Tensile Properties and Viscoelastic Model of a Polyimide Film*

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Abstract

This paper presents tensile properties of a polyimide thin film used in electronic devices. Tensile tests were performed to determine Young's modulus, proportional limit, yield stress, ultimate tensile strength and elongation of the polyimide film. Effects of strain rate and temperature on the tensile properties were discussed. There was a little effect of strain rate on Young's modulus but proportional limit, yield stress and ultimate tensile strength increased with increasing strain rate. Only elongation decreased with strain rate. Young's modulus, proportional limit, yield stress and ultimate tensile strength decreased with increasing temperature, but elongation increased. Applicability of a viscoelastic model for describing the stress-strain curves of the polyimide film was discussed.

Key words: Polyimide, Thin Film, Tensile Properties, Viscoelastic Model

1. Introduction

Build-up circuit board, consisting of laminated resin and copper thin films, has been widely used in various electronic devices to achieve high density mounting of electronic parts. Build-up circuit board undergoes cyclic temperature variation and the mismatch of the coefficient of thermal expansion between the different films causes cyclic thermal stress. In some cases, failure in the laminated circuit board was reported by the thermal crack initiation and propagation in the films ^{(1), (2)}. Numerical analysis, like finite element analysis, has been widely used to estimate the thermal stress and strain ranges in electronic devices and the ranges have been used to predict the thermal fatigue life. In this procedure, accurate tensile properties of resin films are essential for accuracy of the analysis resulting in the quality assurance of laminated circuit board. Since circuit boards in most practical electronic devices undergo temperature variation, tensile properties of resin films at actual temperature ranges are needed.

Tensile properties of conventional bulk materials are well known but they are not necessarily same as those of thin films, because the properties of thin films may vary with fabrication process, texture and thickness ⁽³⁾⁻⁽⁷⁾. Therefore, tensile properties as used in practice should be measured and used for a reliable estimate of cyclic thermal stresses in electronic devices ⁽⁸⁾. Polyimide films have been attracted attention recently in electronic industries to use as a material for resistant layers in build-up circuit board because of their excellent heat resistance and low coefficient of thermal expansion ⁽⁹⁾⁻⁽¹¹⁾. In addition, tensile

*Received 28 Nov., 2011 (No. 11-0723)
[DOI: 10.1299/jmmp.6.668]

properties of polyimide films have been extensively studied ⁽¹²⁾⁻⁽¹⁶⁾. For example, Saeed et al. ⁽¹²⁾ investigated effect of monomer structure on mechanical properties for 4 kinds of polyimide films. In other works, Deng et al. ⁽¹³⁾ and Musto et al. ⁽¹⁴⁾ studied the mechanical properties of polyimide-silica hybrid films. However, there are little researches studying the effect of temperature on tensile properties of polyimide films.

The objective of this paper is to study tensile properties of the polyimide film used in laminated circuit board. Tensile tests were conducted to obtain Young's modulus, proportional limit, yield stress, ultimate tensile strength and elongation at three strain rates at three temperatures. Effects of strain rate and temperature on the tensile properties were discussed. Applicability of a viscoelastic model for describing the stress-strain curves of the polyimide film is discussed.

2. Experimental Procedure

The material tested was a polyimide film called Kapton of which the molecular structural is shown in Fig.1 and was used as received from Du Pont-TORAY Co., Ltd. The polyimide film was made by injection process, leading to orientation of polymer chains in the film. The amount of polymers oriented in the injection direction is slightly high. For that reason, specimens cut in the injection direction (ID) and in its transverse direction (TD) were tested to examine the anisotropy of the polyimide film as shown in Fig.2. In addition, removing internal stress treatment was conducted for the polyimide film after injection process. Other detailed information about the fabrication process was not shown in this paper because of commercial secret of the company.

Figure 3 shows the shape and dimensions of tensile specimen with 70mm parallel length, 8mm width and 25 μ m thickness. Two small white dots with 1mm in diameter with 70mm span were marked on the parallel part of the specimen to measure elongation during tensile test. Aluminum plates with 200 μ m thickness were glued on the both sides of the grip part of specimen to avoid breaking of the film from the grip part. Since specimen sometimes broke in loading stage of gripping, the shadow area shown in the figure was strengthened with copper sheet to protect from breaking and the protected part was cut just before starting tests.

Fig.1 Molecular structural of polyimide used

Fig.2 Cutting directions of specimen

Fig.3 Shape and dimensions of specimen tested (mm)

Fig.4 Schematic of tensile apparatus

Fig.5 Stress-strain curves of polyamide films

Figure 4 shows a schematic of a tensile tester (SHIMADZU/EZ Test) used in this study⁽⁸⁾. The resolution and the maximum capacity of load cell of the apparatus were 0.025N and 500N, respectively. The displacement of the two dots was measured by an image sensor with two CCD cameras with 1.5 μ m displacement resolution. Temperature of the specimen was raised by an electric resistance furnace with 900W. A small fan was installed in upper part of the furnace to improve the temperature distribution of the specimen and the temperature variation in the gage part was within 1.7K during test. Tensile tests were performed at strain rates of 0.01%/s, 0.1%/s and 1%/s at 298K, 353K and 398K. Five specimens were tested under each testing condition.

3. Results and Discussion

3.1 Tensile Properties

Figure 5 depicts stress-strain curves of the polyimide film (a) in two cutting directions, (b) at three strain rates and (c) at three testing temperatures. All the polyimide films showed clear strain hardening and ruptured at the maximum stress. The stress-strain curve in the ID is nearly the same as that in the TD, Fig.5 (a). The stress increased with increasing the strain rate but decreased with increasing the temperature as shown in Fig.5 (b) and Fig.5 (c).

Table 1 summarizes tensile test results obtained in this study. The data in parenthesis show the standard deviation of the mean for each value. Figure 6 presents the variation of

Young's modulus (E) with strain rate at 298K in the ID and the TD. There was a little difference in Young's modulus between the ID and the TD so that slight anisotropy of Young's modulus exists in the polyimide film. Young's moduli at 0.1%/s were the largest and Young's moduli at 1%/s were slightly larger than those at 0.01%/s. Since Young's modulus of polymer is mainly depends on the molecular structure, the strain rate has little effect on Young's modulus of the polyimide film.

Comparing Young's modulus of the polyimide film to that of bulk material with 3.1mm thickness measured by Notomi et al. ⁽¹⁷⁾, Young's modulus of bulk polyimide was 3.18GPa at 298K and was similar to those of the film. This result means that the thickness of specimen has little effect on Young's modulus of the polyimide film, which is consistent with the reference data from company ⁽¹⁸⁾. That is, Young's moduli of the polyimide films with thickness ranged from 12.5 μ m to 125 μ m were almost same.

Table 1 Summary of tensile test results

T , K	Cutting direction	Strain rate $d\varepsilon/dt$, %/s	Young's modulus E , GPa	Proportional limit σ_p , MPa	Yield stress $\sigma_{0.2}$, MPa	Ultimate tensile strength σ_B , MPa	Elongation δ , %
298	ID	0.01	3.19 (0.23)	33.9 (2.6)	46.6 (3.0)	264 (16.4)	67.1 (3.7)
		0.1	3.73 (0.12)	33.1 (0.9)	50.9 (0.7)	300 (6.7)	64.3 (3.5)
		1.0	3.32 (0.20)	43.3 (2.5)	61.3 (2.1)	309 (8.0)	59.2 (1.6)
	TD	0.01	3.21 (0.10)	31.8 (1.3)	44.0 (1.7)	260 (12.7)	64.5 (2.0)
		0.1	3.56 (0.14)	34.4 (1.6)	50.8 (1.4)	279 (10.8)	60.9 (4.6)
		1.0	3.19 (0.16)	47.5 (2.9)	61.5 (1.4)	294 (9.4)	60.2 (4.1)
353	ID	0.1	2.87 (0.09)	32.6 (1.4)	41.8 (2.0)	263 (17.7)	67.2 (8.6)
	TD	0.1	3.07 (0.11)	32.1 (0.6)	42.1 (1.0)	253 (16.4)	67.7 (9.2)
398	ID	0.1	2.59 (0.18)	30.1 (0.7)	38.8 (1.1)	235 (19.8)	70.4 (11.0)
	TD	0.1	2.42 (0.09)	28.3 (0.6)	39.6 (1.6)	228 (11.6)	73.1 (8.7)

Fig.6 Variation of E with strain rate

Fig.7 Variation of E with temperature

Fig.8 Variations of σ_p , $\sigma_{0.2}$ and σ_B with (a) strain rate and (b) temperature

Fig.9 Variation of elongation with (a) strain rate and (b) temperature

Figure 7 plots Young's modulus against testing temperature at 0.1%/s in the ID and the TD. Young's modulus decreased monotonically with the testing temperature in both the ID and the TD. This results from that the thermal activation process of molecular increases with increasing of temperature.

Figure 8 shows variations of proportional limit (σ_p), yield stress ($\sigma_{0.2}$) and ultimate tensile strength (σ_B) in the ID and the TD with (a) strain rate at 298K and (b) testing temperature at 0.1%/s. Proportional limits and yield stresses in the ID were comparable to those in the TD but ultimate tensile strengths in the ID were somewhat larger than those in the TD, because the amount of polymers oriented in the ID is slightly higher than that in the TD. Since the ultimate tensile strength of bulk polymer was about 88MPa⁽¹⁷⁾, ultimate tensile strengths of the polyimide film were larger than that of the bulk polyimide. Ultimate tensile strength of polyimide film decreases with increasing the thickness of the film, which is consistent with the result of other report⁽¹⁸⁾.

The proportional limit, yield stress and ultimate tensile strength increased with the strain rate and decreased with the testing temperature. Increasing strain rate has the same effect on the proportional limit, yield stress and ultimate tensile strength as decreasing temperature, which can be easy explained by the thermal activation process of molecular.

In order to discuss the effects of strain rate and testing temperature on elongation, the elongations in the ID and the TD are plotted against the strain rate at 298K and the testing temperature at 0.1% in Figs.9 (a) and (b), respectively. The elongations in the ID were mostly the same as those in the TD at all the tests, indicating no anisotropy of the polyimide film in elongation. The elongation of bulk polyimide was more than 100%⁽¹⁷⁾ so the elongations of all the polyimide films were smaller than that of the bulk polyimide. In addition, the elongations slightly decreased with the strain rate and significantly increased with the temperature.

3.2 Viscoelastic model

A viscoelastic model ⁽¹⁹⁾ was used to describe the stress-strain curves of the polyimide film obtained in this study. This paper employed 3-element Maxwell model consisting of two spring elements and a dashpot element as shown in Fig.10 from its simplicity and easiness to apply in numerical analysis, like finite element analysis. Stress response for the 3-element Maxwell model at a constant strain rate (C) is given by the following equation.

$$\sigma = C\eta(1 - e^{-\frac{k_2 \varepsilon}{\eta C}}) + k_1 \varepsilon, \quad C = \frac{d\varepsilon}{dt} \quad (1)$$

k_1 and k_2 are the spring constants of the two spring elements, η the viscosity constant of the dashpot element and t the time.

Figure 11 presents the method of determining the constants in the 3-element Maxwell model. Eq. (1) is reduced to simple equation below at high strain regions in the stress-strain curve.

$$\sigma = C\eta + k_1 \varepsilon, \quad C = \frac{d\varepsilon}{dt} \quad (2)$$

Therefore, k_1 and η is easily calculated by the following equation.

$$k_1 = \tan \theta, \quad \eta = \frac{D}{C} \quad (3)$$

k_2 is determined by the regression analysis to have the best fitting of the data.

Table 2 lists the values of k_1 , k_2 and η in the 3-element Maxwell model under each testing condition performed here. Note that only k_2 is influenced by the testing temperature but is not by the strain rate. The cutting direction has also no influence on k_2 . Figures 12 (a) and (b) show the variations of k_1 with strain rate and temperature, respectively. k_1 linearly increased with strain rate and decreased with increasing testing temperature. The linear relationship is expressed by the following equation.

$$k_1 = (C_1 \frac{d\varepsilon}{dt} + C_2) \exp(\frac{\Delta Q_1}{RT}) \quad (4)$$

Fig.10 3-element Maxwell model

Fig.11 Method for determining the constants in the model

Figure 13 plots k_2 against temperature at 0.1%/s in the ID. k_2 decreased with testing temperature and the relationship is equated as follow,

$$k_2 = C_3 \exp\left(\frac{\Delta Q_2}{RT}\right) \quad (5)$$

The variation of η with strain rate and temperature are presented in Figs.14 (a) and (b), respectively. η is almost inversely related with strain rate and is decreased with testing temperature. η is therefore equated below,

$$\eta = C_4 \left(\frac{d\varepsilon}{dt}\right)^{-0.98} \exp\left(\frac{\Delta Q_3}{RT}\right) \quad (6)$$

Table 2 Values of k_1 , k_2 and η in 3-element Maxwell model

T , K	Cutting direction	Strain rate $d\varepsilon/dt$, %/s	Spring constant k_1 , MPa	Spring constant k_2 , MPa	Viscosity constant η , GPa·s
298	ID	0.01	231	2860	1280
		0.1	241	2860	140
		1.0	286	2860	13.9
	TD	0.01	210	2860	1270
		0.1	227	2860	141
		1.0	249	2860	14.4
353	ID	0.1	197	2510	129
	TD	0.1	175	2510	134
398	ID	0.1	153	2110	120
	TD	0.1	145	21110	116

Fig.12 Variation of k_1 with (a) strain rate at 298K and (b) temperature at 0.1%/s

Fig.13 Variation of k_2 with temperature at 0.1%/s

Table 3 Constants in Eq. (7)

Cutting direction	C_1 , MPa·s	C_2 , MPa	C_3 , MPa	C_4 , MPa	ΔQ_1 , J/mol	ΔQ_2 , J/mol	ΔQ_3 , J/mol
ID	921	40.1	898	82.9	4360	2900	1500
TD	570	37.0	898	79.6	4370	2900	1800

Fig.14 Variation of η (a) strain rate at 298K and (b) temperature at 0.1%/s

Fig.15 Comparison of stress-strain curves between test data and the model in the ID

Substituting Eqs. (4), (5) and (6) into Eq. (1) gives the stress-strain relationship of the 3-element Maxwell model as follow.

$$\sigma = C_4 \left(\frac{d\varepsilon}{dt} \right)^{0.02} \exp\left(\frac{\Delta Q_3}{RT}\right) \left(1 - e^{-\frac{C_3 \varepsilon}{C_4 \left(\frac{d\varepsilon}{dt} \right)^{0.02} \exp\left(\frac{\Delta Q_2 - \Delta Q_3}{RT}\right)}} \right) + \left(C_1 \frac{d\varepsilon}{dt} + C_2 \right) \exp\left(\frac{\Delta Q_1}{RT}\right) \varepsilon \quad (7)$$

Table 3 lists values of the constants in Eq. (7).

In order to check the accuracy of estimating the stress-strain relationship by Eq. (7), Figure 15 compares predicted stress-strain curves by Eq. (7) with the experimental data at three strain rates and three testing temperatures in the ID. Predicted curves were indicated by the solid lines in the figure. The predicted curves were almost same as those of the experimental data, although the predicted stresses were slightly smaller than those of the experiment at 298K and 0.1%/s. The results indicate that the 3-element Maxwell model applied in this study satisfactorily describes the stress-strain curve of the polyimide film.

4. Conclusions

(1) There was little anisotropy of Young's modulus, proportional limit, yield stress, ultimate tensile strength and elongation in the polyimide film used in this study. Young's modulus of the polyimide film was similar to that of the bulk polyimide. Ultimate tensile strength of the film was larger than that of the bulk polyimide, but elongation was smaller.

(2) There was a little effect of strain rate on Young's modulus but proportional limit, yield stress and ultimate tensile strength increased with increasing strain rate. Only elongation decreased with strain rate. Young's modulus, proportional limit, yield stress and ultimate tensile strength decreased with increasing temperature, but elongation increased.

(3) A 3-element Maxwell viscoelastic model consisting of two spring elements and a dashpot element was applied. The model satisfactorily described the stress-strain curve of the polyimide film.

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