65 vol.% HNO_3 and Al was removed with 30 vol.% NaOH aqueous solutions. The films were continuously loaded in tension under quasi equi-biaxial strain up to 3% at the DiffAbs beamline at the synchrotron SOLEIL (France), following a similar procedure as in [4,17]. The tensile test device available at the DiffAbs beam line comprises four independent motors and load sensors with a limit of 200 N. To avoid drift, preloading of the mounted samples in the range of 4 N is required. To get an accurate reading of the actual strain on the sample, digital image correlation (DIC) was performed every 10 s (experiments may take 2900 to 5900 s) [31,37] (Pixelfly camera from PCO), using a random speckle pattern (spray paint consisting of crystalline rutile-type TiO_2) on the backside of the sample [4]. The Debye-Scherrer rings of the crystalline stress free TiO_2 were also used for calibration of the diffraction patterns during the in-situ test. XRD experiments were performed with a beam energy of 9.66 keV ($\lambda = 0.124$ nm, spot size 290 × 280 µm², XPAD-S140 detector). For the in-situ XRD stress analysis, the stress is calculated from the measured strain in the crystal lattice using elastic constants [38]. As Mo and Al are only very slightly anisotropic, the isotropic elastic constants were used for the analysis of the Mo {110} and Al {111} diffraction peaks ($E_{Mo} = 320$ GPa, $\nu_{Mo} = 0.32$ and $E_{Al} = 70$ GPa, $\nu_{Al} = 0.33$, respectively [39]). Diffraction peaks during biaxial loading were only recorded in one principal direction (one azimuthal angle) instead of two to increase the density of measured points. Assuming a linear elastic distortion of the crystal lattice, the $\sin^2\psi$ method (five ψ angles: 30, 40, 50, 70, and 90°) was applied to determine the evolution of macroscopic stresses in the film. In a custom routine with Python 2.7, the position (2θ) and full width at half maximum (FWHM) of the diffraction peaks were determined using a Pearson VII function and a linear background. In addition to the macroscopic stress in the individual layers, the FWHM evolution provides information about the lattice defect density and strain heterogeneities through changes of the peak width and thus, contributes to the determination of different behavior (domains) in the sample [20].However, the collection of contributing factors that are reflected in the FWHM are challenging to differentiate. Therefore, in this work the focus is on a general interpretation of the FWHM evolution as a function of biaxial strain.

Following the tensile experiments, all samples were examined with scanning electron microscopy (SEM, Zeiss LEO 1525) and focused ion beam (FIB, Zeiss Leo 1540XB workstation) cross-sections. For selected samples, namely the 250 nm Al series, transmission electron microscopy (TEM) was performed. TEM cross section samples were prepared within a Zeiss Auriga workstation by means of a FIB liftout after coating the top with a protective Pt layer. The samples were analyzed in a JEOL 2200 FS operating at 200 kV using dark field scanning TEM (DF-STEM) mode to reveal the grains in the Al film.

3. Modelling of the sample geometry

Finite element modelling of substrate deformation for biaxial straining is crucial for sample design development and validation. This was satisfied by implementing a finite elements model built in Abaqus via Python 2 programming. For the sake of simplicity and symmetry, a quarter shell model of the polymer substrate (mainly S4 and some S3 elements) was used. Stress-strain curves obtained by the uniaxial tensile testing procedure as described in [40] were used as an input for the materials model. For the evaluation of the elastic constants four Upilex samples (6 mm × 45 mm × 50 µm) were tested using four loading-unloading cycles in the elastic range of 30-60 MPa. The resulting mean elastic modulus of 8.7 GPa with a Poisson's ratio of 0.34 and a yield stress of 147 MPa were used as an input for the simulation. To keep the evaluation straightforward, the von Mises stress was used as an indicator for the biaxiality of the stress. The von Mises stress along a 45° path starting at the center of the sample going to the middle of the radius was evaluated for the cruciform (Fig. 1a). These stresses ultimately lead to a flattening of the circular biaxial region and the first plastic deformation of the substrate will happen in this area at the curved radius outer edge between the arms. Therefore, the selection of the radius is an important parameter to consider for the sample design. For evaluation, the constant von Mises stress was defined as 1% deviation from the stress in the center of the sample, leading to a radius of approximately 6.5 mm at maximum applied strain. This exceeds the area that is measured via XRD. Fig. 1b shows that even at maximum strain plastic deformation of the substrate does not occur in the center of the sample where the measurements were made.

4. Results

Based on the film stress and FWHM data, four different deformation domains were identified for the investigated thin film systems, following the work of Faurie *et al.* [4], on biaxial tensile testing of Ni thin films. To identify and determine the position of the domain transitions indicated in Figs. 2, 3, 4 and 6 most accurately, all possible types of cross-correlations were considered, including comparisons between film stress and FWHM curves of respective Al/Mo pairs in one bilayer, as well as correlations between different Al (and Mo) layers within one architecture and between Al layers of the same thickness. The domains match a recently published work on Al/PI, Mo/Al/PI , and Al/Mo/PI systems with 250 nm Al layers and 50 nm Mo layers [20]. The data of [20] were obtained during the same beam time and were added to the results of this publication to yield a more comprehensive understanding of the influence of the Al layer thickness in different thin film architectures and how the different interfaces control the mechanical behavior.

The deformation domains will first be briefly introduced, then further explained during the discussion of the results of the individual thin film architectures presented in Figs. 2, 3, 4 and 6. Domain I is defined by elastic behavior and displays a linear stress increase and an ideally constant FWHM. Each film elastically deforms and no dislocations are stored as demonstrated by the constant FWHM. The following Domain II can be identified by a continued linear increase to bending over of the stress as well as an increase of the FWHM. Depending on the material, ductile (Al) or brittle (Mo), different processes take place in the thin films. Domain II is mainly attributed to micro-plasticity in the case of ductile films (Al) and to strain heterogeneities transmitted from interfaces in case of brittle films (Mo) [10]. FWHM increase indicates dislocation storage in materials that deform plastically. This will be further elaborated for the specific thin film architectures. Domain III only occurs in Al layers and is composed of constant or slightly decreasing stress levels as well as constant or slightly increasing (lower than Domain II) FWHM. This domain is characterized by localized plastic deformation. It is a characteristic deformation mechanism of polymer supported ductile thin films [3,12], also known as necking. The FWHM changes slope because fewer dislocations are being stored, because they are moving out of the Al film either to the film surface, interface, or as through thickness crack form with increased applied strain. In Domain IV, thin films fragment and develop through thickness cracks. This results in stress relaxation and also the evolution of FWHM changes compared to the previous domain. Depending on the architecture (layer order) and the specific material, the FWHM can either increase, decrease or remain constant in Domain IV.

4.1 Al/PI systems

Under biaxial load, the 250 nm thick Al film displays the full sequence of the four deformation domains (Fig. 2a). The elastic regime (Domain I) is followed by micro-plasticity (Domain II), necking (Domain III), and finally fragmentation (Domain IV). Sudden drops in film stress and FWHM at the end or during an experiment stem from instrumentation and are not characteristics of film deformation.

In comparison, the thinner 125 and 75 nm Al films only display the first three domains with nearly constant stress in Domain III (Fig. 2b,c), meaning only plastic deformation (e.g. necking or grain boundary sliding) and no fragmentation occurs. SEM micrographs and FIB cross-sections (Fig. 2d-f) verify cracks for the thickest 250 nm Al film (Fig. 2d) as well as the absence of Domain IV for the 125 nm and 75 nm Al films with only necking observed (Fig. 2e-f). Apart from the fragmentation of the 250 nm thick Al layer, the three Al films of different thicknesses display an overall very similar behavior (Fig. 2a-c). The boundaries between Domains I-III are at very similar and at nearly the same applied strain values

The increasing FWHM in Domain III gives a qualitative estimate on the growing plasticity in Al, such as, but not limited to, dislocation storage, during necking. The range of the FWHM within Domain III is similar in all three samples (Fig. 2, Table A1). Thinner Al films (125 nm and 75 nm) did not display fracture within the investigated strain regime. The lack of fracture behavior under monotonic straining is similar to that found for 50 nm Au on PI [11]. The continued FWHM increase at a lower rate (change in slope) in Domain III is due to the fact that in the thinner Al films more defects are stored during straining before through thickness crack formation occurs.

4.2 Mo/Al/PI systems

The addition of a 50 nm Mo layer on top of Al/PI influences the behavior of the underlying Al film. The Al film results will be presented first, however, will be displayed in Fig. 3 below the Mo film results to better illustrate the architecture difference to Fig. 4. In the Mo/Al/PI system with 250 nm Al, both stress and FWHM behavior of the Al layer (Fig. 3d) are similar to the single 250 nm thick Al layer in Fig. 2a. Domain III (necking) is shorter (strain range 1.2 - 1.6 %; Al-PI: strain range 1 - 2%), and fragmentation (Domain IV) starts earlier, at 1.6% strain (2% for Al-PI), in the Mo/Al/PI system. Quite obviously, these changes at equivalent film thicknesses are induced by the addition of the Mo overcoat [20].

Upon decreasing the Al layer thickness in the Mo/Al/PI system, the 125 nm thick Al layer in Fig. 3e shows a similar behavior compared to the 250 nm layer and also undergoes all four deformation domains, whereas the 75 nm thick Al layer still shows no Domain IV (Fig. 3f). Comparing all three Al thicknesses in the Mo/Al/PI systems, the range of stress and FWHM values of Domain I and II are similar, while Domain III differs in length. It should be noted that Domain III becomes longer and fragmentation is shifted to higher applied strains with decreasing Al layer thickness, until fragmentation is completely absent for the 75 nm Al layer within the investigated strain regime. This trend, combined with the behavior of the single Al films (Section 4.1), indicates that the Mo overlayer has a stronger influence on the plastic deformation of the ductile film with increasing ductile layer thickness. The fracture of the Mo layer induces through thickness crack formation in ductile Al and a similar behavior has been observed in the Inconel/Ag film system [30]. Dislocations can move to cracks or interfaces, especially the Al-PI interface that acts as a dislocation sink directly under Mo cracks where the formation of voids (Fig. 4g,h) aids in through thickness crack formation. This behavior related to the film thickness is opposite to what has been previously observed for bilayer systems with brittle interlayers subjected to uniaxial loading [3,11] and to the behavior observed with reversed layer order, as further discussed in Section 4.3. The total range of stress and FWHM in the Al layers of the Mo/Al/PI system move to lower values with decreasing Al layer thicknesses (Table A1), indicating that thinner Al layers in the Mo/Al/PI system reach lower maximum stresses under biaxial loads (Tables A1), similar to the Al/PI systems. While interpretation of the absolute FWHM values is difficult due to the manifold influencing factors (grain size, thickness, strain state, etc.) the increased relative FWHM for 250 nm Al indicates more plasticity related to dislocation activity as compared to thinner 125 nm and 75 nm films.

The Mo layers in the Mo/Al/PI system all experience Domain I and Domain II followed by Domain IV. All three film systems appear qualitatively similar to each other (Fig. 3a-c). However, it is also evident from the film stress and FWHM evolution that the decreasing underlying Al layer thickness does influence the behavior of the 50 nm Mo films. While for 250 nm and 125 nm Al layers, the beginning of Domain III (Fig. 3d, necking) in the Al layer and Domain IV of the Mo layer (Fig. 3d, fracture) coincide well, this is not the case when the Al layer thickness is further decreased to 75 nm. There, the beginning of Domain III (necking) of the Al layer (Fig. 3f) precedes the fracture in the Mo layer (Domain IV, Fig. 3c) by about 0.2% strain. This could indicate that necking in the Al layer occurs before fracture of Mo. In general, determining the Domain boundaries for Mo in this configuration (as the top layer, Mo/Al/PI) becomes more difficult, compared to the reversed layer order (Al/Mo/PI, discussed in section 4.3), owing to the offset between the maxima of the film stress and FWHM curves (Fig. 3a-c) that becomes more pronounced with decreasing Al thickness. During post-mortem SEM analysis, the Mo/Al/PI systems all show a mud crack pattern, where the crack spacing increases with the Al layer thickness (Fig. 3g-i, left side). It should be noted that it is much easier to observe the crack patterns in Mo/Al/PI compared to Al/Mo/PI. Relaxation of the polymer substrate after loading poses a challenge to imaging the deformation and cracks in the strained film systems. For 250 and

125 nm Al, cracks penetrate both layers, whereby the crack spacing λ in both layers seems to be the same (λ_{Mo} *vs.* λ_{Al}) which was previously observed for Al/Mo bilayers [29]. However, in the 75 nm Al layer system (Fig. 3i) cracking is only observable in the top Mo layer. Cracks that form could be closed due to the elastic recovery of the substrate [41] or as the evaluation of the XRD data suggests, the Al layer did not fracture and showed necking until the end of the experiment.

4.3 Al/Mo/PI systems

In the reverse bilayer order, the Al layers of the Al/Mo/PI systems display more qualitative similarity to the single Al films (Al/PI systems) with increasing Al layer thickness. The main reason for any differences is now attributed to the underlying Mo layers and the impact of the different interface on the Al behavior. In the Al/Mo/PI system the biaxial load is transferred from the PI substrate via the Mo layer [42] to the Al, thus the conditions and properties of the Mo layer severely impact the behavior of the Al layer. The 250 nm thick Al layer in the Al/Mo/PI system (Fig. 4a) experiences the full sequence of all four deformation domains and qualitatively resembles the single 250 nm Al film in Fig. 2a, including lower start values of the FWHM compared to the other Al/Mo/PI systems (Fig. 4a-c). However, the 125 and 75 nm Al layers (Fig. 4b,c) do not experience Domain III and transition directly from Domain II (micro-plasticity) to Domain IV (fragmentation). Fine cracks going straight through both the Al and Mo layers were found in cross-sectional post mortem SEM micrographs for all bilayer thicknesses (Fig. 4g-i). Cracks are hardly visible in both top view and cross-sections potentially due to the relaxation of the substrate after straining. A pronounced 2D crack pattern on the surface, comparable to Fig.3 g- i, is only observed for the thinnest, 75 nm Al film (Fig. 4i), whereby the crack spacing is significantly larger compared to the same Al thickness in the reversed order (Fig. 3i) and even larger than the largest crack spacing observed in Mo/Al/PI (250 nm Al, Fig. 3g). This difference in lateral crack morphology further demonstrates the significant effect of layer order and film thickness on crack formation and propagation. From the XRD data, it is evident that the stress relaxation in the Al layers caused by the fragmentation (Domain IV) is more abrupt the thinner the Al layer (Fig. 4a-c), in agreement with literature [3,11,26]. It should be noted that Domain III might be present for the 125 nm Al (Fig. 4b, box). The range is small and appears at the same time that experimental problems occurred causing a loss of data points and a sudden, slight decrease in both the stress and FWHM. Additional experiments are necessary to determine the behavior, and for simplicity, the discussion will assume that Domain III is not observed for the 125 nm Al film system. The stress increase in the 125 nm Al layer at the end of the experiment is still under debate, but could be related to the shear lag model and crack density saturation [43]. The peak stresses of the Al layers in the Al/Mo/PI system are very similar, but the 75 nm thick Al layer is slightly higher (Table A1). During fragmentation of the 250 and 125 nm Al layers, the FWHM reached saturation, indicating the end of dislocation storage in the Al films. Comparing the start of Domain IV of the Al layers, there is no direct relation between fracture strain and Al layer thickness. While it seems that thicker Al films fracture later, the decrease in fragmentation onset stagnates at 150 nm thickness. Even though fragmentation is quite clear from the XRD data, cracks on the surface of the 250 and 125 nm Al layer are very short, possibly discontinuous and hardly visible in the micrographs (Fig. 4g,h). Instead, shallow surface scratches from sample preparation, marked by white arrows, are visible (Fig. 4g).

All Mo layers in Al/Mo/PI (Fig. 4d-f) start in the elastic regime (Domain I) and fracture (Domain IV) within the experiment. The elastic regime and length of Domain I in Mo, both in terms of applied strain and film stress, seem to be independent of the overlaying Al layer thickness. However, upon further straining, the thickness of the overlaying Al layer does influence the deformation behavior of Mo. Only the Mo layer with 250 nm Al (Fig. 4d) displays a Domain II, which is attributed to strain heterogeneities arising from the interface with Al (continuity of displacement). The mechanical equilibrium between a thin layer of Mo and an upper thick layer of Al is certainly complex, and the interface with Al (for which plasticity occurs) must have a role in the strain heterogeneities observed in Mo. The fracture sites of the Mo layers (beginning of Domain IV) are shown in Fig. 4g-i and coincide with cracks in the Al layers. It can be seen that the stress relaxation at the beginning of Domain IV is more abrupt as the Al thickness decreases. Indeed, the multi-cracking of a brittle film must be accompanied by a relaxation of the average stress. But here, being in mechanical equilibrium with the flexible substrate and the upper Al layer, its behavior is more influenced as the upper Al layer is thick. Comparing the Mo and Al layers' Domain IV, it is evident that despite the stress relaxation due to fragmentation in the Al/Mo/PI system, the FWHM of the Mo layers continue to increase. The FWHM increase after Mo fracture is believed to occur because the Al induces strain heterogeneities into the Mo film due to the Mo being confined between the Al film and PI substrate. The Mo layer with the 250 nm thick Al layer reached the highest tensile strength of about 4.1 GPa, followed by 3.2 and 3.3 GPa for the Mo layers with the 125 nm and 75 nm thicker Al layers (Table A2), respectively.

In the Al/Mo/PI system with the 250 nm Al layer the domain boundaries of both layers match well. While the Al layer starts necking (Domain III), the Mo is simultaneously building up strain heterogeneities (Domain II). Fracture starts at around 2.0% strain for both layers. The domain boundaries of the Al/Mo/PI systems with thinner Al layers also match well. Bilayers with 125 nm and 75 nm Al show a similar elastic regime (Domain I) in both layers (Al and Mo) and no Domain III. While the Al layers experience Domains I and II (elastic regime and micro-plasticity), the Mo layers only experience Domain I during this same range of strain, after which a nearly simultaneous start of Domain IV (fracture) occurs. It is debatable if in Fig. 4e the small range at the end of Domain I, where the stress in the Mo layer is bending over, could be a Domain II (strain heterogeneities) with a possible Domain III in the Al layer as previously mentioned, but the FWHM of the Mo film does not show any recognizable change to further indicate Domain II. All Al thicknesses in the Al/Mo/PI system start to fracture (Domain IV) simultaneously with the Mo layer generating through thickness cracks in both layers (Fig. 4g-i). This is evident from the fact that the 125 nm and 75 nm Al do not exhibit fracture in the single layer geometry (Fig. 2b-c). For the thicker 250 nm Al film, the results suggest a more mutual, complex interaction of the Al and the Mo layer at the interface. Thereby, the ductile layer thickness is observed to be directly related to the occurrence and length of Domain III in Al and also the occurrence of Domain II in Mo. Even though there is a free surface for dislocations to exit the Al film, necking is not observed in the thinner films further illustrating that the Mo controls the Al behavior.

5. Discussion

The biaxially loaded Al/Mo/PI systems (Fig. 4) show that thinner Al layers assume a behavior similar to the brittle Mo layers, indicated by the lack of necking (Domain III) in the Al layers with decreasing Al layer thickness (Fig. 4). Except for the position and range of the Domains III and IV, the different Al layer thicknesses in the biaxially strained Al/Mo/PI systems did not have too much impact on the relative stress and FWHM changes (change between minimum and maximum value, Table A1, A2) during the experiments. On the other hand, Al films with a Mo overlayer (Fig. 3) exhibit a more ductile behavior (length of Domain III and absence of Domain IV) with decreasing Al layer thickness, similar to single Al films. Within the investigated strain range embrittling effects from the Mo overlayer are only observed for thicker Al films (125 and 250 nm). Additionally, the biaxial straining of the Al/PI, Mo/Al/PI, and Al/Mo/PI film systems causes the stress evolution to behave similar to uniaxial experiments of Au/PI, Au/Cr/PI, Cu/PI, Cu/Mo, Cu/Cr/PI, Cu/Ta, Cu/Nb/PI and Inconel/Ag/Teflon [3,11,13,15,26–28,30,44]. Similar characteristics include a stress plateau with necking of single ductile films and stress relaxation after fracture when a brittle layer is present (interlayer or top layer). More recently it was reported that for a sputter deposited Cu/Mo/PI architecture that the apparent mode I fracture toughness of the Mo layer increased with increasing Cu thickness [26], illustrating in another system that a thicker ductile film can improve the fracture resistance of brittle interlayer. Contrary to literature on brittle overcoats [30], no two stage cracking mechanism with bimodal crack onset strains and crack spacings was observed in this study. Regarding the mud crack pattern observed on the film surface, it is important to note that the crack spacing in the Mo/Al/PI systems scales with the thickness of the underlying Al film, clearly indicating the influence of the Al deformation on the fracture behavior of Mo. At an identical Al thickness (75 nm) the crack spacing observed with a Mo interlayer is much larger compared to Mo/Al/PI (Fig. 4i vs. 3i), indicating that the interlayer is dominating the cracking behavior in the latter case.

The microstructure of the 250 nm Al film systems were characterized with TEM cross-sections due to the large difference in the maximum stresses achieved (Fig. 5a-c, respectively). All three 250 nm Al layers – regardless of layer order – reveal a bimodal grain size distribution throughout the film thickness with smaller grains situated close to the interface on which the Al film was grown. This “seed layer” with a much smaller average grain size is approximately 100 nm thick and is followed by a larger grained second “sublayer” in the Al layer towards the top of the film. It is reasonable that the grain structure is similar to the seed layer presented in Fig. 5a-c at 75 nm and 125 nm thickness for the Al film systems (e.g. smaller grains) [10]. For the 125 nm and 75 nm single and bilayers, the maximum stresses of the Al films (Table A1) are relatively constant providing further evidence that similar microstructures exist for both thicknesses. Additionally, when the Scherrer equation [45] was applied to determine the smallest coherently diffracting domain size, the respective Mo and Al layers are similar with a slight trend to larger crystal domain size with thicker Al films (Table A3). It should be noted that only one sample of each film system was tested and more experiments are necessary for further statistically relevant discussions. Moreover, the Scherrer equation is only used here as a comparison for general trends.

To facilitate comparison between all different film thicknesses and architectures, the results are summarized in Fig. 6. In the Mo/Al/PI bilayer systems, the Mo layer controls the failure behavior of the Al layer. As shown in Fig. 6b and Fig. 3, necking in the Al films occurs at approximately the same time that fracture of the Mo layer is observed. It has been demonstrated that the residual stress of brittle layers can be tailored to increase the fracture strain by having high compressive stresses [22]. When Mo is the top layer, the residual stress of the Mo layer could be a dominating factor [22]. The residual stress of the Mo layer (Table A2, Mo/Al/PI, minimum stress) is almost double for the 75 nm Al film (-1.18 GPa) compared to the 125 nm and 250 nm films (approx. -0.52 GPa). Under biaxial loading, the Mo layer in the 75 nm film system reaches the highest stress (1.73 GPa) and higher fracture strain (approx. 1.4% strain, start of Domain IV) compared to the 125 and 250 nm Al films (both approx. 1.2%). In the bilayers, the maximum stresses in the Al films (Table A1, Fig. 6) only vary slightly for Al thicknesses above 125 nm. The thinnest 75 nm film shows a slightly lower (Mo top) or higher (Mo bottom) maximum stress depending on the architecture, indicating an increased sensitivity to the presence of the brittle Mo layer. The stresses in the Mo films do not vary with Al thickness (Fig. 6b, bottom). In contrast to the Mo/Al/PI system, the fracture strain of the Mo layers in Al/Mo/PI decreases with decreasing Al thickness of 250, 125 and 75 nm to 2.0, 1.4, and 1.4%, respectively (Fig. 4d-f), indicating that thick ductile overlayers are beneficial for the fracture behavior of brittle films on polymers. In this configuration the compressive residual stresses (Table A2, minimum stress) of the Mo layers at the beginning of the biaxial straining experiments only vary by a few hundred MPa, with the 125 nm Al system having the highest Mo residual stress. If the residual stress of the Mo layer was the main factor controlling the necking and crack formation in the Al films, the 125 nm Al system should have the highest fracture strain. But, this film system failed at about the same strain as the Mo film with the lowest residual stress (75 nm Al). Analogous to the fracture strains, the maximum stresses achieved in the Mo layers decreases with thinner Al layer thicknesses (Table A2). As shown in Fig. 6a-c, the maximum stresses of the Al films (Table A1) do not statistically vary significantly as a function of architecture. This further indicates that the Al grain structures are similar and that the Al failure is more influenced by the Mo layer due to its interface with the polymer substrate.

In comparison to the Mo layers in the Al/Mo/PI system (Fig. 6c), where the FWHM increases in Domain IV, the Mo layers in the Mo/Al/PI system show a decreasing FWHM trend (Fig. 6b). Also, the FWHM values in the Mo layers are generally lower in the Mo/Al/PI systems, with the exception of 75 nm Al/Mo/PI system. The significant difference in the 75 nm Al/Mo/PI system is not known since all Mo films were deposited using the same sputter parameters. Any further interpretations at this point would be highly speculative and require further experiments. The general difference in FWHM behavior is observed to be caused by the layer order. When the Mo layers are on top of the multilayer system, the interface with Al is much rougher as results of the Al grain size and topography (Fig. 5b vs. 5c) demonstrate. Furthermore, as the top layer is only constrained on one side (Al layer), the inherently brittle Mo layers in the Mo/Al/PI systems are less hindered during straining than their sandwiched counterparts in the Al/Mo/PI systems. There the influence of strain heterogeneities is less at the Mo/Al interface than the Mo/PI interface. The decreasing trend of the FWHM in the Mo layers in the Mo/Al/PI architecture is related to the progressive relaxing stress due to fragmentation. Moreover, as much as the fragmentation of a brittle film on a flexible substrate in its elastic domain induces an increase in heterogeneities due to a stiffness contrast of several orders of magnitude, here the Mo is adherent to a metal underneath which itself is fragmented along the same cracks. On the other hand, the Al is still in contact with the flexible substrate and continues to deform, which explains why the FWHM of the Al continues to increase during this time. It should also be noted that the maximum stress that the Mo achieves is about 2 times higher for the Al/Mo/PI system than for the Mo/Al/PI system (Table A2 and Fig. 6b-c) and is a further indication that when the Mo film is constrained between the Al and PI more energy is needed to cause fracture. The Al layer behaves more ductile (length of the Domain III) when the film is constrained by the Mo film (Mo/Al/PI bilayer). When Al is on top of the Mo interlayer, the 125 nm and 75 nm thick Al films have little or no Domain III (Fig. 6c). But, when Mo is on top of Al, all three Al film thicknesses have a Domain III with the longest being that for the 75 nm Al (Fig. 6b and 4a-c). Similarly, single Al films without any Mo layer (Fig. 6a and 2) exhibit a Domain III at all investigated film thicknesses, but no fragmentation below 250 nm Al.

A noteworthy point is how the ductile Al layer thickness influences fracture of the Mo layer and plasticity in the Al layer. As single Al layers, as the film thickness decreases, more plasticity is observed and for the maximum applied strain, the 125 nm and 75 nm films did not achieve fragmentation (Domain IV). For the Mo/Al/PI architecture, the Mo layer fractures first and as the Al thickness decreases the amount of plasticity increases (more necking, Domain III), illustrating more fracture resistance. Conversely, in the Al/Mo/PI architecture, both layers fracture at nearly the same strain and bilayer fracture strain depends on the thickness of Al. In this architecture, thicker films are more fracture resistant than thinner.

6. Conclusions

In-situ biaxial straining with XRD of single and bilayer thin film systems of Al and Mo supported on polymer substrates was performed to better understand the role of film thickness and layer architecture on the mechanical behavior. Four different deformation domains identified by the stress and FWHM evolution were used to describe the mechanisms found in the Al and Mo layers simultaneously. Depending on the Al thickness the Al layers in the Al/PI and Mo/Al/PI architectures experience three (I-III, thin Al) or all four (I-IV, thick Al) deformation domains, whereby the presences of the Mo overlayer has a stronger influence on thicker Al films (here, 250 nm Al), governing and shifting the occurrence of Domain IV (fragmentation) to lower strains. In the reversed bilayer order (Al/Mo/PI system) the occurrence of Domain III (necking) in the Al layers is affected by the Al thickness, whereby the thick 250 nm Al shows all four domains (I-IV), and thinner Al layers (125 and 75 nm) only experience three domains (I, II, IV). Mo films generally experience three domains (I, II, IV) when Mo is a top layer (Mo/Al/PI) and when the overlying Al film is sufficiently thick (i.e. 250 nm Al/Mo/PI). In the Al/Mo/PI architectures with 125 nm and 75 nm Al, only Domains I and IV were observed in the Mo layer. Domain boundaries in Al were found to correspond well with domain boundaries in the Mo for both bilayer architectures. The change of the Al layer thickness has an impact on the maximum stress and FWHM behavior of the Al layers under biaxial load for all architectures. However, the Al layer thickness impacts the stress behavior of the Mo layers depending on the architecture, with more stress needed to cause cracking in the Mo layer when the Mo is confined between the polymer substrate and the Al layer (Al/Mo/PI). The measured Mo stresses in the Al/Mo/PI system were about 2 times higher than the reversed Mo/Al/PI system. Additionally, the FWHM of the Mo layers illustrates a reproducible and characteristic behavior. When Mo is an interlayer the FWHM increases in Domain IV and when Mo is a top layer the FWHM decreases after fracture (Domain IV). In both cases the behavior is independent of the Al thickness. These results together demonstrate that the mechanical behavior of a ductile film (here, Al) is controlled by the fracture of the brittle film (here, Mo) and that thicker ductile films can improve the fracture resistance of brittle interlayers (here, Al/Mo/PI) because the smaller stiffness of Al is compensated by a higher thickness. Additionally, when Mo is on top of Al (Mo/Al/PI), thinner Al films (here, 75 nm Al) improve the fracture resistance. With this new knowledge, multilayer systems of ductile and brittle metals can now be designed with thicker ductile layers or a reversed bilayer architecture to achieve higher fracture resistance.

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8. Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Figure Captions

Figure 1: Finite element simulation of the stress conditions in the polymer substrate under equi-biaxial stresses. a) von Mises stress(in MPa) and b) equivalent plastic strain (PEEQ) at 3% overall applied strain. The SNEG fraction specifies the direction of the view of the shell, namely the bottom surface of the shell. This figure is in color in the online version.

Figure 2: Al/PI systems: (a-c) stresses and FWHM measured from in-situ XRD biaxial straining experiments indicating the different deformation domains present (I: Elastic, II: Micro-Plasticity (Al), III: Necking, and IV: Fragmentation). Al {111} diffraction analysis of (a) 250, (b) 125 and (c) 75 nm thick Al film of the Al/PI system. (d-f) Post mortem SEM micrographs of the surface (left side) and film cross-sections (right side). The black arrows in (d) mark a fine crack in the Al layer and necking in (e,f). The straight lines marked by white arrows in (f) are shallow surface scratches from transportation of the samples prior to testing. (color online)

Figure 3: Mo/Al/PI systems: (a-f) stresses and FWHM measured from in-situ XRD biaxial straining experiments indicating the different deformation domains (I: Elastic, II: Micro-Plasticity (Al) or Strain Heterogeneities (Mo), III: Necking, and IV: Fragmentation). (a-c) Mo {110} diffraction analysis of Mo/Al/PI. (d-e) Al {111} diffraction analysis of Mo/Al/PI. (g-i) Post mortem SEM micrographs of the surface (left side) and film cross-sections (right side). The black arrows in (g,h) mark a fine crack in the Mo and Al layer, and in (i) a crack on the Mo film. The white arrow in (i) indicates either necking or where the Al crack closed due to substrate relaxation. (color online)

Figure 4: Al/Mo/PI systems: (a-f) stresses and FWHM measured from in-situ XRD biaxial straining experiments indicating the different deformation domains present. Domains are the same as in Fig. 3. (a-c) Al {111} diffraction analysis of the Al/Mo/PI. (d-f) Mo {110} diffraction analysis of the Al/Mo/PI. (g-i) Post mortem SEM micrographs of the surface (left side) and film cross-sections (right side). The black arrows in the cross-sections mark fine cracks in the Al layers and coinciding cracks in the Mo layer appear closed due to substrate relaxation. The straight lines marked by white arrows in (g) are shallow surface scratches from transportation of the samples. (color online)

Figure 5: BF-STEM cross-sections of (a) 250 nm Al/PI, (b) 50 nm Mo/250 nm Al/PI and (c) 250 nm Al/50 nm Mo/PI illustrating the microstructure of the Al films with the corresponding high-mag insets (DF-STEM) below each film system. A thin Pt top layer is visible to protect the thin films during the FIB cutting.

Figure 6: Comparison of stresses and FWHM in Al and Mo films ordered by (a-c) architectures with different Al layer thicknesses. In the top row the Al stresses and FWHM and in the bottom row the Mo stresses and FWHM for (a) Al/PI, (b) Al/Mo/PI, and (c) Mo/Al/PI are displayed. For better visibility only every third data point is displayed. (color online)

Appendix A

Table A1: Minimum, maximum and relative change (between minimum and maximum) of stress and FWHM data in the Al layers of the different thin film systems from biaxial testing.

Al data	**Al 250 nm** Mo 50 nm	**Al 125 nm** Mo 50 nm	**Al 75 nm** Mo 50 nm	
Al/PI	0.03	0.02	0.02	min. stress [GPa]
	0.80	0.58	0.58	max. stress [GPa]
	0.76	0.56	0.56	ΔStress [GPa]
	0.149	0.139	0.167	min. FWHM [°]
	0.319	0.315	0.351	max. FWHM [°]
	0.170	0.176	0.184	ΔFWHM [°]
Mo/Al/PI	0.09	0.09	0.10	min. stress [GPa]
	0.71	0.68	0.60	max. stress [GPa]
	0.62	0.59	0.49	ΔStress [GPa]
	0.131	0.194	0.217	min. FWHM [°]
	0.280	0.327	0.342	max. FWHM [°]
	0.148	0.133	0.125	ΔFWHM [°]
Al/Mo/PI	0.06	0.00	0.09	min. stress [GPa]
	0.68	0.69	0.77	max. stress [GPa]
	0.62	0.69	0.68	ΔStress [GPa]
	0.142	0.201	0.204	min. FWHM [°]
	0.273	0.280	0.280	max. FWHM [°]
	0.131	0.079	0.076	ΔFWHM [°]

Table A2: Minimum, maximum and relative change (between maximum and minimum) of stress and FWHM data in the Mo layers of the different thin film systems from biaxial testing.

Mo data	Al 250 nm **Mo 50 nm**	Al 125 nm **Mo 50 nm**	Al 75 nm **Mo 50 nm**	
Mo/Al/PI	-0.49	-0.57	-1.18	min. stress [GPa]
	1.37	1.11	1.73	max. stress [GPa]
	1.86	1.68	2.91	ΔStress [GPa]
	0.371	0.391	0.400	min. FWHM [°]
	0.427	0.444	0.474	max. FWHM [°]
	0.056	0.053	0.074	ΔFWHM [°]
Al/Mo/PI	-2.20	-2.41	-1.83	min. stress [GPa]
	4.06	3.17	3.30	max. stress [GPa]
	6.26	5.58	5.13	ΔStress [GPa]
	0.473	0.465	0.378	min. FWHM [°]
	0.527	0.523	0.411	max. FWHM [°]
	0.054	0.058	0.033	ΔFWHM [°]

Table A3: Estimation of the individual layers' smallest crystal domain size, *D*, determined by the Scherrer equation at the beginning of the experiments.

Architecture	**D Al [nm]**	**thickness Al [nm]**	**D Mo [nm]**	**thickness Mo [nm]**
Al/PI	46	250	-	-
	47	125	-	-
	41	75	-	-
Mo/Al/PI	52	250	19	50
	34	125	18	50
	30	75	17	50
Al/Mo/PI	48	250	15	50
	34	125	15	50
	33	75	18	50