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Research paper

Influence of extreme thermal cycling on metal-polymer interfaces

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ABSTRACT

Thin metal films on polymer substrates are key components for a variety of terrestrial and space applications such as flexible microelectronics or multilayer insulation systems. Although these applications seem to be quite different, there is a similarity in terms of thermal stress. In both cases the material undergoes a considerable amount of thermal cycling of ± hundreds of degree Celsius during its lifetime. Due to the composite nature of the material it is important to investigate interfacial properties as a function of this thermal cycling. This work investigates interface strength and electro-mechanical behaviour as a function of thermal cycling for vapour deposited Aluminum (Al) films (150 nm) on 25 μm Polyimide (PI). The material is subjected to various numbers of thermal cycles of ± 150 °C in a gaseous N_2 atmosphere. Tensile induced delamination with in-situ resistance measurements is used to measure adhesion values as well as the electro-mechanical failure behaviour. It was found that although the adhesion of Al-PI is unaffected by the applied thermal load, a strong decrease in the electro-mechanical properties of the system was observed. Since good electro-mechanical properties are crucial for flexible electronic applications the resistance against thermal cycling needs to be taken into account when assessing the suitability of a material for flexible applications.

1. Introduction

Thin metal films on flexible polymer substrates are a highly attractive field of research. One of their key applications is the area of flexible electronics where they are used as electrical connections between semiconducting islands and other operating features [1,2]. During the lifetime of a device those connections can undergo up to 10,000 thermal cycles of ± hundreds of degree Celsius due to current induced heating. Another area of use is multilayer insulation systems (MLI), which are key components for today's and future space missions. To ensure optimal operational conditions for the very sensitive scientific equipment on board of a satellite, MLIs are used to decouple the interior of the spacecraft from the thermal conditions outside with temperatures ranging from −70 and +125 °C, depending on the specific purpose and position of the spacecraft in relation to the sun [3,4]. Typically a Low Earth Orbit (LEO) spacecraft encounters 16 thermal cycles a day leading to about 6000 thermal cycles during one year of operation [5]. In both cases the material sees a considerable amount of thermal cycles during its lifetime. Because of the composite nature of the material system it is important to investigate interfacial properties as a function of this type of thermal cycling. Enhanced diffusion along interfaces in combination with high temperatures and the mechanical stresses generated by different thermal expansion coefficients and changing temperatures can alter the interface behaviour. At worst, delamination of the metal film from the polymer substrate and the loss of the specific function of the material can be consequences of these processes.

Adhesion is the parameter characterizing the strength of a metal-polymer interface. The adhesion between two materials is mainly determined either by mechanical interlocking, or interfacial diffusion, or chemistry [6]. Surface roughening and modification of the polymer surface prior to film deposition are two techniques used to promote mechanical interlocking or chemical reactions [7]. There are different techniques to measure adhesion such as tape test, peel test, spontaneous buckling, nanoindentation induced buckling or tensile induced delamination, giving either qualitative or quantitative adhesion values [6,8,9]. Tensile induced delamination is particularly suitable for flexible substrates. When thin films on polymers are strained in uniaxial tension localized delamination in the form of buckles initiate parallel to the straining direction if laterally compressive stresses occur [10,11]. An adhesion model, presented by Cordill et al. [8] allows the calculation of interfacial adhesion values from the buckle geometry. The model has been used to study thin Cr and Ti films on flexible polyimide (PI) and polyethylene terephthalate (PET) substrates [8,12–14]. Important for thin films used in flexible electronics is the electro-mechanical behaviour, the evolution of the electrical film resistance, *R*, as a function of applied mechanical load on the film-substrate couple [15,16]. The film resistance is a very powerful tool to detect changes in the thin film such as grain

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growth or the formation of localized thinned regions and cracks as a result of mechanical load [17–20]. This work uses tensile induced delamination with in-situ resistance measurements to investigate the adhesion and electro-mechanical failure behaviour of thin Al films on PI as a function of thermal cycling. The material under investigation can be classified as an Al-on-PI type of interface [21] and was specifically designed for the use as a MLI. The combination of interface strength and electro-mechanical response will give detailed insight into the processes occurring at the metal-polymer interface during thermal cycling. This information can be used to improve the resistance of metal-polymer interfaces against thermal cycling and increase the reliability of flexible electronic devices and MLI systems.

2. Materials and methods

Vapour deposited Al films on 25 μm Polyimide (PI) substrate (Upilex-S®) were supplied by UBE Europe GmbH. The Al metallization consists of two individual Al layers of 50 nm and 100 nm for the bottom and the top layer, respectively. The initial grain size of the films was on the order of the film thickness for both Al layers. Film thicknesses and grain size were measured from focused ion beam (FIB, Zeiss AURIGA crossbeam, operation currents: 500 pA coarse-cut, 50 pA polishing, operation voltage SEM: 5 kV) cross-sectional images. Thermal cycling (0–200 cycles) was performed with a Weiss Enet Temperature Chest Facility in a gaseous N_2 environment. The maximum and minimum temperature for one thermal cycle were ± 150 °C with a heating rate of 10 °C per minute. At the maximum temperature a hold period of 15 min was applied.

After thermal cycling rectangular samples used for tensile straining were cut with a scalpel to the dimensions of 5 × 40 mm. Tensile induced delamination [8] was used to measure the adhesion of the Al-PI interface as a function of cycle number. Rectangular samples (width 5 mm, initial gauge length $L_0 = 20$ mm) of each cycle number were continuously strained to a maximum strain of 40% under uniaxial tension to induce film cracking and delamination. Straining was performed with an MTS Tyron250® universal testing machine with a 4 point probe (4PP) resistance measurement setup (Keithley 2000 multimeter, current: 1 mA) incorporated into the tensile grips following the process of Glushko et al. [18]. Joule Heating during the in-situ resistance measurements is negligible due to the low applied current. A constant displacement rate of 5 μm/s was used for all experiments. The constant contact resistance of the film under the grips, measured with a second 4PP setup, was subtracted from the recorded resistance values in order to not underestimate the tensile induced resistance increase [18]. After straining the sample surfaces were investigated with scanning electron microscopy (SEM, LEO 1525, operation voltage: 5 kV) and atomic force microscopy (AFM, Veeco Dimension 3100). SEM and AFM images were analysed with ImageJ [22] and GWYDDION [23], respectively, to quantify the amount of film damage and to measure the dimensions of the delaminations. At least 30 buckles were measured for each cycle number. The buckle height, δ and the half buckle width, b, were measured from extracted surface profiles and used to calculate the adhesion energy. Cross-sectional FIB cuts were used to identify the delaminating interface.

3. Results and discussion

There was no significant difference in the initial resistance R_0 of cycled and uncycled samples that would indicate a change in microstructure or point defect population caused by thermal cycling. Small variations in R_0 can be attributed to slightly different sample dimensions. The normalized values of R_0/(sample width) remain constant at around 0.5 Ω/mm. However, the normalized resistance curves R/R_0 (Fig. 1) recorded during tensile straining show a clear change in the electro-mechanical behaviour of the Al-PI system due to thermal cycling. At the beginning all curves approximately follow the theoretical curve for perfect plastic deformation $R/R_0 = (L / L_0)^2$. In theory R/R_0 should follow the theoretical curve until mechanical damage such as cracks or deformation in the film occur which can be referred to as the failure strain, ε_f. After 100 and 200 thermal cycles, R/R_0 deviates from the theoretical curve at lower applied strains ($\varepsilon = 1.8\%$) compared to zero cycles ($\varepsilon = 3.2\%$). The failure strain of a film depends on various parameters such as grain size, film thickness, strain rate, residual stresses and interface adhesion [24–26]. A decrease of the fracture or deformation strain due to thermal cycling, as observed here, can be considered an embrittlement of the material. Embrittlement of thin films as a result of thermal treatment can be found for other thin film systems in literature. Cordill et al. [27] found similar results for Cu/Ti thin films on PI. In-situ SEM straining of Cu/Ti films revealed that annealed films have a reduced fracture strain compared to the as-deposited films. In the case of 100 nm Cu/10 nm Ti the failure strain decreases from 4.9% for the as-deposited material to 2.3% after annealing for 90 min at 350 °C. Similarly, transparent conducting oxide (TCO) films on polyethylene naphthalate (PEN) have a reduced crack onset strain when strained in uniaxial tension at elevated temperature from 1.55% at 23 °C to 1.2% when tested at 150 °C [20]. This decrease was attributed to tensile stresses controlled by the thermal expansion of PEN, as also to the increasing elastic contrast resulting from the softening of the polymer [20].

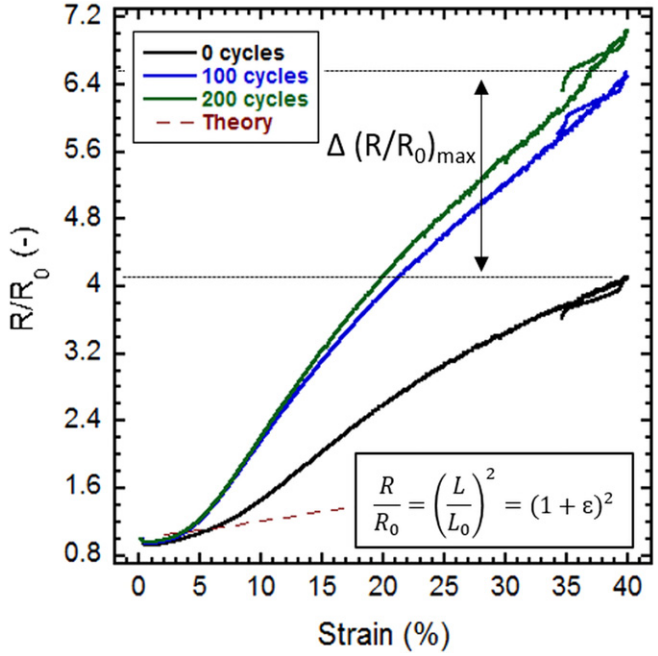

Fig. 1. Normalized resistance as a function of applied strain demonstrating a clear change in the electro-mechanical behaviour after 100 thermal cycles. For cycled samples the normalized resistance at equivalent strains is higher and the failure strain ε_f is lower.

Upon further straining after the film initially fails, the resistance increases as more through thickness cracks (TTCs) form in the film. The saturation strain, from which the crack spacing remains constant (typically 8–10% for ductile metal films [17,28]) cannot be assigned to a certain strain value in the normalized resistance curves. The phenomena of the further increase of the film resistance after the saturation state is reached can be found for ductile and brittle thin films [19] and is currently the subject of further investigations. The normalized resistance at equivalent applied strains for the cycled material is about 1.5 to 2 times higher compared to the non-cycled state. After unloading the normalized resistance decreases slightly but remains high compared to the initial normalized resistance. This result corresponds well to the cracks and necks in the Al film that are observed with AFM and SEM after straining.

After straining to 40% the amount of film damage was quantified from SEM micrographs. When a ductile film is strained in tension