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Citation for published version (APA):

Guan, Q., Laven, J., Bouten, P. C. P., & With, de, G. (2013). Subcritical crack growth in SiNx thin-film barriers studied by electro-mechanical two-point bending. *Journal of Applied Physics*, 113, 1-10. [213512].
<https://doi.org/10.1063/1.4809542>

DOI:

[10.1063/1.4809542](https://doi.org/10.1063/1.4809542)

Document status and date:

Published: 01/01/2013

Document Version:

Publisher's PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:

- A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
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Citation: [Journal of Applied Physics](#) **113**, 213512 (2013); doi: 10.1063/1.4809542

View online: <http://dx.doi.org/10.1063/1.4809542>

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Subcritical crack growth in SiN_x thin-film barriers studied by electro-mechanical two-point bending

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(Received 27 February 2013; accepted 20 May 2013; published online 5 June 2013)

Mechanical failure resulting from subcritical crack growth in the SiN_x inorganic barrier layer applied on a flexible multilayer structure was studied by an electro-mechanical two-point bending method. A 10 nm conducting tin-doped indium oxide layer was sputtered as an electrical probe to monitor the subcritical crack growth in the 150 nm dielectric SiN_x layer carried by a polyethylene naphthalate substrate. In the electro-mechanical two-point bending test, dynamic and static loads were applied to investigate the crack propagation in the barrier layer. As consequence of using two loading modes, the characteristic failure strain and failure time could be determined. The failure probability distribution of strain and lifetime under each loading condition was described by Weibull statistics. In this study, results from the tests in dynamic and static loading modes were linked by a power law description to determine the critical failure over a range of conditions. The fatigue parameter n from the power law reduces greatly from 70 to 31 upon correcting for internal strain. The testing method and analysis tool as described in the paper can be used to understand the limit of thin-film barriers in terms of their mechanical properties. © 2013 AIP Publishing LLC.

[<http://dx.doi.org/10.1063/1.4809542>]

I. INTRODUCTION

Flexible thin-film technology has great potential to be applied in the electronics industry since it allows electronic products to become thinner, lighter, more portable, and more compatible with low-cost roll-to-roll manufacturing processes.¹ Usually a flexible device is constructed by thin-film multilayers. For instance, in flexible organic light emitting diode (OLED), it is desirable to build a multilayer structure, in which the polymer substrate carries the whole assembly and provides the mechanical flexibility; the active layers perform the device functions and the encapsulation layers prevent the device from environmental degradation. Since functional layers in an electronic device are frequently sensitive to environmental factors, like oxygen and water vapor, inorganic thin-film barrier layers are required to protect the functional layers, due to the fact that the polymer substrate cannot provide sufficient suppression to the permeation.^{2–4}

In flexible multilayers, the inorganic barrier layer is brittle and known to be susceptible to mechanical deterioration. Even a single crack in a high quality barrier layer degrades the barrier quality seriously. Therefore, the mechanical integrity of the barrier layer is a crucial element in determining the lifetime of a flexible device. In recent years, there have been several investigations dealing with the mechanical failure of inorganic barriers.^{5–7} As reported in literature, stress-induced cracking is the dominant reason for mechanical failure in barrier layers and subsequent device degradation. The cracking phenomenon is influenced by many factors, such as defect size and location,^{5,7–9} internal stress,^{10,11} layer

thickness,^{12,13} and loading type.^{11,14} It is important to indicate that crack extension in barrier layers is greatly affected by environmental conditions, due to the fact that the brittle inorganic material is susceptible to environmentally assisted subcritical crack growth.¹⁵ In addition, the considerable hydro-thermal-mechanical contrast between the inorganic barrier layer and the polymer substrate also contributes to the cracking.

Analysis of mechanical performance of thin brittle films on a compliant substrate is mostly carried out by tensile tests in combination with optical observation to determine the cohesive and adhesive properties of the multilayers.^{10,12} Another method, which is called electro-fragmentation, combines optical and electrical measurements to observe mechanical failure of conductive inorganic films on a polymer substrate.¹³ In this method, the continuous electrical resistance measurement identifies the critical failure for unstable growing cracks. However, the electrical resistance measurement restrains the observation to conductive films. To detect mechanical failure of dielectric layers, a conducting layer such as amorphous graphite,⁶ ITO,¹⁶ or ITO-Ag¹⁷ is sputtered or deposited as an electrical probe. The influence of various electrical probes on the internal strain in the multilayers and the accuracy of the crack onset strain observation have also been investigated.¹⁷

The mechanical two-point bending method was first developed to study the strength of optical fibers and the fracture of the fiber was sensed by an acoustic detector.^{18–20} Recently this method was modified to characterize the fracture of thin conductive layers, such as ITO, on a polymer substrate by measuring its electrical resistance.²¹ The electro-mechanical two-point bending method has great advantages

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in determining critical failure strains for a number of reasons: (i) The sample has no clamping problem, such as slippage and stress gradient at the clamping jaws, and when the sample is under pure elastic deformation²¹ it has a well specified geometry; in this case, the applied strain at any location along the curvature can be accurately determined. (ii) The resistance measurement is global, which shows a fast response of the whole deforming sample; the optical measurement is a detailed observation which requires to observe the crack opening, so only local observation is possible; compared with the resistance measurement, the optical observation is a laborious undertaking and hardly achievable on a deforming curved sample. (iii) In this method, the failure criterion is defined as a specific level of electrical resistance increase; although the failure criterion is not accurately indicating the state of cracks, such as the population, location, and geometry, significant resistance increase can be used as a marker for well-developed cracks without verifying these uncertain parameters; using the resistance as a probe, the two-point bending method produces data on crack formation in such a consistent way that Weibull statistical analysis becomes accessible to study the failure probability distribution.

For this research an electro-mechanical two-point bending device was developed to study the mechanical reliability of the inorganic SiN_x barrier layer on a polyethylene naphthalate (PEN) foil. A thin conducting ITO layer on top of the barrier layer acts as an electrical probe. During the bending test crack formation in the barrier layer was effectively detected by monitoring the electrical resistance of this ITO probe. The objective of this work is to predict the failure and estimate the lifetime of the inorganic barrier layer, which is closely related to the overall functional failure time of flexible devices. The testing method and analysis tool in this paper can be used further to understand the limit of thin-film barriers in term of their mechanical properties and thus to optimize manufacturing parameters leading to improved mechanical durability in specific applications.

II. THEORY

A. Thin-film fracture mechanics for crack channelling

In the present study, we are dealing with the fracture mechanics of a thin brittle film of thickness h containing a defect of characteristic size c . For crack initiation and propagation in thin brittle films, two regimes are recognized: (i) For $c \ll h$, the fracture mechanics for bulk materials is applied; under loading a crack of length c locally amplifies the applied stress σ , resulting in a stress intensity factor given by

$$K_I = Y\sigma\sqrt{c}, \quad (1)$$

where Y is a geometry factor for the crack shape;²² (ii) for $c \gg h$ the crack channelling model is applied; instead of crack length, the film thickness h becomes the dominant element which influences the stress intensity factor, i.e.,

$$K_I = Z\sigma\sqrt{h}, \quad (2)$$

where Z is a geometry factor that depends on the elastic mismatch parameters between the thin brittle film and the substrate.²³ An estimate for the minimum crack size for which the crack channelling model can be applied was reported in the literature.⁷ It tells that for significant elastic mismatches, the crack is channelling at $c > 40h$. This condition is frequently fulfilled for inorganic films on polymer substrates. It is known that when the stress intensity factor K_I is larger than the fracture toughness K_{Ic} , the material fails instantaneously. For the situation $K_I < K_{Ic}$ subcritical crack growth is known to be widely involved in the failure of brittle materials, which usually contain small flaws and are susceptible to stress corrosion. Various kinetics models describing the relationship between the subcritical crack growth rate and the stress intensity factor have been proposed in the literature.^{24–28} In the present study, a power law is considered, according to which the crack growth velocity is

$$v = AK_I^n. \quad (3)$$

Here the fatigue parameter n represents the stress corrosion susceptibility in terms of the sensitivity of the involved chemical reaction to the stress. The parameter A depends on the environment and the material.

In a dynamic fatigue test, the sample is loaded at a constant stress rate $\dot{\sigma}$, so that the stress becomes

$$\sigma = \dot{\sigma}t, \quad (4)$$

where t represents the time. If a brittle film is susceptible to subcritical crack growth, a crack with length c extends during dynamic loading, and the crack growth rate can be described by combining Eqs. (2)–(4)

$$v = \frac{dc}{dt} = AZ^n \dot{\sigma}^n t^n \sqrt{h}^n. \quad (5)$$

In the present analysis, we adopt a crack extending over a distance b , normal to the tensile direction as failure criterion. Assuming that the crack starts at one edge of the sample, this results in

$$\sigma_f = \left[\frac{b(n+1)}{AZ^n \sqrt{h}^n} \right]^{\frac{1}{n+1}} \dot{\sigma}^{\frac{1}{n+1}}, \quad (6)$$

from which we can see that the measured failure stress σ_f depends on the stress rate during loading. The failure time during a dynamic fatigue test t_f^d is derived from the failure stress and the loading stress rate and is given by

$$t_f^d \cong B(n+1)\sigma_f^{-n}, \quad (7)$$

where

$$B = \frac{b}{AZ^n \sqrt{h}^n} \quad (8)$$

which is slightly different from the failure time estimation for bulk materials.²²

In a static fatigue test, a constant loading stress is applied. Combining Eq. (2) and Eq. (3), the crack growth rate under an applied stress σ_s is given by

$$v = \frac{dc}{dt} = AZ^n \sigma_s^n \sqrt{h}^n. \quad (9)$$

Using the same failure criterion as in the dynamic fatigue test, the failure time of static fatigue test evolves to

$$t_f^s = \frac{b}{AZ^n \sqrt{h}^n} \frac{1}{\sigma_s^n} \cong B \sigma_s^{-n}, \quad (10)$$

where B is the same as in Eq. (8). Therefore, by comparing the expressions for dynamic failure time in Eq. (7) and static failure time in Eq. (10), these two types of failure test can be related via the fatigue parameter n . When the applied stresses are the same, $\sigma_f = \sigma_s$, the failure times involved are related resulting to

$$t_f^s(\sigma_s) = \frac{t_f^d(\sigma_f)}{n+1}, \quad (11)$$

which shows that a characteristic failure stress, thus failure time in the dynamic test, can be translated into an equivalent static fatigue time at a specific applied stress.

B. Weibull statistics

Weibull statistics is widely applied to describe the fracture probability of brittle materials caused by intrinsic defect variability.²² For thin brittle films, suppose that a number of defects on a characteristic area A_0 exists. Then according to Weibull statistics the failure probability P of a thin brittle film of area A , for an applied stress σ , is given by

$$P = 1 - \exp \left[-\frac{A}{A_0} \left(\frac{\sigma - \sigma_u}{\sigma_0} \right)^m \right], \quad (12)$$

where m is the Weibull modulus, which describes the homogeneity of the flaw distribution; σ_0 is a characteristic failure stress; and σ_u is an offset stress above which the crack extension mechanisms start to be active. In practical applications, $\sigma_u = 0$ is frequently used, which means no offset is assumed and a two-parameter Weibull distribution is obtained. In the main part of the present study, we will follow this approach. In Sec. IV E, the internal stress in the barrier layer will be considered to be the cause for an offset, and its consequence will be analyzed. To estimate the Weibull parameters, a linear regression method is used. Assuming $\sigma_u = 0$ in Eq. (12), rearranging and taking logarithms twice yields

$$\ln \left[\ln \left(\frac{1}{1-P} \right) \right] = m \ln(\sigma) - m \ln(\sigma_0) + \ln \left(\frac{A}{A_0} \right). \quad (13)$$

This relation can be used to determine the Weibull modulus by plotting $\ln \left[\ln \left(\frac{1}{1-P} \right) \right]$ versus $\ln(\sigma)$ on linear scales for a series of samples with identical shape. The slope of the linear fit yields the Weibull modulus m , and from the intercept the characteristic strength σ_0 at a failure probability of 63% is

obtained. Clearly, to apply this approach to a set of experimental data an estimate for P must first be made. Various estimators for the failure probability are known.²² However, the empirical estimator

$$P_i = \frac{i - 0.5}{N} \quad (14)$$

yields the least biased result with minimum variance in a least-squares fitting of the stress data.²⁹ In Eq. (14), i is the rank of the tested specimen and N is the total number of tested specimens. In a similar way, a Weibull distribution for analyzing a static failure test can be represented as

$$\ln \left[\ln \left(\frac{1}{1-P} \right) \right] = m' \ln(t) - m' \ln(t_0) + \ln \left(\frac{A}{A_0} \right), \quad (15)$$

where the failure probability is represented as a function of the static failure time t , t_0 is a characteristic failure time, and m' is the corresponding Weibull modulus. Here we have two Weibull moduli, i.e., m from plotting the failure stress in a dynamic fatigue test and m' from plotting the failure time in a static fatigue test. According to the subcritical crack growth theory described in Sec. II A, when a stress applied in a static test σ_s is identical to the failure stress σ_f in a dynamic test, i.e., $\sigma_f = \sigma_s$, substituting Eq. (10) into the Weibull distribution for static failure (Eq. (15)) leads to

$$m' = \frac{m}{n}, \quad (16)$$

which shows that the Weibull moduli for dynamic and static fatigue failure are connected via the fatigue parameter n .

III. MATERIALS AND EXPERIMENTAL METHOD

A. Sample preparation

A non-stoichiometric SiN_x layer with a typical thickness of 150 nm was deposited by low-temperature plasma-enhanced chemical vapour deposition on a PEN substrate. The PEN substrate had a size of approximately $150 \times 150 \text{ mm}^2$ with a thickness of 125 μm and was laminated on a glass carrier during the deposition process. In the literature,⁶ a 10 nm thin amorphous graphite layer was found to accurately reproduce the cracking of the dielectric SiN_x film. However, in this paper a thin ITO layer with a nominal thickness of 10 nm is used as an electrical probe for which a much higher sputtering deposition rate is possible. The sheet resistance of the conducting ITO film was about 1000 Ω/square . The multi-layered structure (PEN substrate and SiN_x -ITO layers) was kindly provided by the Holst Centre, Eindhoven.

Several aspects of the ITO film are worth mentioning. First, the observed critical failure strain¹⁶ of this 10 nm ITO film is around 1.5%, which is much higher than that of the SiN_x film (from 0.7% to 0.8%). Second, in the manufacturing of this polymer- SiN_x -ITO structure, the highest temperature is reached during the deposition of the SiN_x layer, which is the main cause of the internal strain in the structure. The ITO layer is sputtered at room temperature, and thus the thermal load is minimal. Besides, there are no clear changes in

the sample curvature due to ITO deposition. Therefore, the expectation is that the internal strain in the SiN_x film is not significantly influenced by the deposition of ITO layer. Last but not least, the energy release rate in the fracture of SiN_x layer will not change by adding the ITO layer on top, as has been addressed in the paper from Vellinga *et al.*¹⁶ All in all, the 10 nm ITO layer has proved to be an effective probe to monitor the fracture failure of the SiN_x layer beneath.

After deposition, the sheet was released from the glass carrier and cut into strips of typical size $150 \times 10 \text{ mm}^2$. The sample strips were subsequently numbered and stored at 21°C and 50% relative humidity for more than 15 days before starting the measurements. In order to eliminate the bias in analyzing samples caused by manufacturing variability, sample strips from all sheets were taken in a fully randomized way to improve the reliability of the statistical analysis. In practice, 16 polymer sheets were coated in one deposition series, and 192 samples were cut and numbered from these sheets. A random sample selection procedure was used, and samples were divided in 8 batches. Each batch containing 24 samples was prepared to test at one specific test condition, from which 22 samples were tested at each condition.

B. Electro-mechanical two-point bending test

Since an inorganic brittle layer behaves linearly elastic, the stress σ and strain ϵ are linearly related by Hooke's law as $\sigma = \epsilon E$, where E is the Young's modulus for the brittle layer. In the two-point bending geometry, it is more convenient and accurate to determine the strain than the stress considering the device property and the time-dependent behavior of the polymer substrate. Due to the fact that the effective elastic modulus E of the polymer is a function of time, the stress in the polymer substrate is time-dependent as well, typically decreasing 3% per decade in time. Upon bending the stress in the polymer substrate along the bending curve is slightly but uniformly decreasing with time due to the viscoelasticity, but the shape of the bending curve is well preserved. The shape in the polymer substrate dictates the strain in the SiN_x film, which is also well preserved and independent of time. Moreover, an inorganic brittle layer like SiN_x behaves linearly elastic, and this allows us to describe the applied loading condition in terms of strain and strain rate instead of by stress and stress rate. The equations for Weibull distributions can be rewritten accordingly while preserving the values of m , m' , and n .

1. Geometry

The geometry of the two-point bending device employed in the present study is shown in Fig. 1. Upon two-point bending, a tensile strain is induced in the outer layer of the sample and a compressive strain at the inside. At the position of the neutral line, no bending strain is induced. For a symmetrical sample, the neutral line is located in the middle of the sample.

The sample strip is mounted between two parallel plates with the brittle layers (SiN_x and ITO) outside, thus tensile

FIG. 1. Geometry of the electro-mechanical two-point bending device.

loaded. The maximum tensile strain is related to the smallest local curvature radius R and is given by

$$\epsilon = f_n \frac{h}{2R}, \quad (17)$$

where h is the thickness of the sample strip and the factor f_n corrects for the position of the neutral line in the sample ($f_n = 1$ when the neutral line is in the middle). If the substrate is elastic, the smallest curvature radius R is uniquely related to the distance between the neutral lines L in the specimen where the specimen is in contact with the loading plates, as shown in Fig. 1

$$R = \frac{L}{2C_b}, \quad (18)$$

where C_b is an elastic constant²¹ as $C_b = 1.1985$. From Eqs. (17) and (18), we derive for the maximum local strain in the two-point bending

$$\epsilon = f_n C_b \frac{h}{L}. \quad (19)$$

Note that the distance between the neutral lines L slightly differs from the distance between the two parallel plates L_0 since the sample thickness and the location of the neutral line need to be considered, so that

$$L = L_0 - 2(1 - f_n)h. \quad (20)$$

In the bending test, the failure strain of the brittle barrier layer will be derived from the distance L at which the failure event occurs. Two basic test modes are used: (i) In dynamic fatigue, the plates move at a specific speed, and the distance of the plates at failure indicating the failure strain ϵ_f^d is determined. (ii) In static fatigue, a certain strain is applied, and the time t_f^s required for the failure to occur is determined. The average failure strain ϵ_f^d and failure time t_f^s depend on the strain rate $\dot{\epsilon}$ (Eq. (6)) and the applied strain ϵ_s (Eq. (10)), respectively.

2. Implementation

In this study, we utilize a newly designed two-point bending equipment that has the possibility to program the

velocity of the parallel plates. For instance, in order to apply a constant strain rate $\dot{\epsilon}$ in a dynamic fatigue test, velocity control is used in which a voltage profile is sent to the motor and the position profile is measured. In this mode the maximum speed of the motor is 20 mm/s, corresponding to a 10 V supply voltage to the motor. In a static fatigue test, position control is preferred since here the sample has to be loaded quickly to a pre-defined distance, thus a pre-defined applied strain ϵ_s . In the position controlled mode, the standard velocity used to bring the parallel plate to a specified position is 33.33 mm/s. The fatigue tests can be terminated in two ways: one is by a pre-defined endpoint in time, and the other is when the measured resistance exceeds a pre-defined limit $R_{\max}$. At the end of the test, one plate returns to the start position, which is called the unloading step. During the loading and subsequent unloading step in the test, time, position, and sample resistance are recorded at defined time intervals.

In the dynamic fatigue test, a velocity is given as a function of time or plate distance as shown in Fig. 2. Due to the velocity limitation of the voltage controlled motor (20 mm/s), a constant velocity might be required before the profile starts, which is illustrated in the velocity profile for an imposed strain rate of 10%/min in Fig. 2(a). Parameters used with the present samples are foil thickness $h = 125 \mu\text{m}$, initial plate distance $L_0 = 60 \text{ mm}$, and strain rates of 0.5%/min, 2%/min and 10%/min, respectively. Figure 2(b) clearly shows that at a strain rate of 10%/min the targeted profile is

FIG. 2. Velocity profiles for dynamic fatigue test at various strain rates (a) as a function of time and (b) as a function of distance.

realized at and below a plate distance of 42.4 mm, which is equivalent to an applied strain of 0.35% for a 125 μm foil. The failure strain of 150 nm SiN_x layers on PEN substrate is expected to be 0.70%–0.80%. Therefore, reliable dynamic fatigue data are obtained where the displacement profile to be used starts significantly below the displacement associated with the failure strain. Based on this discussion, a strain rate of 10%/min is used as the maximum strain rate in the dynamic fatigue test.

In the present test, the resistance of the conductive layer on the sample surface is used to monitor the evolution of cracks. Figure 3 shows a typical resistance-strain curve in a dynamic fatigue test. The sample was loaded at a strain rate of 0.5%/min, and a significant resistance increase was observed due to fracture in the SiN_x layer (Stage 1–2). Upon unloading (Stage 3) a hysteresis in resistance is observed until a strain at which the cracks close and thus the ITO layer regains its conductivity (Stage 4). Finally the resistance recovers to the initial level (Stage 5). Upon reloading, the resistance curve “remembers” its track from the last cycle, and there are two remarkable regimes to be distinguished: (i) At Stage 6–7 where the applied strain is less than or equal to the internal strain, i.e., $\epsilon \leq \epsilon_{\text{internal}}$, the existing cracks that are closed by the internal strain open gradually with increasing applied strain. (ii) At Stage 8 where the applied strain is larger than the internal strain, i.e., $\epsilon > \epsilon_{\text{internal}}$, all existing cracks are open, and the increase in the applied strain at this level causes no further crack growth until it reaches the value closed to the failure strain. Consequently, the reloading resistance curve can be used to determine the internal strain in the SiN_x film, as will be discussed more in detail later.

To characterize the failure in the barrier layer, a specific relative increase of the electrical resistance has been used as a failure criterion. Because the level chosen is somewhat arbitrary, three levels of resistance increase were considered to learn the effect that the level chosen has on the results. The relative resistance increase levels $\Delta R/R$ considered are 2%, 10%, and 20%, and the corresponding strain and time are defined as failure strain and failure time. For the presently used sample size these levels correspond to the increase in sheet resistance $\Delta R/R_{sq}$ of 0.28, 1.4, and 2.8, respectively.

FIG. 3. Resistance-strain curve in a dynamic fatigue test by the two-point bending method: (1–2) loading; (3–5) unloading; (6–8) reloading.

IV. RESULTS AND DISCUSSION

A. Electrical resistance increase in dynamic and static fatigue tests

In the present study, three different strain rates, i.e., 10%/min, 2%/min, and 0.5%/min, were applied to perform the dynamic fatigue tests. The loading strain profiles as a function of time are shown in Fig. 4(a). As aforementioned, the two-point bending test can be terminated in two different ways, and here the maximum resistance $R_{\max} = 28 \text{ k}\Omega$ is used to stop the test in consideration of the initial resistance of the conductive ITO layer $R_0 \cong 16 \text{ k}\Omega$. Figure 4(b) shows resistance responses in dynamic fatigue tests at various strain rates, which clearly indicates a strain rate dependent behavior in the cracking of the SiN_x barrier. Note that upon loading, the resistance at a higher strain rate tends to access a higher resistance increase level although the same terminal resistance $R_{\max}$ was programmed in all cases. The reason is that upon reaching $R_{\max}$ the cracks that are initiated do not stop growing at the exact moment of changing the settings of the device, especially for the test at high strain rate. As described in Eq. (6), the failure strain is strain rate dependent, which explains why in Fig. 4(b) the strain reaches a higher value when the sample is loaded at a high strain rate using the same terminal resistance $R_{\max}$. In the power law description in Eq. (3) the crack growth speed is closely

FIG. 4. (a) The strain profiles at various strain rates and (b) the corresponding electrical resistance profiles as a function of strain in dynamic fatigue tests.

related to the applied strain which represents the stress intensity factor K_I . Therefore, cracks exposed to a higher strain grow much faster and even continue growing for some time upon subsequent unloading. This is also observed in the resistance curve of 10%/min in Fig. 4(b) where the resistance curve bends to low strain direction when it approaches the plateau indicating the increase of resistance upon unloading.

In a static fatigue test, the sample is loaded at a constant applied strain, and the loading time and the evolution of electrical resistance are recorded. Figure 5 shows examples of static strain profiles and the corresponding resistance curves at various levels of applied strain, from which failure times at failure criteria $\Delta R/R = 20\%$, $\Delta R/R = 10\%$, and $\Delta R/R = 2\%$ were extracted. Figure 5(b) shows that the failure time varies over four decades while the strain window varies only about 0.08% (from 0.708% to 0.783%). This provides us important information about the critical strain window for the SiN_x layer.

B. Weibull statistics on subcritical crack growth in dynamic and static fatigue tests

Fig. 6 shows the Weibull distribution of failure strains of the SiN_x barrier using various strain rates in the dynamic fatigue tests. The failure strains given in this figure were extracted from dynamic fatigue tests based on a failure criterion of $\Delta R/R = 20\%$. The data are represented in terms of $\ln[\ln(\frac{1}{1-P})]$ as a function of $\ln(\epsilon)$ in Weibull plots according

FIG. 5. (a) The strain profiles at various applied strains and (b) the corresponding electrical resistance profiles as a function of time in static fatigue tests.

FIG. 6. Weibull statistical distribution for the dynamic fatigue result of SiN_x barrier at a failure criterion of $\Delta R/R = 20\%$.

to Eq. (13). The failure probability is obtained from Eq. (14), and the lines through the data are fitted with a common value for the Weibull slope m . The characteristic failure strain is extracted at failure probability of 63%. From the three characteristic failure strains obtained for these three different strain rates, a value of 64 for the fatigue parameter n was derived, according to the relation between failure strain ϵ_f and strain rate $\dot{\epsilon}$ given by

$$\frac{\dot{\epsilon}_1}{\dot{\epsilon}_2} = \left(\frac{\epsilon_{f1}}{\epsilon_{f2}} \right)^{n+1}. \quad (21)$$

One might be inclined to distinguish two regimes in the Weibull plot, due to, respectively, surface and bulk defects. However, even for materials where fractography can be done without any problems, such as glass, it appears that identification of the type of defect from the Weibull graph is impossible.³⁰ Moreover, in this case failure is dominated by crack channelling. Hence we consider the plot to show a single distribution. In Table I the fatigue parameter and Weibull modulus results are given for three failure criteria, defined at relative resistance increases of 2%, 10%, and 20%. At each failure criterion, it is possible to fit the Weibull plots for the three different strain rates with a common Weibull modulus. In view of the accuracy of the values of Weibull modulus it can be concluded that its value is not sensitive to the exact level of the resistance increase. Therefore, the conclusions regarding failure are independent of the somewhat arbitrarily chosen resistance criterion.

As shown in Fig. 7, the failure strain depends on the strain rate, and their relation can be described by a power law in accordance with Eq. (7) for all failure criteria. The high value of the fatigue parameter indicates a low influence of the strain rate on the failure strain, which is also observed

TABLE I. Weibull modulus m and fatigue parameter n values for dynamic fatigue of the SiN_x barrier.

Failure criteria $\Delta R/R(\%)$	m	n	Predicted m'
20	77 ± 17	64	1.2 ± 0.3
10	76 ± 16	68	1.1 ± 0.2
2	69 ± 15	66	1.1 ± 0.2

FIG. 7. The characteristic failure strain at various strain rates in dynamic fatigue tests.

in Fig. 7. The failure strain reduces by less than 5% for a strain rate reduction of 1.5 decades, and it is independent of the chosen resistance level.

As an application of the power law, Eq. (11) relates the dynamic to the static failure through a common fatigue parameter n . The dynamic failure strain at a certain strain rate can be transformed to the corresponding static failure time at a particular failure strain. In Fig. 8 the three open points are dynamic fatigue results at strain rates of 0.5%/min, 2%/min, and 10%/min. The corresponding failure strains are 0.779%, 0.795%, and 0.815%, respectively. When these three strain levels are applied in static fatigue tests, the failure time are estimated according to Eq. (11) as 1.437 s, 0.367 s, and 0.075 s, respectively. For the prediction of the lifetime of a barrier, much longer failure times should be considered, thus lower failure strains. According to the relation between the static failure time t_f^s and applied strain ϵ_s

$$\frac{t_{f1}^s}{t_{f2}^s} = \left(\frac{\epsilon_{s2}}{\epsilon_{s1}} \right)^n. \quad (22)$$

A complementary series of four static tests were carried out, the results of which are represented by the four solid

FIG. 8. Targeted static failure as based on the dynamic failure results.

FIG. 9. Weibull statistical distribution for the static fatigue result of SiN_x barrier at a failure criterion of $\Delta R/R = 20\%$.

points in Fig. 8. The intention is to estimate the strain levels to be applied in the static fatigue tests using the dynamic fatigue data. The boundary conditions of the static fatigue tests are that the shortest static fatigue test is made to be comparable to the slowest dynamic fatigue test, and the longest static test is about 1000 s. The resulted applied strains are 0.783%, 0.762%, 0.734%, and 0.708% in static fatigue tests. To perform the static fatigue tests, one can validate the estimate of the failure based on the power law. More importantly, it leads to a great expansion in the failure strain window and failure time scale.

At each failure criterion failure times were analyzed by Weibull statistics. Fig. 9 shows the Weibull distribution of the data from the static fatigue tests at the failure criterion of $\Delta R/R = 20\%$. The failure at high strains, like 0.783% and 0.762%, yields higher slopes, thus higher Weibull moduli, in the Weibull plot than that at low strains. Probably, at low applied strains, the failure is influenced by both the time-dependent behaviour of the subcritical crack growth in the SiN_x layer and the visco-elastic behavior of the polymer substrate. Further study is still needed to validate the explanation of this phenomenon.

In a similar way as in the dynamic fatigue analysis, the characteristic failure times at various failure criteria were extracted from the static Weibull distributions at a failure probability of 63%, and results are shown in Fig. 10.

FIG. 10. The characteristic static failure strain under various strain rates.

Depending on the failure criterion, the static failure time at the strain window from 0.783% to 0.708% varies from about 1 s to about 1000 s.

C. Lifetime and mechanical reliability of the SiN_x barrier layer

In Fig. 11 the results of static fatigue tests are plotted together with those of the dynamic tests. In order to plot them both along the same time scale, dynamic failure times were converted in expected static equivalents, according to Eq. (11). By the combination of dynamic and static failure, failure times over a much larger range, more than four decades, are realized. The facts that all data for each individual failure criterion in Fig. 11 fall on a single curve and that the failure strain and failure time show a power law behavior supports the validity of the model description for subcritical crack growth in thin-film barrier layers. For industrial applications, the lifetime prediction will be on the time scale of 10^9 s. This is definitely out of the range of the present mechanical two-point bending test and, thus, has to be made based on an extrapolation of the tested range. However, the combination of dynamic and static data as performed in the present study will drastically increase the reliability of the lifetime prediction.

D. Weibull modulus and fatigue parameter

By least-squares fitting to the dynamic Weibull data and using Eqs. (13) and (21), the Weibull modulus m and fatigue parameter n for dynamic fatigue were determined as shown in Table I. Similarly, the static Weibull modulus m' and fatigue parameter n were determined by using Eqs. (15) and (22), and the results are shown in Table II. Due to the fact that we observed a difference in the slope of Weibull distribution at high and low strains in the static fatigue tests shown in Fig. 9, the data at high strains (0.783% and 0.762%) and low strains (0.734% and 0.708%) were fitted separately so as to extract Weibull moduli m'_1 and m'_2 , respectively.

It should be noted that the Weibull moduli obtained in the dynamic fatigue tests are fairly high since for defect

FIG. 11. Failure strain and static failure time of SiN_x barrier layer. The dynamic failure results were converted in static ones according to Eq. (11).

TABLE II. Weibull modulus m and fatigue parameter n values for static fatigue of the SiN_x barrier.

Failure criteria $\Delta R/R$ (%)	m'_1	m'_2	n
20	1.1 ± 0.2	0.6 ± 0.1	72
10	1.1 ± 0.2	0.7 ± 0.1	68
2	1.1 ± 0.2	0.8 ± 0.2	61

populations mostly $m < 20$ is obtained. Only for highly deterministic failure mechanisms like cracking channelling in this case, high Weibull moduli are obtained. The dynamic Weibull modulus m represents the width of the failure strain distribution, while the static Weibull modulus m' describes the width of the failure time distribution and the relation between m and m' is given by Eq. (16). From the m values in Table I, predictions of m' were made by using Eq. (16). These values, also shown in Table I, match excellently with the values m'_1 extracted from the sets of data in Table II. The conclusion is that the statistics of static and dynamic measurements match well, and it suggests a single mechanism over the whole failure strain or failure time domain. However, the results from m'_2 suggest a transition in the statistical variation of failure at a static failure time of the order of the magnitude 10^2 s. It suggests another mechanism or at least a combination of another mechanism in failure. This will be a subject of future research.

In both dynamic and static fatigue tests, the fatigue parameter n yield high values between 61 and 72. As we known, a high fatigue parameter indicates a low sensitivity for environmental effects and high mechanical reliability, which tells that SiN_x can perform well as a barrier layer from a mechanical point of view.

E. The internal strain and intrinsic strain in the SiN_x layer

So far a two-parameter Weibull statistical analysis was performed in this study, which means we assumed that the value of the offset strain ϵ_u is 0. However, there is a built-in compressive internal strain in the barrier layer. Actually, the built-in internal strain is the main reason for $\sigma_u \neq 0$, and it plays an important role in determining the failure strain level and fatigue parameter n .

To investigate the effect of an internal strain on the failure of the barrier layer, we define

$$\epsilon_{\text{exter}} = \epsilon_{\text{intrin}} + \epsilon_{\text{inter}}. \quad (23)$$

As described in Sec. III B 2, Fig. 3 provides a typical resistance-strain curve to analyze the external, internal, and intrinsic strains. Upon loading, a significant resistance increase is observed due to crack formation in the SiN_x layer, and the external failure strain is determined at a specific resistance increase level. A hysteresis in resistance is observed in the unloading stage due to the fact that the cracks formed during loading remain open as long as the applied strain is larger than the compressive internal strain in the barrier. When the strain reduces to equal to and below the level of the internal strain, cracks close and the resistance drops

to the initial level. Upon reloading, the curve does not retrace the unloading curve as expected, but one must realize that the unloading curve might be affected by many extra factors, such as the local deformation due to the stress concentration at the crack tips and settling of the local fracture surface of the thin ITO. Moreover, crack closure might behave differently from crack opening due to the time-dependent behavior of the polymer substrate. Hence, the reloading curve using the same speed as for the initial loading is more suitable to determine the internal compressive strain in the SiN_x layer by observing opening of the existing cracks formed during the loading.

Figure 12 shows the determination of external, internal, and intrinsic strains in the SiN_x barrier layer based on the above method. The external strain indicates the characteristic failure strain derived from the two-parameter Weibull distribution at the failure criterion of $\Delta R/R = 20\%$, from which a strain-rate dependent behavior is observed, as discussed in Sec. IV B. The internal strain is determined by the average strain value on the reloading curve at the same failure criterion $\Delta R/R = 20\%$. Fig. 12 suggests for the internal strain an absence of strain rate dependence. This is what can be expected due to the fact that the strain-rate dependent behavior represents the subcritical crack growth in the barrier layer. Once a crack develops, the barrier layer loses its local cohesion. The internal compressive strain can close the opening cracks to restore the conductivity but not the mechanical integrity. Therefore, no strain-rate dependence is expected for the strain to reopen the cracks during reloading. The internal strain in this case is $0.47\% \pm 0.02\%$. Vellinga *et al.*¹⁶ performed tensile tests on another batch of the same barrier-polymer combination and found an internal compressive strain of $0.45\% \pm 0.05\%$ in the barrier, which is very close to the value here.

The intrinsic strain at failure is determined by subtracting the internal strain from the imposed external strain in the experiment for each individual test. The obtained strains are then analyzed by Weibull statistics resulting in the intrinsic failure data plotted in Fig. 12. The line through the intrinsic strains is parallel to the one through the external strains. The

FIG. 12. Determination of the external strain (imposed strain in the experiment at failure), the internal strain (the built-in compressive strain in the barrier), and the intrinsic strain (the real failure strain of the barrier).

difference in strain between them is constantly at 0.47% which represents well the internal strain determined on the reloading curve. The scatter in the internal strain at low strain rate may be due to the local variation caused by the deposition process of the barrier, which induces negligible local variation in the intrinsic failure strain.

The fatigue parameter n was found to be 64 in dynamic failure and 72 in static failure at the failure criterion of $\Delta R/R = 20\%$ (Tables I and II). After subtracting the internal strain, the fatigue parameter n in both dynamic and static failure becomes 31. This is expected since the fatigue parameter is a material inherent property, which should be identical in dynamic and static failure. When designing a barrier layer, it is clear that a built-in internal strain can not only influence the critical failure strain in view of its mechanical integrity but also its sensitivity to the environmental effect on subcritical crack growth. In this application, on one hand, a built-in compressive internal strain (0.47%) in the SiN_x barrier layer strengthens the layer under tensile load. On the other hand, it also enhances the resistivity of the barrier layer to the environmentally assisted cracking, thus its mechanical stability.

V. CONCLUSION

An electro-mechanical two-point bending device is used to study the mechanical failure caused by subcritical crack growth in a SiN_x barrier layer. The well-controlled loading rates and plate distances achieved with the two-point bending device made a series of systematic dynamic and static fatigue tests possible. Failure was detected by monitoring the electrical resistance of a 10 nm ITO probe. The results were analyzed by the Weibull statistical distribution, and the dynamic and static failure was correlated by a power law description to represent the failure over a large strain domain and timescale.

As a result, failure times over more than four decades were realized, and the failure strain and failure time show a power law behavior, which supports the validity of the theoretical description for subcritical crack growth in thin-film barrier layers. A prediction on the failure on the large time scale could be made to fulfill the industrial need by making an extrapolation based on the tested range.

More importantly, a built-in internal strain is beneficial for both the mechanical reliability and the stability of the barrier layer. First, the failure strain in tensile loading is raised by the compressive internal strain in the barrier layer. Moreover, a high fatigue parameter n results by a built-in compressive internal strain, which increases the value from 31 to 70. Thus the compressive strain enhanced the resistivity of the barrier to the environmental degradation and could perform better as a diffusion barrier in hostile environmental conditions.

ACKNOWLEDGMENTS

This research was carried out under project number MC7.07299 in the framework of the Research Program of the Materials innovation institute (M2i) (www.m2i.nl). The authors acknowledge Ferdie van Assche (Holst Centre) for the sample preparation.

- ¹G. P. Crawford, *Flexible Flat Panel Displays*, edited by G. P. Crawford (Wiley, Chichester, 2005).
- ²J. Lewis, *Mater. Today* **9**, 38 (2006).
- ³F. Papadimitrakopoulos, X.-M. Zhang, and K. Higginson, *IEEE J. Sel. Top. Quantum Electron.* **4**, 49 (1998).
- ⁴Y. F. Liew, H. Aziz, N. X. Hu, H. S. O. Chan, G. Xu, and Z. Popovic, *Appl. Phys. Lett.* **77**, 2650 (2000).
- ⁵Y. Leterrier, *Prog. Mater. Sci.* **48**, 1 (2003).
- ⁶A. Pinyol, B. Meylan, D. Gilliéron, V. Mewani, Y. Leterrier, and J.-A. E. Månson, *Thin Solid Films* **517**, 2000 (2009).
- ⁷J. M. Ambrico and M. R. Begley, *Thin Solid Films* **419**, 144 (2002).
- ⁸J. Andersons and Y. Leterrier, *Probab. Eng. Mech.* **20**, 103 (2005).
- ⁹Y. Leterrier, D. Pellaton, D. Mendels, R. Glauser, J. Andersons, and J.-A. E. Månson, *J. Mater. Sci.* **36**, 2213 (2001).
- ¹⁰Y. Leterrier, L. Boogh, J. Andersons, and J.-A. E. Månson, *J. Polym. Sci., Part B: Polym. Phys.* **35**, 1449 (1997).
- ¹¹Y. Leterrier, A. Mottet, N. Bouquet, D. Gilliéron, P. Dumont, A. Pinyol, L. Lalande, J. Waller, and J.-A. E. Månson, *Thin Solid Films* **519**, 1729 (2010).
- ¹²Y. Leterrier, J. Andersons, Y. Pitton, and J.-A. E. Månson, *J. Polym. Sci., Part B: Polym. Phys.* **35**, 1463 (1997).
- ¹³D. R. Cairns, R. P. Witte, D. K. Sparacin, S. M. Sachsman, D. C. Paine, G. P. Crawford, and R. R. Newton, *Appl. Phys. Lett.* **76**, 1425 (2000).
- ¹⁴A. A. Abdallah, P. C. P. Bouten, J. M. J. den Toonder, and G. de With, *Surf. Coat. Technol.* **205**, 3103–3111 (2011).
- ¹⁵J. Vlassak, Y. Lin, and T. Tsui, *Mater. Sci. Eng. A* **391**, 159 (2005).
- ¹⁶W. P. Vellinga, J. T. M. De Hosson, and P. C. P. Bouten, *J. Appl. Phys.* **110**, 044907 (2011).
- ¹⁷Y. Leterrier, A. Pinyol, L. Rougier, J. H. Waller, and J.-A. E. Månson, *J. Appl. Phys.* **106**, 113508 (2009).
- ¹⁸P. W. France, M. J. Paradine, M. H. Reeve, and G. R. Newns, *J. Mater. Sci.* **15**, 825 (1980).
- ¹⁹M. J. Matthewson, C. R. Kurkjian, and S. T. Gulati, *J. Am. Ceram. Soc.* **69**, 815 (1986).
- ²⁰P. C. P. Bouten, Ph.D. dissertation, Technische Hogeschool, Eindhoven, The Netherlands, 1987.
- ²¹J. de Goede, P. C. P. Bouten, L. Médico, Y. Leterrier, J.-A. E. Månson, and G. Nisato, in *MRS Proceedings*, edited by R. Vinci, R. Schwaiger, A. Karim, and V. Shenoy, 2004, Vol. 854.
- ²²G. de With, "Structure, deformation, and integrity of materials," in *Structure, Deformation, and Integrity of Materials: Plasticity, Visco-Elasticity, and Fracture* (Wiley-VCH Verlag GmbH, 2008), Vol. II, Chap. 24, pp. 757–780.
- ²³J. Hutchinson and Z. Suo, *Adv. Appl. Mech.* **29**, 63 (1991).
- ²⁴K. Jakus, J. E. Ritter, and J. M. Sullivan, *J. Am. Ceram. Soc.* **64**, 372 (1981).
- ²⁵Y. S. Shiue and M. J. Matthewson, *J. Appl. Phys.* **89**, 4787 (2001).
- ²⁶M. A. H. Donners, L. J. M. G. Dortmans, and G. de With, *J. Mater. Res.* **15**, 1377 (2000).
- ²⁷M. A. H. Donners, L. J. M. G. Dortmans, G. de With, and M. J. M. de Graaf, *J. Mater. Res.* **16**, 3575 (2001).
- ²⁸T. A. Michalske, W. L. Smith, and B. C. Bunker, *J. Am. Ceram. Soc.* **74**, 1993 (1991).
- ²⁹L. J. M. G. Dortmans and G. de With, *J. Am. Ceram. Soc.* **74**, 2293 (1991).
- ³⁰M. H. Dielhof, L. J. M. G. Dortmans, and G. de With, *J. Eur. Ceram. Soc.* **12**, 215 (1993).