Residual Stress and Thermal Expansion of Spun-On Polyimide Films

GERHARD ELSNER, *IBM Deutschland GmbH, PR NFS, Department 0837 / 7032-53, P.O. Box 266, D-7032 Sindelfingen, West Germany*

Synopsis

The residual stress in spin coated films and the effect of heat treatments on this stress were determined by the Fizeau interference method. Three types of spin coated polyimide (PI) films have been studied. Two of them were prepared by thermal conversion of their poly(amic acid)s (PAAs) and the third one by solvent evaporation of the soluble preimidized PI. For the imidized PI films the residual stress vs. bake temperature shows an inclined steplike behavior while this function for soluble PI is approximately linear. The room temperature stress relative to silicon substrates in fully baked films (400°C) is between 40 and 70 MPa and nearly independent of the film thickness. From the stress measurement, the thermal expansion coefficient and Young's moduli have also been obtained. The thermal expansion coefficient and the Young's modulus are in the order of 9.0×10^{-6}°C^{-1} and 10 GPa, respectively. These values deviate from those published for bulk material which is explained by the in-plane orientation of the molecular chains in spun-on PI films.

INTRODUCTION

For the electronics industry polymers become more and more important as insulating materials.[1-3] For applications requiring high thermal stability, polyimides (PI) are used having attractive dielectric and thermal properties. PI has a low-frequency dielectric constant of $\simeq 3.5$ at 20°C and is stable at temperatures even above 400°C.[4] The heat resistance of PI results from the cyclic structure, and the low number of mobile groups which are sensitive to heat and oxidation. The glass transition temperature T_g of PI is above 300°C.

A further advantage is the possibility to use the well-known spin-on process for coating.[5-7] Generally, PI is not soluble in most of the common solvents. Therefore, they are usually applied in the form of their precursors, i.e., as poly(amic acid)s (PAA) which are solvent-soluble. The spun-on films are then imidized thermally to obtain the desired properties. In case of soluble PI the spun-on films are thermally exposed to remove the solvent. The thermal treatment results in a loss of film volume and eventually produces intrinsic stresses in the film. However, Goldsmith et al.[8] have shown that the intrinsic stress is zero. They have measured *in situ* the stresses generated in spun-on PI films during the imidization cycle. The stress measurements were made using an X-ray double crystal lattice curvature technique. The PI investigated in that paper was the ester of benzophenonetetracarboxylicdianhydride (BTDA) and 4,4′-methylene dianiline (MDA). The stresses after cooling to room temperature were found to be tensile. The magnitude of these stresses increased linearly with increasing bake temperature up to 70 MPa at 400°C.

Journal of Applied Polymer Science, Vol. 34, 815–828 (1987)
© 1987 John Wiley & Sons, Inc.
CCC 0021-8995/87/020815-14$04.00

Also, the stress was found to be independent of the film thickness for thicknesses of up to 24 μm. However, the influence of different chemical structures on the stresses in the films were not investigated.

The stress in the film arises from a mismatch of the coefficient of thermal expansion between the film and the coated substrate. For PI imidized at 400°C, T_g is approximately 400°C. Since the coefficient of thermal expansion for PI is higher than for Si in this temperature range, one expects that cooling from the T_g will generate tensile stresses in the PI films.

This paper presents results of an investigation on the residual stresses in spin-coated PI films, depending on both the thermal treatments and the chemical structure. The stress in the films coated onto Si wafers were measured by the Fizeau interference method with grazing incidence.[9] The relationship between the residual stress and the thermal treatment as well as the chemical structure is discussed, including the results of thermogravimetric analysis (TGA) measurements. Furthermore, the thermal expansion coefficient and Young's modulus for the spin-coated films are calculated.

EXPERIMENTAL

Film Preparation

Three different types of PI with different chemical structures were investigated. Two of the PI's were used as PAA precursors because they are insoluble in the fully imidized state in most common solvents. The third one is a PI which is soluble in relatively non-polar solvents. Table I summarizes the properties of the PIs, which are the subject of this paper. One of the insoluble PI is the cyclodehydration product of poly[*N'*,*N*-bis (phenoxyphenyl)–pyromellitamic acid] (PAA), made from pyromellitic dianhydride (PMDA) and oxydianiline (ODA). The reaction scheme for the production of this PI from PAA Allotherm 610-16 is shown in Figure 1. The other insoluble PI is a poly(imide-co-siloxane); the chemical structure of PIX-3400 is shown in Figure 2(a).[10]

The soluble PI is the polymerization product of 5(6)-amino-1-(4'-aminophenyl)-1,3,3-trimethylindane (DAPI) with benzophenone tetracarboxylic dianhydride (BTDA).[11] The solubility of the PI is caused by the nonplanar nature of the two amino groups in the (DAPI) diamine. Figure 2(b) shows the chemical structure of Probimide 293.

TABLE I
Properties of the Investigated Polyimides

Trade Name	Allotherm 610-16	PIX-3400	Probimide 293[a]
Supplier	BASF/Beck	Hitachi	Ciba-Geigy
Feature	PAA solution	PAA solution	Imide solution
Solvent	NMP[b]	NMP	GBL[c]
Viscosity	70 P	40 P	75 P

[a] Previously XU 293.
[b] *N*-methyl-2-pyrrolidone.
[c] γ-Butyrolactone.

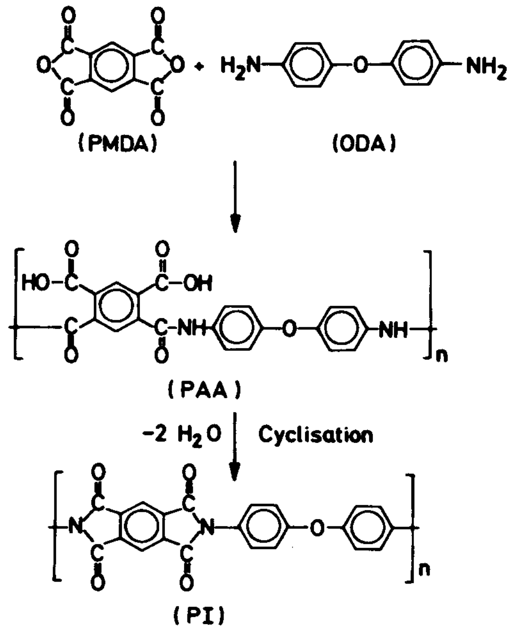


Fig. 1. Polycondensation reaction between pyromellitic dianhydride and oxydianiline.

All solutions were spin-coated onto (100)-oriented 3.25 in. Si wafers and onto 3.25 in. SiO_2 (fused quartz) substrates using a "Convac" photo resist spinner, Model 1001 GS. The spin coating process was performed with a constant acceleration of 5×10^4 rpm/s up to the final spin speed ω, which was applied for 50 s. In order to get proper adhesion between the wafer and PI coating, the following pretreatment was performed: A dilute solution of an organosilane was applied to the wafer and spun to dryness. A typical log–log plot of the PI film thickness d_f after full bake vs. spin speed ω is shown in Figure 3.

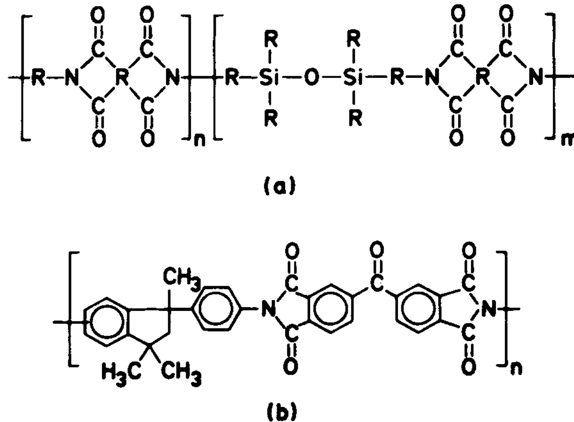


Fig. 2. Chemical structures of (a) Poly(imide-co-siloxane) and (b) BTDA/DAPI polyimide.

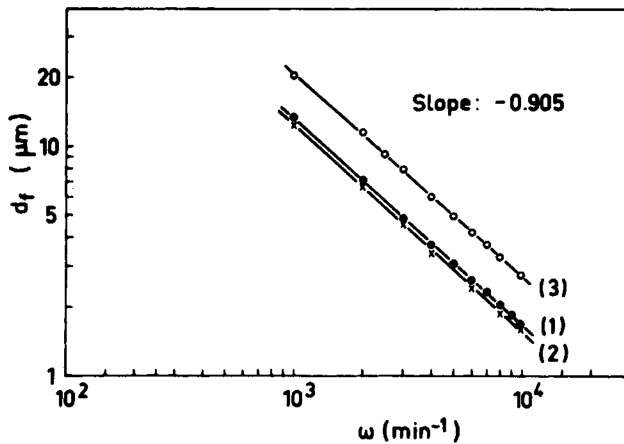


Fig. 3. Film thickness d_f as a function of spin speed ω after full bake (30 min at 60°C + 50 min at 120°C + 50 min at 345°C) for: (1) Allotherm 610-16; (2) PIX-3400; (3) Probimide 293.

All heat treatments were performed in two steps depending on temperature on a hot plate or in an oven under nitrogen or argon atmosphere. All bake temperatures were held constant within ±3°C. The bake procedure was performed by a prebake followed by a final bake at higher temperatures. The prebake time was always 30 min; the final bake time always 50 min. Thermogravimetric analysis (TGA) was carried out on a Perkin-Elmer TGS-2 unit. Samples for the TGA investigation were prepared by casting the PAA and the PI solutions onto glass plates and by adjusting the coating thickness to 100 µm by doctor blading. Thereafter, these samples were prebaked at 60°C for 30 min followed by an additional bake at 120°C for 50 min.

Stress Measurements

The stress in the PI was determined by measuring the radius of curvature of the substrates without and with the PI coating using the Fizeau interference method with grazing incidence.[9] In this experimental setup, an optical flat was used as reference surface instead of the reflection grating used in Ref. 9. The measurements of the radius of curvature were performed at room temperature. In order to guarantee that the measured stress produced by the thermal treatment is only caused by the PI films and not influenced by instabilities of the substrates, some uncoated substrates were baked and measured for comparison. The fringe pattern analysis showed no such instabilities.

The relation between the residual film stress σ_f and the radius of curvature in the case of $d_f \ll d_s$ is[12]

$$\sigma_f = \frac{1}{6} \cdot \frac{E_s}{(1 - \nu_s)} \cdot \frac{d_s^2}{d_f}\left(\frac{1}{R_f} - \frac{1}{R_0}\right) \tag{1}$$

where R_0 and R_f are the radii of curvature for the Si substrate before and after the film deposition, E_s and ν_s are Young's modulus and Poisson's ratio

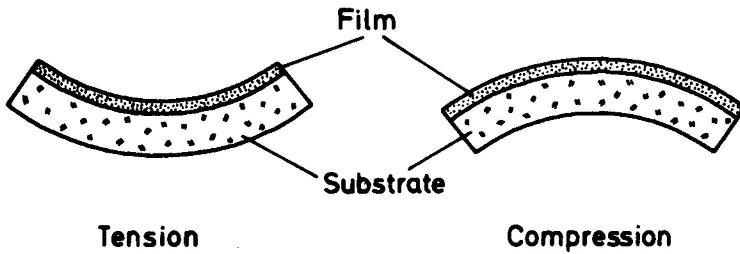


Fig. 4. Convention showing tensile and compressive stresses.

for the substrate, and d_s and d_f are the substrate and film thickness, respectively. The film stress in the plane of the substrate is assumed to be isotropic. The substrates used were (100)-oriented Si wafers and SiO_2 wafers (fused quartz, Synsil from Westdeutsche Quarzschmelze GmbH), both 3.25 in. in diameter and 400 μm in thickness. For (100)-oriented Si substrates $E_s/(1 - \nu_s)$ is 1.81×10^5 MPa(13) and for SiO_2 substrates $E_s/(1 - \nu_s)$ is 8.68×10^4 MPa.[14] Figure 4 shows the convention used for tensile and compressive stresses.

Thermal stress σ_t, caused by the difference between the thermal expansion coefficients for the film and the substrate, was calculated from[15]

$$\sigma_t = \frac{E_f}{(1 - \nu_f)}(\alpha_f - \alpha_s)(T_b - T) \tag{2}$$

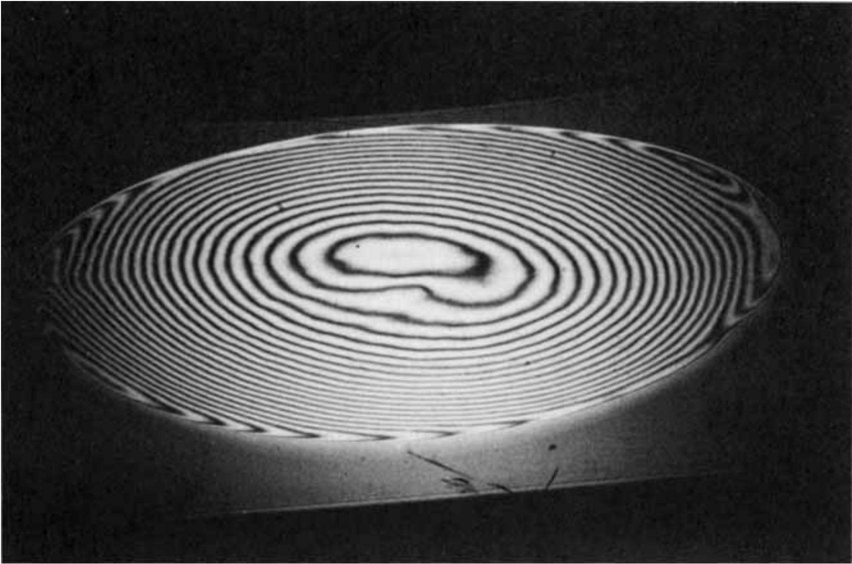
where α_f and α_s are the thermal expansion coefficients for film and substrate and E_f and ν_f are Young's modulus and Poisson's ratio for the film, respectively. T_b is the film bake temperature and T is the temperature at which the stress is measured.

PI film thickness for bake temperatures above 175°C were measured by an Alpha-Step (Tencor Corp.) profilometer. Below approximately 175°C the measurement is inaccurate due to the sink-in of the stylus into the partly baked PI film. Therefore, an IBM Film Thickness Probe (FTP) was used.[16] The FTP uses a white light interferometry and need the refractive index to calculate film thickness. The refractive index of the baked PI was assumed to be $n = 1.72$.[17] In the case of bake temperatures below 100°C, the refractive index decreases to 1.62, the value of PAA.[18] However, this variation can be neglected because of its small influence on the stress results.

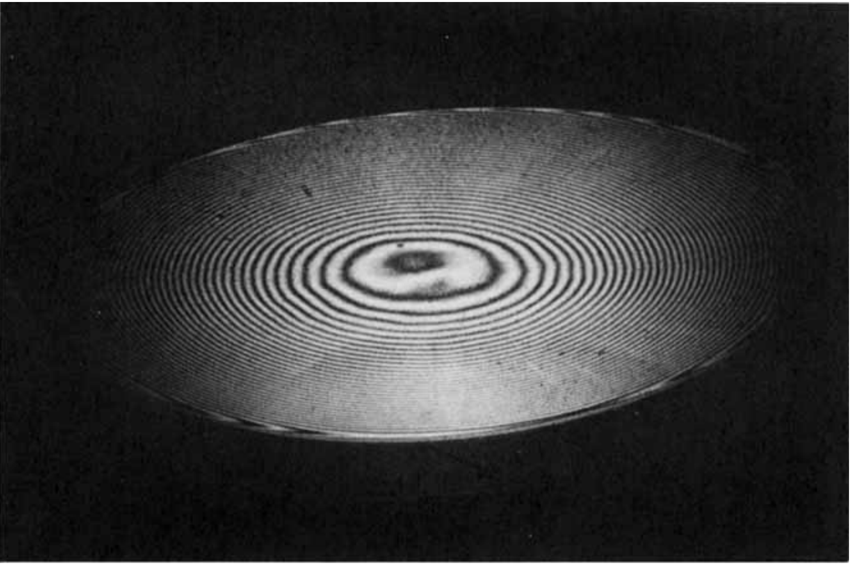
RESULTS

Residual Stress in Films

Photographs of Fizeau interference fringe patterns of uniformly curved Si substrates are shown in Figure 5: a wafer without PI film in Figure 5(a), and a wafer with PI film baked at 345°C in Figure 5(b). Due to the inclination of the beam to the substrate, the fringe pattern is elliptical. It is evident that the number of fringes is increased and the radius of curvature decreased when the solution of PAA and PI is deposited on the substrate and baked. Figure 6 shows the residual stress σ_f for fully imidized Allotherm 610-16 PI films as a



(a)



(b)

Fig. 5. Interference fringe pattern of uniformly curved Si substrate. The beam is inclined 20° to the substrate normal (fringe spacing corresponds to 0.9 μm): (a) without PI film; (b) with PI film baked at 345°C.

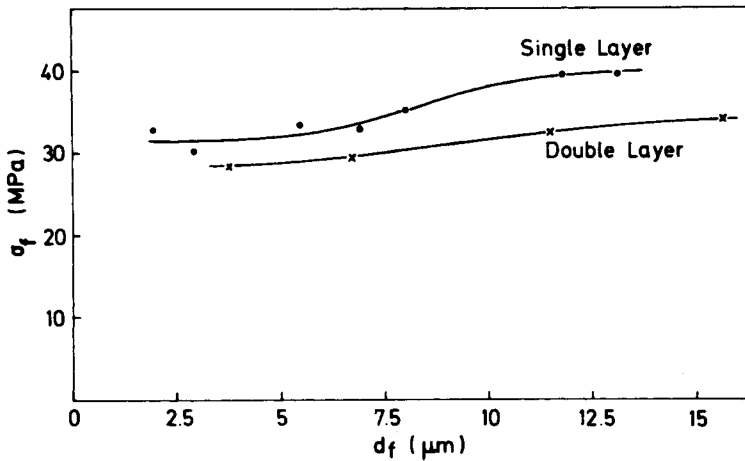


Fig. 6. Residual stress σ_f for fully imidized Allotherm 610-16 PI film as a function of film thickness d_f for single and double layer.

function of the film thickness d_f. The stresses were measured at room temperature. In order to analyze the influence of multiple coatings on the film stress, the substrates were coated with single layers as well as with double layers. In case of double layers the first layer was prebaked at 85°C for 30 min before applying the second one. Then, the final bake at 345°C for 50 min for full imidization was performed. The results in Figure 6 show for Allotherm 610-16 that the effect of the film thickness on the residual stress is small for both cases. However, a double coating process reduces the film stress slightly. Similar behavior is observed with the PIX-3400 PI films as well as the Probimide 293 PI films.

Figure 7 shows the residual stress σ_f for Allotherm 610-16 PI films measured at room temperature as a function of the final bake temperature T_b for

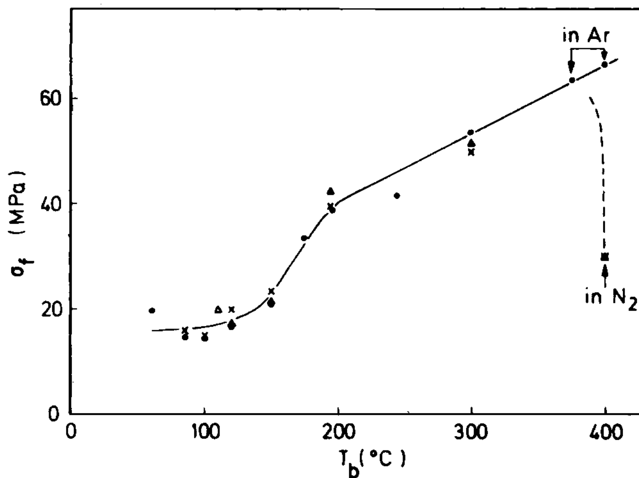


Fig. 7. Residual stress σ_f for Allotherm 610-16 PI film as a function of final bake temperature T_b for different prebake temperatures (°C) (prebake time: 30 min): (●) 61; (×) 85; (△) 100.

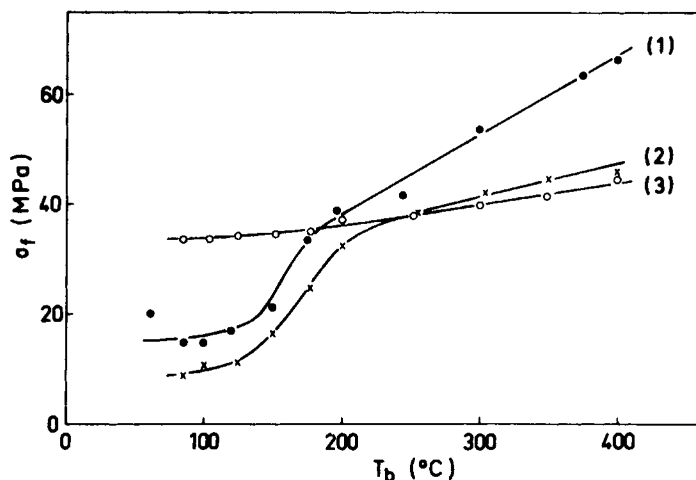


Fig. 8. Residual stress σ_f as a function of final bake temperature T_b for : (1) Allotherm 610-16; (2) PIX-3400; (3) Probimide 293. Prebake temperature 85°C for 30 min.

different prebake temperatures. All films exhibited tensile stresses in the temperature range studied and the samples become concave as viewed from the film side (see convention in Fig. 4). Further on, the film stress increases in a steplike shape with increasing final bake temperature. Above 200°C the stress increases linearly. However, it is independent of the prebake temperature. This independence on the prebake temperature was observed also with the PIX-3400 PI films and Probimide 293 PI films. In order to investigate the influence of film decomposition on film stress, a final bake at 400°C was performed under nitrogen instead of argon atmosphere. A small amount of oxygen was added to the nitrogen in order to induce film decomposition. As a result, the stress decreases to approximately 40% of its original value.

In Figure 8 the residual stress σ_f as a function of the final bake temperature T_b measured at room temperature for all PI films (Allotherm 610-16, PIX-3400, Probimide 293) is shown. The prebake temperature was 85°C. As already explained for Allotherm 610-16 PI films in Figure 7, the PIX-3400 PI films also show the same inclined steplike stress shape. However, the stress curve is shifted to somewhat lower values. For Probimide 293 the stress shows a nearly linear increase in the entire temperature range. Below 150°C the stress σ_f is approximately two times higher compared to the other two materials.

During bake a loss of film volume takes place and TGA indicates a corresponding weight loss. Because the PI films adhere to the substrates during bake the loss of volume is essentially in one dimension, i.e., in the film thickness. This is shown in Figure 9 where the film thickness d_f of Allotherm 610-16 is plotted vs. the final bake temperature T_b for different prebake temperatures. It is evident from this figure that the value of d_f decreases with the final bake temperature T_b up to 200°C and is constant for $T_b > 200$°C. Moreover, the film thickness d_f is independent of the prebake temperature as was the residual stress. The same is valid for PIX-3400 films and Probimide 293 films.

Figure 10 shows the not-normalized film thicknesses d_f as a function of the final bake temperature T_b for all PI films (Allotherm 610-16, PIX-3400,

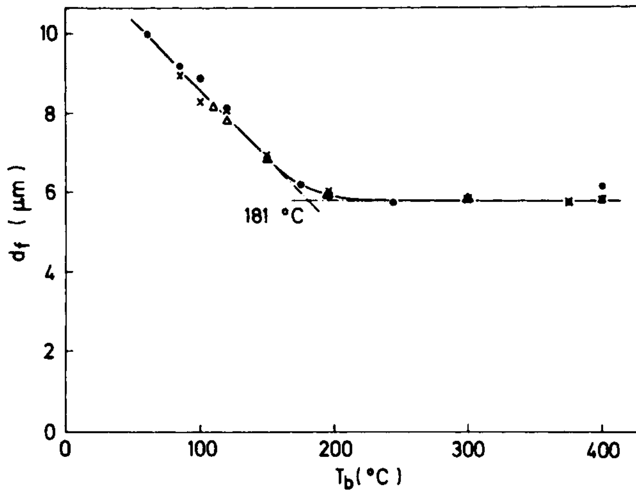


Fig. 9. Film thickness d_f for Allotherm 610-16 PI film as a function of final bake temperature T_b for different prebake temperatures (°C) (prebake time: 30 min): (●) 61; (×) 85; (△) 110.

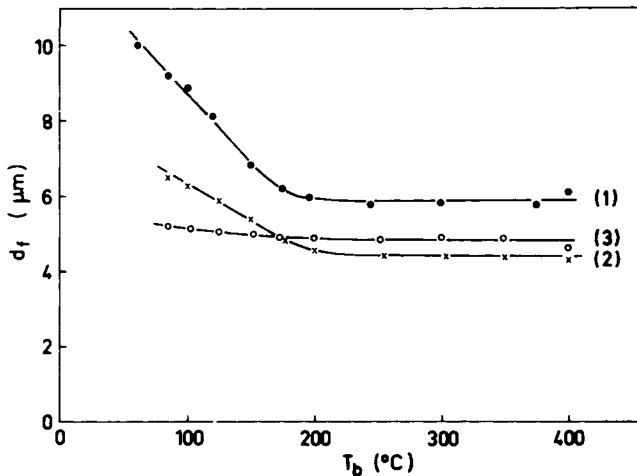


Fig. 10. Film thickness d_f (not normalized) as a function of final bake temperature T_b for: (1) Allotherm 610-16: (2) PIX-3400; (3) Probimide 293. Prebake temperature 85°C for 30 min.

Probimide 293) for a prebake temperature at 85°C. The thickness of the PIX-3400 PI films shows nearly the same behavior as that of Allotherm 610-16 PI. However, the thickness of the Probimide 293 PI film shows a slight decrease up to 200°C and is then constant.

Thermogravimetric Analysis

A thermogravimetric analysis (TGA) of all PI (Allotherm 610-16, PIX-3400, Probimide 293) prebaked in nitrogen twice, for 30 min at 60°C and 50 min at 120°C is shown in Figure 11 (full line). Three major regions of weight loss are observed for Allotherm 610-16 PI and for PIX-3400 PI films [Figs. 11(a) and 11(b)]: Approximately 20–30% is lost between 100 and 230°C, another 5–8% is lost between 230 and 360°C at a comparatively slow rate, and beyond 500°C

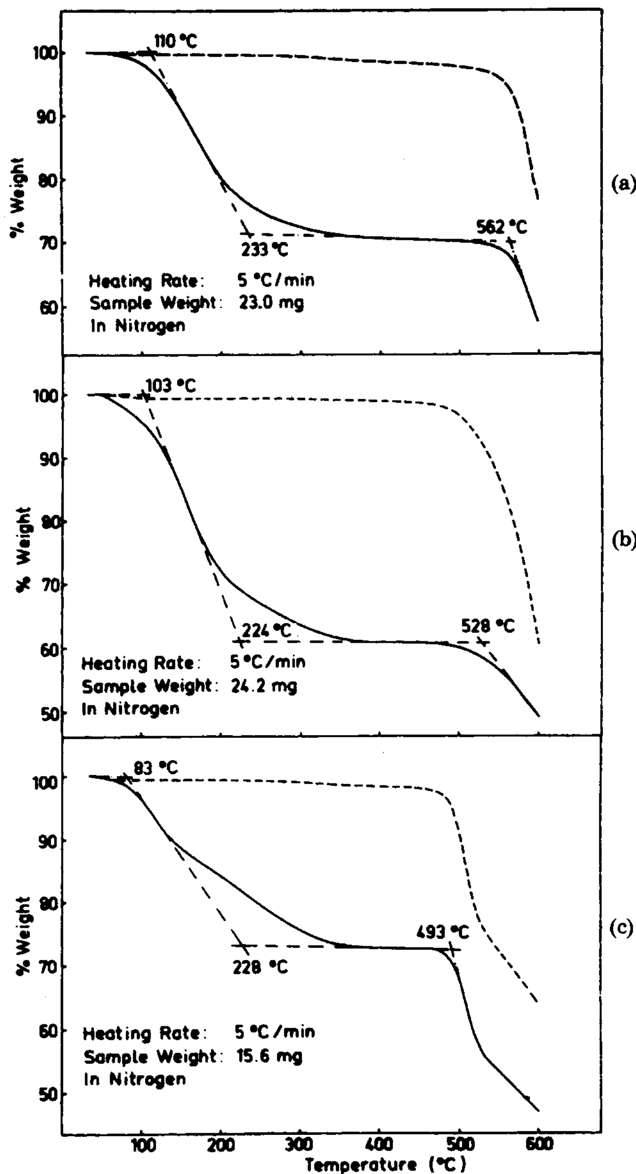


Fig. 11. Thermal gravimetric analysis of (a) Allotherm 610-16, (b) PIX-3400, and (c) Probimide 293: (—) baked at 60°C (30 min) + 120°C (50 min); (---) fully baked at 60°C (30 min) + 120°C (50 min) + 345°C (50 min).

both PI films decompose. In the region between 360 and 500°C the films are stable and lose no weight. A slightly different behavior shows the Probimide 293 PI film [Fig. 11(c)]. Only 15–18% weight is lost between 80 and 230°C. However, in contrast to the foregoing PI films, a more pronounced 12–14% loss is between 230 and 360°C at a relatively high rate. Above 500°C the PI film decomposes. For comparison a fully baked sample is also shown in Figure 11 (broken line).

Thermal Expansion Coefficient and Young's Modulus

Figure 12 shows the temperature dependence of the curvature $1/R_f$ for all PI films (Allotherm 610-16, PIX-3400, Probimide 293) on Si and SiO_2 substrates measured in a temperature range from 55°C to 215°C. All PI films were exposed to a prebake at 85°C for 30 min and a final bake at 345°C for 50 min. As discussed by Goldsmith et al.,[8] once the films were fully baked, the stresses change reversibly with temperature, and all stresses are caused by

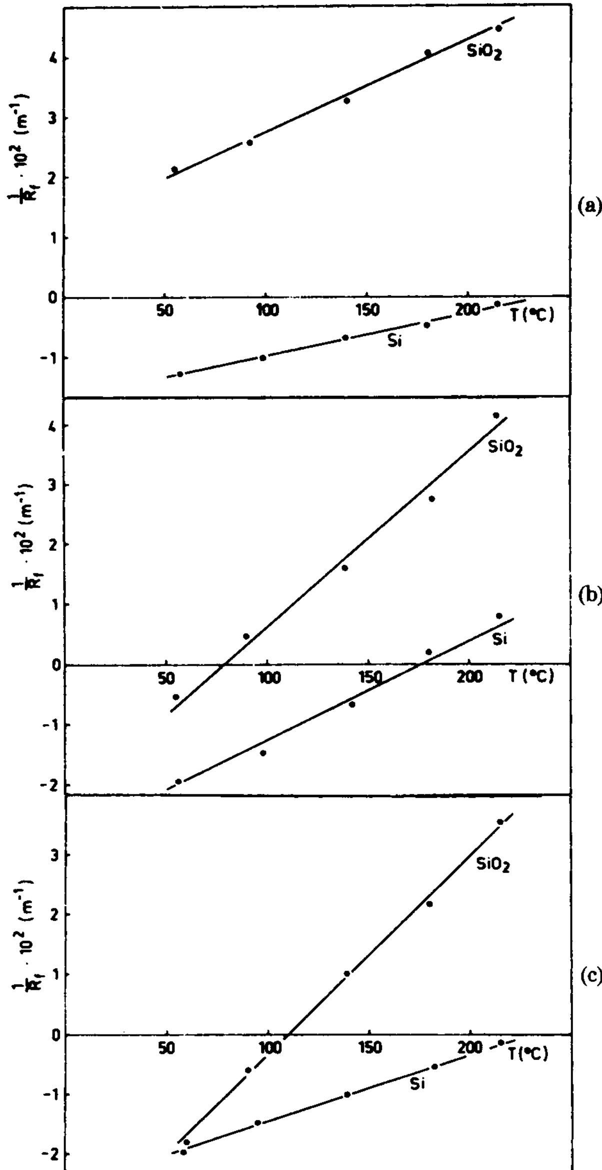


Fig. 12. Temperature dependence of curvature $1/R_f$ for Si and SiO_2 substrates for fully baked (a) Allotherm 610-16, (b) PIX-3400, and (c) Probimide 293. Bake conditions: 60°C (30 min) + 120°C (50 min) + 345°C (50 min).

thermal expansion mismatch. Therefore, we get, by combining eqs. (1) and (2),

$$\left(\frac{1}{R_f} - \frac{1}{R_0}\right) = 6 \cdot \frac{E_f}{E_s} \cdot \frac{d_f}{d_s^2} \cdot \frac{(1 - \nu_s)}{(1 - \nu_f)}(\alpha_f - \alpha_s)(T_b - T)$$

As shown in Figure 12, $1/R_f$ increases linearly with increasing T. This means that α_f, ν_f, E_f, α_s, ν_s, and E_s are independent of the temperature in the temperature range from 55 to 215°C. Accordingly, from the difference in the slopes for the two curves in Figure 12(a), α_f and E_f for the Allotherm 610-16 PI film are estimated to be 9.0×10^{-6}°C^{-1} and 7.5 GPa, respectively. In the same way as the Allotherm 610-16 PI film, the values of α_f and E_f for the PIX-3400 PI film in Figure 12(b) are estimated to be 11.0×10^{-6}°C^{-1} and 9.8 GPa. Finally, the values of α_f and E_f for the Probimide 293 PI film in Figure 12(c) are estimated to be 8.2×10^{-6}°C^{-1} and 13.4 GPa, respectively.

DISCUSSION

Figure 6 shows that the residual stress σ_f in fully baked PI films is nearly independent from the film thickness d_f for a single coating as well as for double coating. The lower stress for double layers is believed to be caused by a molecular chain relaxation process originating from the partial dissolution of the underlying PI layer during application of the second layer.

One of the most important variables in the processing of PI coatings is the bake cycle. During heating, absorbed water and solvent (NMP, GBL) evaporate simultaneously. In case of Allotherm 610-16 PI and PIX-3400 PI additional water evaporates created by the imidization reaction. The residual stress σ_f and the film thickness d_f are independent of the prebake temperature as shown in Figures 7 and 9. This behavior indicates that only solvent evaporation takes place during prebake in the 60–110°C temperature range. Structural changes influencing the residual stresses occur only above 110°C where the imidization process starts.[19] Despite the independence of both the film thickness d_f and the residual film stress σ_f on the prebake temperature a prebake of spun-on films is necessary to avoid the formation of brittle films.

As shown in Figure 10, the film thickness for Allotherm 610-16 and PIX-3400 decreases with increasing final bake temperature up to 200°C and is constant above. This decrease below 200°C is caused by evaporation of the water which is formed during the imidization process and also by evaporation of residual solvents (bp NMP: 202°C). This loss was also measured by TGA in the temperature range of 100–230°C [Figs. 11(a) and 11(b)]. The imidization process is almost complete at 200°C.[19] Above 200°C, there is only little change in weight, and the film thickness d_f remains constant.

A different behavior is observed with the Probimide 293 film. An insignificant decrease of the thickness up to 200°C is observed. Because the soluble Probimide 293 does not imidize, only residual GBL evaporation (bp 204°C) takes place as measured by TGA in the temperature range of 80–230°C [Figure 11(c)]. Above 200°C the film thickness remains constant.

In Figure 8 the residual film stress σ_f is shown as a function of the final bake temperature T_b. Only a little stress is seen in the 60–100°C temperature range for both the Allotherm 610-16 and PIX-3400, because the NMP is almost completely evaporated during the previous prebake step and the

imidization has not fully started yet. With beginning imidization above 110°C, the stress increases rapidly until 200°C is attained. In this temperature range the PI films releases water due to the cyclization process. In addition, the remaining solvents (NMP, GBL) evaporate. The rapid increase in stress can be explained by increasing rigidity of the polymer chains as a result of the imide formation. Beyond 200°C the imidization is complete, and the stress increases linearly with the temperature up to 400°C. In contrast the Probimide 293 is from the beginning a completely developed PI which is soluble. Therefore, no fundamental change in polymer structure takes place. As a result the stress increases nearly linear in the entire temperature range up to 400°C.

Goldsmith et al.[8] have investigated the *in situ* stresses generated during bake. They have shown once the PI was fully baked (above 200°C), the stress changes reversibly and linearly during cooling from final bake temperature to room temperature and vice versa. At final bake temperature the stress value is always approximately zero. This behavior indicates that no intrinsic stress is generated during baking and that the residual stress is only due to the difference in the thermal expansion coefficients between the PI film and the substrate. This justifies our assumption that the residual film stress is equal to the thermal stress, i.e., $\sigma_f = \sigma_t$. Even if the intrinsic stress is not zero, the values for the thermal expansion coefficient and the Young's modulus will not be influenced because an intrinsic stress only shifts the $1/R_f$ curve.

Based on this result and the measurements presented in this paper we can extract the thermal expansion coefficients α_f and the Young's moduli E_f in the temperature range from 55 to 215°C. These values are estimated from the slopes in Figure 12. The results of $\alpha_f \simeq 9.0 \times 10^{-6}{}^\circ C^{-1}$ is by a factor of 4 smaller, and the result for $E_f \simeq 10$ GPa is by a factor of 4 higher than the corresponding values for commercially available PI Kapton (supplier DuPont). The values are $31.0 \times 10^{-6}{}^\circ C^{-1}$ and 2.5 GPa for the thermal expansion coefficient and Young's modulus, respectively.[20]

Numata et al.[21] have investigated two categories of PIs which are different in their main chain structure. The first has directly bonded para-benzene rings and the other one more flexible linkages between the benzene rings. The lower thermal expansion coefficient of the first category has been assumed by these authors to depend on linear conformational structure of para-benzene units. In contrast, our material shows a lower thermal expansion coefficient in spite of flexible linkages so that we cannot explain the effect of lower thermal expansion coefficient according to the above-mentioned authors. We suppose the lower thermal expansion coefficient in our case to be due to in-plane orientational effect.

Spun-on PI films have an in-plane orientation of the molecular chains.[18] Because the thermal expansion coefficient in the molecular chain direction is lower than that in perpendicular direction, the obtained value deviates from that for isotropic oriented chains. Both the thermal expansion and the elasticity depend on the degree of molecular chain orientation.

The Allotherm 610-16 PI used in the experiments described was provided by S. Pohl, Manufacturing IBM Boeblingen. F. Druschke, H. Hinkel, and J. W. Bartha contributed to this paper by helpful discussions and W. Jaerisch by Fizeau interference measurements. This assistance is hereby gratefully acknowledged.

References

1. E. D. Feit and C. W. Wilkins, Jr., Eds., *Polymer Material for Electronic Applications*, ACS Symposium Series 184, Am. Chem. Soc., Washington, DC, 1982.

2. K. Mukai, A. Saiki, K. Yamanaka, S. Harada, and S. Shoji, *IEEE J. Solid-State Circ.*, **SC-13**, 462 (1978).

3. M. A. Zuegel, in *Methods and Materials in Microelectronic Technology*, Joachim Bargon, Ed., Plenum, New York, 1984, p. 269.

4. Yu. N. Sazanov, *J. Thermal Anal.*, **23**, 65 (1982).

5. B. D. Washo, *IBM J. Res. Dev.*, **21**, 190 (1977).

6. F. L. Givens and W. J. Daughton, *J. Electrochem. Soc.*, **126**, 269 (1979).

7. W. J. Daughton and F. L. Givens, *J. Electrochem. Soc.*, **129**, 173 (1982).

8. C. Goldsmith, P. Geldermans, F. Bedetti, and G. A. Walker, *J. Vac. Sci. Technol.*, **A1**, 407 (1983).

9. W. Jaerisch and G. Makosch, *Appl. Opt.*, **17**, 740 (1978).

10. Hitachi Chemical Data Sheet, Sep. 30, 1984

11. J. H. Bateman, W. Geresy, Jr., and D. S. Neiditch, *Coatings Plast. Prepr.*, **35**(2), 77 (1975).

12. R. J. Jaccodine and W. A. Schlegel, *J. Appl. Phys.*, **37**, 2429 (1966).

13. J. J. Wortman and R. A. Evans, *J. Appl. Phys.*, **36**, 153 (1965).

14. Data Sheets, Schl. 6.79/5, Westdeutsche Quarzschmelze GmbH, D-2054 Geesthacht, West Germany.

15. W. R. Hoffman, in *Physics of Thin Films*, G. Haas and R. E. Thun, Eds., Academic, New York, 1966, Vol. 3, p. 211.

16. H. E. Korth, *J. Phys. (Paris)*, **44**, C10–101 (1983).

17. W. A. Pliskin, private communication, unpublished data, 1980.

18. T. P. Russel, H. Gugger, and J. D. Swalen, *J. Polym. Sci., Polym. Phys. Ed.*, **21**, 1745 (1983).

19. Y. K. Lee and J. D. Craig, in Ref. 1.

20. Data Sheets, E-42727 6/81, DuPont Co., Wilmington, DE 19898.

21. S. Numata, S. Oohara, K. Fujisaki, J. Imaizumi, and N. Kinjo, *J. Appl. Polym. Sci.*, **31**, 101 (1986).

Received October 23, 1986
Accepted December 2, 1986