Advanced Characterization Techniques for Coatings and Thin Films
Room Royal Palm 1-3 - Session H2
Advanced Mechanical Testing of Surfaces, Thin Films and Coatings

Moderators: Benoit Merle, Friedrich-Alexander-University Erlangen-Nürnberg (FAU), Marco Sebastiani, University of Rome "Roma Tre"

1:30pm **H2-1 In Situ Observation of Strain Transfer and Crack Formation in Evaporated and Printed Thin Films and Devices on Compliant Substrates***, **Patric Gruber,*** Karlsruhe Institute of Technology (KIT), Institute for Applied Materials (IAM-WBM), Germany

Compliant substrates enable the fabrication of flexible electronics for numerous applications like flexible displays, solar cells, batteries or wearable/biocompatible electronics. However, the reliability of such devices is limited by the stretchability of the inorganic components. So far, little experimental work has been carried out to investigate the mechanical properties of thin inorganic films on compliant substrates at high strains and cyclic loading. Here, we present experimental results for the flow stress, fracture strain and fatigue behavior of evaporated and printed Ag films as well as printed thin film transistors on compliant substrates. The film systems have been tested by a synchrotron-based tensile testing technique (up to 10% total strain) as well as cycling loading (50 Hz, strain amplitude up to 2.5%) and have been characterized by SEM and FIB microscopy. The synchrotron experiments yield the stress evolution and strain transfer within the film systems whereas the cyclic tests give the fatigue lifetime. On the other hand, *in situ* electro-mechanical testing, *in situ* tensile tests in the SEM and stationary FIB investigations reveal the evolution of electrical performance, crack morphology and crack density as well as fatigue damage in the individual films. First, results of electro-mechanical testing of printed and evaporated Ag films will be presented. Electrical conductivity and mechanical reliability are investigated with respect to the inherently nanoporous microstructure, and are compared to those of evaporated Ag films of the same thickness. It is shown that there is an optimized nanoporous microstructure for inkjet-printed Ag films, which provides a high conductivity and improved reliability. It is argued that the nanoporous microstructure ensures connectivity within the particle network and at the same time reduces plastic deformation and the formation of fatigue damage. Furthermore, results on printed In_2O_3 thin film transistors are presented. Here, the interplay of the polymer substrate and solid polymer electrolyte with the metallic and ceramic interlayers within the transistor structure will be discussed based on the strain evolution within the individual layers determined from the *in situ* synchrotron experiments.

1:50pm **H2-2 Comparison of Different Methods for the Investigation of Thin Film Adhesion***, **Felix Schiebel,*** Fraunhofer Institute for Mechanics of Materials IWM, Germany*; C Eberl,* University of Freiburg, Germany

The reliability of power electronic devices (e.g. in solar industries and electric automotive applications) depends strongly on their electrical and mechanical interconnections. Thin metallic films deposited on semiconductor substrates are used to improve the connection at its most critical interface. The performance of these thin films, with dimensions typically of a few hundred nanometers, therefore greatly influences the reliability of the entire device.

To evaluate the adhesion of such films, a typical stack of thin metallic films consisting of an adhesion layer, a solderable NiV layer, and an Au oxidation protection layer was deposited on an Si semiconductor substrate. Several experimental methods (e.g. cross sectional nanoindentation (CSN) and scratch test) have been implemented to evaluate the adhesion in both the as-deposited state and an annealed state. The results of those measurements will be used to discuss the influence of the annealing on the adhesion and also on the measurement technique.

Furthermore, a new experimental setup will be presented which allows monitoring the delamination during CSN experiments with two separate cameras. The captured image series provide new experimental data which can be used to further improve fracture toughness calculations and to gain a deeper insight in delamination processes, allowing more the design of more reliable devices in the future.

2:10pm **H2-3 Electro-Mechanical Characterization of Functional Thin Film Metallic Glasses***, M Mühlbacher,* Montanuniversität Leoben, Austria*; O Glushko, **Christoph Gammer,*** Erich Schmid Institute of Materials Science, Austrian Academy of Sciences, Austria*; C Mitterer, J Eckert,* Montanuniversität Leoben, Austria

Thin film metallic glasses, which have a signature disordered structure with metallic bonding characteristics, are starting to emerge as functional materials. To investigate their electro-mechanical behavior, we prepare binary metallic PdSi thin films with varying thicknesses on the order of 5-300 nm by co-sputtering from elemental Pd and Si targets in an unbalanced DC magnetron sputtering device. In order to achieve amorphous film growth on Si(001) and flexible polyimide substrates, deposition proceeds without external substrate heating and film composition is chosen close to the deep eutectic composition in the Pd-Si phase diagram.

Selected samples deposited on flexible substrates are analyzed in a tensile testing set-up with *in-situ* resistance measurements. The strain at which cracks start to form in the brittle amorphous films can be determined exactly as the point where resistance starts to increase. Simultaneous confocal laser scanning microscopy of the strained samples provides more information on the failure mode. In thicker PdSi thin film samples, characteristic shear bands are evident with a 45° angle to the straining direction in addition to large cracks normal to, and film delamination in the form of buckles parallel to the tensile direction after straining to 10%. With decreasing PdSi film thickness down to 5 nm, a more ductile fracture behavior is observed. This is explained by a plasticity size effect, where deformation changes from a highly localized to a homogeneous mode due to geometric limitations. These findings are complemented by *in-situ* tensile tests of free-standing PdSi films in a transmission electron microscope. The gained understanding of the interplay of electro-mechanical properties in functional thin film metallic glasses will contribute to a successful application of these materials in future microelectronic devices.

2:30pm **H2-4 New Pull-off Tensile Tests for Adherence Assessment in Concrete-formwork Coated and Uncoated Contacts***, **Nicolas Spitz,*** Laboratory of Mechanics, Surface and Materials Processing (MSMP-EA7350), France*; N Coniglio, M El Mansori,* Arts et Métiers ParisTech d'Aix-en-Provence, Laboratory of Mechanics, Surface and Materials Processing (MSMP-EA7350), France*; A Montagne,* Arts et Métiers ParisTech de Lille, Laboratory of Mechanics, Surface and Materials Processing (MSMP-EA7350), France*; S Mezghani,* Arts et Métiers ParisTech de Châlons-en-Champagne, Laboratory of Mechanics, Surface and Materials Processing (MSMP-EA7350), France

Nowadays buildings construction is performed by pouring concrete into molds called formworks that are usually prefabricated metallic modules. Defects such as stripping may possibly form during the removal of the formwork if the interfacial bonding between the concrete and the formwork is high. Making use of a new pull-off tensile test designed in our laboratory, a correlation has been established between the formwork superficial functional signatures and its adherence susceptibility to concrete. The originality of this near-to-surface test was to characterize the concrete-to-formwork adherence by measuring the required force to pull the concrete from the formwork surface. The design of the test coupon was validated by finite element analysis that proves the small deformation of the tested formwork specimen under the tensile loading and the homogeneity of the applied tensile stress at the interface. The interfacial bonding to concrete has been compared between bare and coated formwork. Both metallic and polymer coatings have been studied. The analyses of the pull-off test results enabled us to understand the bonding mechanisms at the concrete-coating interfaces. The pull-off tensile test was proven capable of ranking formwork coatings according to their adherence to concrete.

2:50pm **H2-5 In-situ-squared: Combined Electro-mechanical Behavior of Thin Films with One Experiment***, **Megan Cordill,*** Erich Schmid Institute of Materials Science, Austrian Academy of Sciences, Austria **INVITED**

Studying the combined electro-mechanical properties of thin metal films on polymer substrates under mechanical load is one way to advance flexible electronic technologies. Ductile films and lines allow current flow between semiconducting islands and other operating features, thus are an integral part of flexible electronics. Flexible electronics also contain brittle layers which can improve adhesion, protect again corrosion or have semiconducting properties. When films on polymer substrates are strained in tension the substrate can suppress some of the catastrophic failure that allows for their use in flexible electronics and sensors. However, the interplay between the different layers can be very different compared to similar films on rigid substrates. In order to improve mechanical and electrical properties of these complex material systems, more work at characterizing the processing-structure-property relationships as well as the thin film architectures should be performed. Studies of strained films on polymer substrates tend to emphasize only the electrical properties or fracture strains effects more than the role of film thickness or multilayer behavior. The thickness of a ductile film will influence the mechanical behavior but also the electrical behavior, while even brittle thin metal layers can greatly affect the failure of ductile films. To address the electro-mechanical and deformation behavior of metal films supported by polymer substrates, in-situ 4 point probe resistance measurements were performed with in-situ confocal scanning laser microscopy imaging of films surface during uni-axial tensile straining. The 4 point probe resistance measurements allow for the examination of the changes in resistance with strain of the more deformable layer, while the surface imaging permits the visualization of localized thinning and crack formation. Furthermore in-situ synchrotron tensile tests provide information about the stresses in individual films and show the yield stress where the deformation initiates and the relaxation of the film during imaging. The combination of electrical measurements, surface imaging, and stress measurements allow for a complete picture of electro-mechanical behavior needed for the improvement and future success of complex flexible electronic devices.

3:30pm **H2-7 Mechanical Behavior of Ductile/Brittle Multilayers Studied with In-situ Straining Methods**, ***Patrice Kreiml**, M Rausch, V Terziyska,* Montanuniversität Leoben, Austria*; H Köstenbauer, J Winkler,* Plansee SE, Austria*; C Mitterer,* Montanuniversität Leoben, Austria*; M Cordill,* Erich Schmid Institute of Materials Science, Austrian Academy of Sciences, Austria

Mo thin films, which are frequently used as back-plane electrodes and as source/drain in thin film transistors, are inherently brittle compared to the electrical charge carrying ductile Cu or Al films. To improve the fracture behavior of stacks containing Mo layers for future flexible devices, and thus the reliability of the whole device, an optimum ductile/brittle layer thickness ratio is necessary. In this work the electro-mechanical behavior of multilayered Al/Mo thin films grown by dc magnetron sputter deposition on polyimide substrates is investigated. Various combinations of thicknesses for bi-layer and tri-layer architectures were evaluated (films of 50 to 450 nm thickness with different ductile/brittle layer thickness ratios). For a comparison of the behavior of the thin films, a combination of in-situ mechanical testing with electrical measurements, X-ray diffraction and 4-point probe resistance techniques were employed. In order to correlate the mechanical and electrical properties and to better understand the connection between crack onset, crack evolution and electrical resistance of the Al/Mo multilayers, in-situ confocal laser scanning microscope imaging was also performed. From X-ray diffraction data, individual film stresses during straining were determined, demonstrating that the ductile film thickness will dictate when through thickness cracks form in the multilayer. Once through thickness cracks form, the electrical conductivity deteriorates quickly and the multilayer stack can be considered failed. The combined in-situ testing also illustrates how brittle layers can cause typically ductile films to behave in a brittle manner due to the fact that cracks initially form in the brittle layers that act as stress concentrators in the ductile films. It will be shown that an optimized film architecture will improve the electro-mechanical behavior for flexible electronic applications.

3:50pm **H2-8 Fracture Behavior of Nanocrystalline BCC High-Entropy Alloys**, *Y Xiao, H Ma, R Spolenak, **JeffreyM. Wheeler**,* ETH Zurich, Laboratory for Nanometallurgy, Switzerland

Refractory high-entropy alloys (HEAs) have attracted significant attention due to their superior mechanical properties at elevated temperature [1]. However, most of them are brittle and suffer from low ductility and toughness at room temperature, and their usage is limited due to the inadequate fracture-resistance property. Grain boundaries play an important role in the extraordinarily high temperature strength and stability [2] of bcc HEAs and can also be potential sites for fracture. Here, strongly textured, columnar and nanometer-size grains NbMoTaW HEA thin films with and without ion beam assisted deposition technique are produced. Mechanical properties, especially fracture toughness are determined by in situ micro-pillar and micro-cantilever tests. Further characterization is conducted by high-resolution SEM images and TEM analysis.

References

[1] O.N. Senkov, et al., Intermetallics, 19 (2011) 698-706.

[2] Y. Zou, et al., Nano Lett, 17 (2017) 1569-1574.

4:10pm **H2-9 Recent Advanves in Microcantilever Bending Experiments**, ***Karsten Durst**,* Physical Metallurgy, TU Darmstadt, Germany*; M Göken,* University Erlangen-Nürnberg, Germany*; J Ast,* EMPA (Swiss Federal Laboratories for Materials Science and Technology), Switzerland **INVITED**

Microcantilever bending experiments are becoming more prominent in testing the local fracture toughness in a wide variety of materials. By focused ion beam milling or other techniques, a notched cantilever with micron sized dimensions is milled into a region of interest of a bulk material or thin coating. By loading the cantilever with a nanoindenter, a crack is nucleated at the notch tip and propagated through the sample and the fracture toughness can be evaluated at the local length scale. Applying conventional fracture mechanics approaches, the local critical stress intensity of the material is determined. With the presentation recent advances in the technique will be presented, focusing on elastic plastic fracture mechanics and crack tip plasticity.

4:50pm **H2-11 Temperature and Loading Rate Influence in Micro-Scale Fracture Experiments**, *J Ast,* Empa, Swiss Federal Laboratories for Materials Science and Technology, Switzerland*; J Schwiedrzik,* EMPA, Swiss Federal Laboratories for Materials Science and Technology, Switzerland*; J Wehrs,* Empa, Swiss Federal Laboratories for Materials Science and Technology, Switzerland*; J Michler,* EMPA, Swiss Federal Laboratories for Materials Science and Technology, Switzerland*; **Xavier Maeder**,* Empa, Swiss Federal Laboratories for Materials Science and Technology, Switzerland

The process zone or plastic zone around a loaded crack tip can significantly influence the fracture behavior of a material. Especially in micro-scale specimens, the plastic zone size may make out a large share of the sample volume and lead to a different fracture behavior than the one usually observed for macroscopic samples of the same material. Furthermore, the theoretical description of the plastic zone according to Irwin is not valid for single crystals. Therefore, a characteristic elastic-plastic fracture behavior is observed depending on crystallographic sample orientation and slip system activation. It is the aim of the study to give insight into the fracture process and behavior in micro-scale specimens in the presence of crack tip plasticity.

Notched micro-cantilevers were prepared by focused ion beam (FIB) milling in a tungsten single crystal. This material has nearly perfect elastic isotropy, a limited amount of activated slip systems and detailed knowledge of the macroscopic fracture behavior is available [1]. The cantilevers have dimensions of 25 µm in length, 5-7 µm in thickness and crack length to thickness ratios a/w of ca. 0.4. Loading rate and temperature are known to influence the fracture behavior decisively in bcc metals. Therefore displacement-controlled fracture tests were performed inside a scanning electron microscope in the temperature range between -100°C and 500°C. Applying the recently presented J-Integral technique [2] to plot continuous crack resistance curves, the fracture toughness and brittle-to-ductile transition (BDT) temperatures, which depend on the applied loading rate, were determined. This allows a thorough investigation of the activation energy of the BDT at the micro-scale.

Crack tip plasticity before and during crack growth was investigated by high-resolution electron backscatter diffraction measurements (HR-EBSD) on FIB cross-sections of the micro-cantilevers after mechanical testing. Plastic zones, which are strongly depending on the activated slip systems, and plastic strain gradients in terms of geometrically necessary dislocations were quantified and linked with the observed BDT behavior. Transmission electron microscopy was used to confirm the EBSD results and to provide dislocation analysis.

[1] P. Gumbsch, J. Riedle, A. Hartmaier, H.F. Fischmeister, Controlling Factors for the Brittle-to-Ductile Transition in Tungsten Single Crystals, Science. 282 (1998) 1293–1295.

[2] J. Ast, B. Merle, K. Durst, M. Göken, Fracture toughness evaluation of NiAl single crystals by microcantilevers - a new continuous J-integral method, Journal of Materials Research. 31 (2016) 3786–3794.

5:10pm **H2-12 Investigating the Local Fatigue Properties of Materials in Small Dimensions by Dynamic Micropillar Compression**, ***Benoit Merle**,* Friedrich-Alexander-University Erlangen-Nürnberg (FAU), Germany

A novel method was develop in order to investigate the local fatigue properties of ultrafine-grained copper on the micrometer-scale up to the high cycle fatigue (HCF) range, relying only on widely available nanoindentation hardware. This breakthrough was achieved by combining the widely used micropillar compression method with the fast actuation

(40 Hz) provided by the continuous stiffness measurement (CSM) module, originally intended for pyramidal nanoindentation. Cyclic testing was performed at constant nominal stress amplitude for up to several million (3.10^6) cycles. The resulting strain amplitude was directly recorded and the plastic strain was evaluated from the phase angle measured by the lock-in amplifier during testing. Defining a threshold for strain amplitude decrease as failure criterion further enabled the determination of S-N curves. The fatigue behavior of the tested ECAP copper micropillars was found to be dominated by cyclic softening, which is in line with previous macroscopic observations on similar samples. The calculated plastic strain amplitude also matches the literature data closely. Generally, the new method has a great potential for studying the local cyclic effects taking place at interfaces in complex micro-architectured materials and coatings.

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