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THERMO-HYGRO-MECHANICAL BEHAVIOUR OF ULTRATHIN MULTILAYER COMPOSITE MATERIALS FOR FLEXIBLE DISPLAY APPLICATIONS

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Abstract:

A theoretical modeling framework is proposed to follow shape and residual stress evolutions of layered flexible display structures with respect to coating deposition process, ambient temperature and relative humidity conditions. In the presented simplified version of this model, the mechanical equilibrium equations of elastic layers is coupled with transient mass transfer equations for the predictions of temperature and moisture content inside the structure. To calibrate this model, the behaviors of nanometric silicon nitride barrier layers on polyimide substrates were investigated. Then experimental measurements of the deflection dynamics of samples subjected to isothermal relative humidity jumps were compared with simulation results by considering different cases of boundary conditions. It is concluded that accurate simulations require accounting for the dependence of the water concentration-dependent diffusion coefficient.

Résumé :

Un modèle théorique est proposé afin de suivre l'évolution de la géométrie et des contraintes résiduelles de structures multicouches d'écrans flexibles en fonction du procédé de déposition des couches, de la température ambiante et des conditions d'humidité relative. Ce modèle, dans la version simplifiée qui est présentée ici, couple des équations d'équilibre mécanique des couches, vues comme étant élastiques, avec une équation d'évolution de la concentration en eau dans la structure. Pour en faire la calibration, le comportement de couches barrières de nitrure de silicium déposées sur un support polymère a été étudié. Des mesures expérimentales de l'évolution de la déflexion d'échantillons au cours de sauts d'humidité relative isothermes ont ensuite été comparées avec des résultats de simulations pour lesquelles différentes conditions aux limites ont été considérées. On peut en conclure que l'exactitude des simulations requiert de prendre en compte la dépendance du coefficient de diffusion avec la concentration en eau.

Key-words:

Flexible displays, Thin layers, FEM modeling

1 Introduction

Thin inorganic films deposited from a vapor phase onto a polymer substrate are used as barrier layers towards ingress of oxygen and moisture in flexible displays. Residual stresses are inherent to such layered composite materials and comprise intrinsic (deposition), thermal and hygroscopic (temperature and relative humidity variations and mismatch between material properties) contributions, see e.g. Leterrier (2003). Prediction and control of residual stresses is key to achieve a high dimensional stability and avoid premature damage in the display structure. Nanometric silicon nitride (SiN_x) layers on polyimide substrates were investigated. Firstly, the thermo-hygro-mechanical behavior of individual materials, including the water diffusion and sorption properties of polyimide were measured. Secondly, these data were used to calibrate a non-linear finite element model designed to follow shape evolutions of display structures with

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respect to relative humidity (RH) conditions. Thirdly, simulation results are compared with curvature measurements of the films subjected to an isothermal RH jump. The influence of i) time-evolving boundary conditions and ii) type of moisture diffusion coefficient on the quality of the simulation is finally discussed.

2. Coupled model

Considered bilayer structures are formed with a coating deposited on a polymer substrate. Each material of these structures is designated n and occupies the domain $\Omega^{(n)}$. Both materials are assumed to be homogeneous and perfectly bonded, with no cracks or defects. Whatever the considered physical phenomenon, all materials are assumed to be isotropic.

Assuming that body and acceleration forces are negligible, the momentum balance equation of each material n writes:

$$\underline{\text{div}} \underline{\underline{\sigma}}^{(n)} = \underline{\underline{0}} \quad (1)$$

The linear stress/strain relationships for each material n is

$$\underline{\underline{\sigma}}^{(n)} = \frac{E^{(n)}}{1+\nu^{(n)}} \left(\underline{\underline{\epsilon}}^{(n)} + \frac{\nu^{(n)}}{1-2\nu^{(n)}} \text{tr}(\underline{\underline{\epsilon}}^{(n)}) \underline{\underline{\delta}} \right) - \frac{E^{(n)}}{1-2\nu^{(n)}} \left(\underline{\underline{\epsilon}}^{(n)th} + \underline{\underline{\epsilon}}^{(n)hy} \right) + \underline{\underline{\sigma}}^{(n)int}, \quad (2)$$

where $E^{(n)}$ and $\nu^{(n)}$ are respectively the Young's modulus and the Poisson's coefficient of each material n and where $\underline{\underline{\delta}}$ is the unit tensor and $\underline{\underline{\epsilon}}^{(n)}$ the linear strain tensor

$$\underline{\underline{\epsilon}}^{(n)} = \frac{1}{2} \left(\underline{\underline{\text{grad}}} \underline{\underline{\mathbf{u}}}^{(n)} + {}^t \underline{\underline{\text{grad}}} \underline{\underline{\mathbf{u}}}^{(n)} \right), \quad (3)$$

where $\underline{\underline{\mathbf{u}}}^{(n)}$ is the displacement field of the material n . The tensor $\underline{\underline{\sigma}}^{(n)int}$ represents the intrinsic stress of the material n related to e.g. the growth of vapor-deposited layers. The thermal and hygroscopic strain contributions are written respectively as:

$$\underline{\underline{\epsilon}}^{(n)th} = \left[\int_{T_{ref}}^{T^{(n)}} \alpha^{(n)}(\theta) d\theta \right] \underline{\underline{\delta}} \quad \text{and} \quad \underline{\underline{\epsilon}}^{(PI)hy} = \epsilon^{(PI)hy}(c) \underline{\underline{\delta}} \quad (4)$$

where θ is the temperature, T_{ref} a reference temperature from which the strain and the stress fields are measured, $T^{(n)}$ the current temperature field and $\alpha^{(n)}$ the coefficient of thermal expansion (CTE) of the material n and $\epsilon^{(PI)hy}$ is determined below.

Assuming that there is no source of moisture in the domains $\Omega^{(n)}$, the evolution of the temperature $c^{(n)}$ of the constituent n is described by

$$\frac{\partial c^{(n)}}{\partial t} + \text{div}(-D^{(n)}(T, c) \underline{\underline{\text{grad}}} c^{(n)}) = 0, \quad (5)$$

where $D^{(n)}$ is the diffusion coefficient. The dependence of the coefficient of diffusion on the temperature $T^{(n)}$ and the moisture concentration $c^{(n)}$ is given by the following general form (Benjelloun-Dabaghi 2001):

$$D^{(n)}(T, c) = D_0^{(n)}(T) \lambda(T, c) \quad \text{with} \quad \lambda(T, c) = e^{\psi(T)c}. \quad (6)$$

In the following, this coupled problem is solved numerically by the finite element method (FEM) using the dedicated software Comsol 3.2. A weak-coupling strategy between the momentum balance equation and the moisture diffusion equation is adopted.

3. Studied structures: experimental determination of mechanical and sorption parameters

A 125 μm thick polyimide (PI) substrate (Upilex S, UBE) was used. The PI foils were annealed at 120°C during 30 min before deposition of SiN_x coating having a thickness $h_c = 400$ nm by

plasma enhanced chemical vapor deposition (PECVD) at a temperature of 200°C. During PECVD, the PI foils was maintained at a temperature $T=200^\circ\text{C}$ and kept flat on the deposition web.

The Young's modulus, Poisson's ratios, CTE's of both coating and substrate materials were determined previously in Dumont et al. (2007). In this paper, a compressive intrinsic stress was also identified in the plane of the silicon nitride coating:

$$\underline{\underline{\sigma}}^{(SiNx)int} = \sigma^{(SiNx)int} (\underline{\underline{e}}_1 \otimes \underline{\underline{e}}_1 + \underline{\underline{e}}_3 \otimes \underline{\underline{e}}_3). \quad (7)$$

The moisture sorption behavior of the polyimide PI was experimentally studied by performing sorption experiments (for the technique used, see e.g. Bhargava et al. (2006)). The moisture diffusion coefficient was determined as a function of the moisture concentration (see figure 1a), i.e. at room temperature:

$$D^{(PI)}(c) = 1.53138 \times 10^{-14} e^{-0.0315c}. \quad (8)$$

An average coefficient can also be used:

$$\langle D^{(PI)} \rangle = 8.385 \times 10^{-15} \text{ m}^2 \text{ s}^{-1}. \quad (9)$$

The sorption isotherm, which determines the relation between the RH ϕ and the moisture concentration c_m at saturation ($t \rightarrow \infty$) of the polyimide with the relative humidity ϕ of the ambient air (see figure 1b) was shown to follow a GAB (Guggenheim-Anderson-de-Boer) model (Bhargava et al. 2006). But here, for sake of simplicity, the following polynomial fit can be used:

$$c_m^{(PI)} = 54.452\phi^2 - 2.0016\phi, \text{ where } 0 \leq \phi \leq 1. \quad (10)$$

In-plane expansion measurements were performed to determine the evolution of the sample length with respect to the RH ϕ . The following hygroscopic in-plane strain was found:

$$\varepsilon^{(PI)hy}(c) = 8 \times 10^{-6} (10c - 0.2c^2). \quad (11)$$

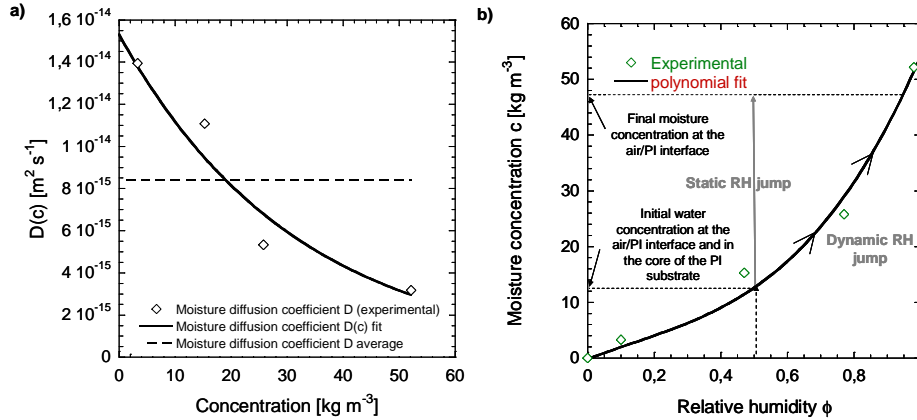


FIG. 1 – Evolution of the moisture diffusion coefficient for the Upilex S polyimide (a) and evolution of the moisture content at saturation as a function of the RH of the ambient air (b).

It was further assumed that there is neither moisture-induced expansions of the SiN_x coating nor moisture flux through the SiN_x coating, thus $D^{(SiNx)}=0$ and $c^{(SiNx)}=0$.

Materials' parameters needed for the simulations presented in the following are given in table 1.

4. Examples of numerical simulations

4.1 Relative humidity “jump” experiments

After SiN_x deposition at $T=200^{\circ}\text{C}$ the foils were cooled-down to room temperature and stored during several weeks in air atmosphere at a RH ϕ of approx. 50%. Rectangular samples of 55×6 mm² were then cut in these foils and placed in an environmental chamber equipped with a RH generator (VTI RH-200) controlled by a hygrometer (Ebro RHT 200). The samples were freely supported by two razor blades with a separation of 40 mm. At the center of the sample, the deflection δ of the point of coordinates $(x_1=0, x_2=(h_s+h_c)/2)$ was measured using a binocular lens (Olympus SZH). Isothermal RH jumps were carried out at 22°C . The chamber was set at 50% RH and the deflection of the sample was measured until moisture uptake equilibrium was reached (typically 2 h). The observed deflection was attributed to the cumulative effect of an intrinsic compressive stress in the coating layer, a thermal stress and a hygroscopic stress, respectively due to the mismatch of thermal and hygroscopic expansions between both materials (Dumont et al. 2007). The RH was then set to 95% (figure 2a) and the resulting change of deflection was measured during 7 hours. In figure 2b, the absolute value of the vertical $u_2(t)-u_2(t=0)$ displacement of the point $(x_1=0, x_2=(h_s+h_c)/2)$ is shown with respect to the time t . The initial deflection of the bilayer film was reduced progressively, due to the hygroscopic expansion of the PI substrate.

Young's modulus $E^{(PI)}$ [MPa]	5638.9-12.41(T-273.15)
Poisson's coefficient $\nu^{(PI)}$	0.25
CTE $\alpha^{(PI)}$ [K ⁻¹]	$(31.773-0.046674(T-273.15)+0.00026144(T-273.15)^2)\times 10^{-6}$
Intrinsic stress $\sigma^{\text{int}(PI)}$ [MPa]	-
Saturation moisture concentration c_m [kg m ⁻³]	cf. above
Moisture diffusion coefficient $D^{(PI)}$ [m ² s ⁻¹]	cf. above
Young's modulus $E^{(SiNx)}$ [MPa]	100000
Poisson's coefficient	0.264
CTE $\alpha^{(SiNx)}$ [K ⁻¹] (400 nm)	13.395×10^{-6}
Intrinsic stress [MPa] ($h_c=400$ nm)	-135.17
Saturation moisture concentration c_m [kg m ⁻³]	-
Moisture diffusion coefficient $D^{(SiNx)}$ [m ² s ⁻¹]	-

TABLE. 1 – Materials' data for the Upilex S polyimide substrate and the silicon nitride coating.

4.2 Geometry, mesh, initial and boundary conditions

For numerical simulation of isothermal ($T=22^{\circ}\text{C}$) RH jumps, due to the symmetry of the problem, one-half of the structure is modeled. A 2D simulation is performed and a plane strain assumption along \mathbf{e}_3 is made. The displacement boundary conditions are given on figure 3.

Quadratic rectangular elements were used for all unknown fields. The mesh size used for accurate results consisted of 2500 elements along the length, 3 elements through the coating layer thickness and 15 elements through the substrate layer thickness.

At time $t=0$, the moisture content is assumed homogeneous in the PI substrate and can be related to the sorption isotherm (see figure 1b).

During RH jump, the surface moisture content of the polyimide layer c_s is related to the RH of the ambient air and the sorption isotherm (figure 1b) of the polyimide: it is assumed that there is a boundary layer, which is fully saturated by the moisture, in the polyimide at the interface air/polyimide. Thus, we have $c_s=c_m$ (see figure 2b). A first case of static boundary conditions can be investigated. This means that the relative humidity jump is seen as being applied to the

boundaries of the sample at time $t=0$ (see figure 1b where the arrow “Static RH jumps” indicates the step seen by c_s with this first case). A second case of dynamic boundary conditions can also be studied. The arbitrary ΔRH jump profile imposed during the experiment (see figures 2a and 1b where the arrows “Dynamic RH jumps” indicate the path followed by c_s with this second case) has to be fitted with respect to the time by the following function $\phi(t)=A+B[1-\exp(-Ct)]$, where the coefficients A , B and C need to be identified for each experiments.

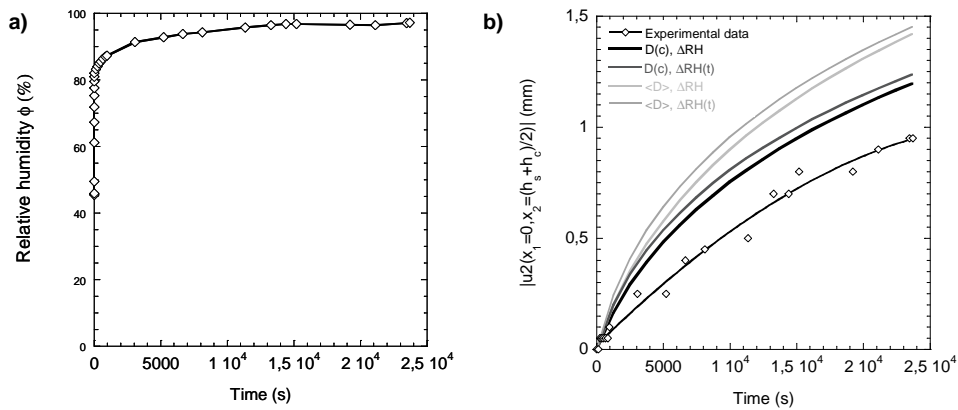


FIG. 2 – Evolution of the imposed RH with respect to the time (a) and evolution of the vertical displacement of the midplane point at the center of the bilayer structure (b).

Four different simulation cases were considered by combining static and time-evolving boundary conditions with either a constant average moisture diffusion coefficient, or a concentration-dependent moisture coefficient.

4.3 Results and comparison with experimental measurements

Figure 2b compares the experimental data with the 4 simulation cases. All scenarios reproduce the measured decay of the bilayer film deflection, a result of the progressive ingress of moisture in the polymer substrate. However, large differences are evident between the four simulation cases. It is evident that the simulation is more sensitive to the moisture diffusion problem (constant coefficient or concentration-dependent coefficient, the latter providing more accurate simulation), than it is to the type of boundary conditions (instantaneous vs. time-dependent RH change). One notices that most accurate simulation overestimates the measured displacement $u_2(t)-u_2(t=0)$. To explain this difference between simulation and experiment, one may invoke for instance the viscoelastic nature of the polyimide substrate, which has not been considered here.

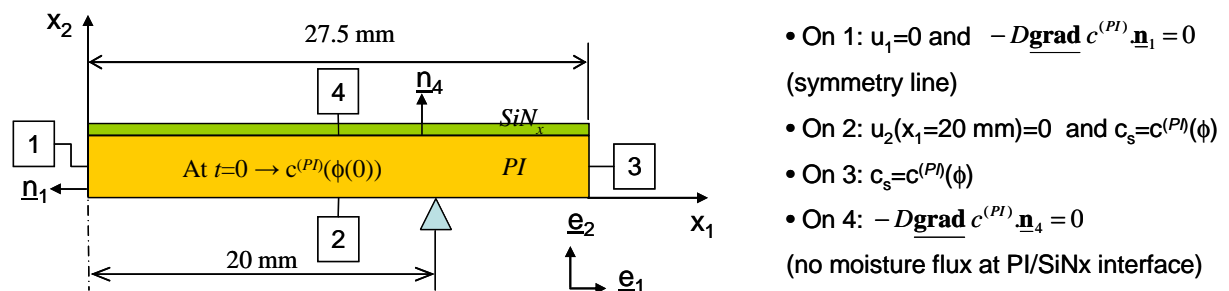


FIG. 3 – Schematic side view of the bilayer structure and of the boundary conditions for displacements and for moisture concentration.

5. Concluding remarks

A FEM framework was developed to simulate the hygrothermal stability of thin film/polymer composite structures, and applied to the case of SiN_x/PI films subjected to an isothermal RH

jump. The quality of the simulation was improved when accounting for a concentration-dependent moisture diffusion coefficient. To further improve the prediction, a detailed investigation of the influence of the viscoelasticity of the polyimide substrate is being currently performed.

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