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Effects of quinoline on the imidization temperature and properties of polyimide

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Abstract
Polyimide (PI) with excellent mechanical, thermal, dielectric properties which has been widely applied in electronics, automotive, high-speed rail and chemical industry. However, the high curing temperature has limited its application in the micro-electronic areas. To address this issue, quinoline (QL) with unique catalytic effect was employed in low temperature curable PI. The changes in chemical structure of the PI films at different temperature was confirmed by FTIR spectroscopy with controlled addition of QL. The effects of QL content on the electrical, thermal and mechanical properties of polyimide were discussed as well. It was found that PI-1.5 has been proved with the best property which exhibited high imidization rate of 99.6% at 200 °C, dielectric constant of 3.20 and a dielectric loss of 5.06 × 10⁻³ (1 MHz). Besides, the Young’s modulus of as much as 1.66 GPa, elongation at break of more than 145% with high tensile strength about 134 MPa have been achieved. Moreover, the T₅ is more than 545 °C and the glass transition temperature is higher than 376 °C. The developed low temperature curable PI is expected to be applied in the advanced electronic packaging such as fan-out wafer package, insulation, interlayer dielectric and so forth.

1. Introduction

Polyimides (PIs) are very promising engineering polymers, exhibiting high-temperature resistance, low-temperature tolerance, outstanding mechanical properties, chemical and radiation resistance, flexibility, and excellent dielectric properties. Hence, Polyimides have been widely applied in many high-tech fields such as electrical insulation, nuclear energy industry and microelectronics industry as shown in figure 1 [1–4]. In traditional, PI preparation process includes two steps: the first step is to synthesize polyamic acid (PAA) precursor from the dianhydride and diamine monomer in polar solvent [5]; the second step is to gradually temperature stepped low-temperature curing at about 350 °C to obtain the final polyimide material [6]. However, as it can be seen, PAA requires a high-temperature imidization process at 350 °C in the traditional preparation method which has the following drawbacks: first, high curing temperature consumes a lot of energy; second, high curing temperature easily destroys the internal stress of the film and diminishes its mechanical properties [7]; third, high curing temperature also result in the solder joint cracking, peeling off, recrystallization of low melting solder and even destroy the devices. Therefore, researches focused on low-temperature curable PI which would possess a great market prospect in the short future.

There are two typical methods for developing low temperature curable (about 200 °C) PIs. (1) One-step method: the dianhydride and diamine are directly polymerized and imidized in a high boiling solvent. For example, Inoue et al [8] employed biphenyl-tetraacarboxylic dianhydride (BPDA) and 4,4′-diaminodiphenyl
ether for the polymerization and imidization in p-chlorophenol and the as-prepared PI exhibited a tensile strength of 205 MPa and elongation at break of 80%. Then, Kuznetsov et al. [9] explored the effect of various factors on the degree of solidification of PI by one-step synthesis in benzoic acid, they found the rate of cyclization of transient polyamic acids is very high in benzoic acid at 140 °C. The key of this method is to choose a suitable solvent, but most high-boiling solvents reported contain carcinogens which are not suitable for large-scale production and the thermal stability is relatively limited. (2) Temperature stepped low-temperature curing: (a) Design of flexible molecules: Low temperature curable polyimide resin CT4112 produced by Toshiba Chemical Company of Japan has been imidized at low temperature (<200 °C) due to the introduction of soft segment structure in PI molecular chain. (b) Control of acidity: the acidity control of amine unit of precursor is very effective. For example, reducing the acidity of the segment can significantly reduce the curing temperature. (c) Low-temperature curing accelerator: In recent years, researchers have turned their attention to investigate the low temperature curable PI in the dehydrating agents and catalysts. With these additives, PAA could realize the dehydration at lower temperatures followed by closed-loop imidation which is a common chemical curing method. The dehydrating agent may be an aliphatic or aromatic acid, and the imidization catalyst could be the aliphatic tertiary amine represented by triethylamine, or aromatic tertiary amine represented by N,N-dimethylaniline, and pyridine or heterocyclic tertiary amine represented by picoline, quinoline or isoquinoline. For example, Oba et al. [10] studied the catalytic effects of 26 additives at 100 °C, and found p-hydroxybenzoic acid, 2,4-dihydroxybenzoic acid, p-hydroxybenzoic acid, those results proved excellent catalytic effect. The reported catalysts are all aromatic compounds with more than two reactive groups. And then, Nelson et al. [11] employed benzimidazole and heated at 100 °C for 24 h, the results showed that PAA was almost completely imidized. Compared with imidation with high-temperature, this method can be carried out at a low temperature (<200 °C) and shortening curing time, so it is widely used in large-scale production on the assembly line. However, adding an excessive amount of curing reagent may affect the performance of the materials.

In this work, investigations on the effects of different weigh ratio of quinoline (QL) in the imidization temperature, electrical, thermal and mechanical properties of polyimides were carried out. The best performance of polyimide was obtained from PI-1.5. This study is expected to have influences on the preparation of low temperature curing polyimide in the respect of both theoretical and applicable system.

2. Experimental details

2.1. Materials
Pyromellitic dianhydride (PMDA) and 4,4′-oxydianiline (ODA) were purchased from TCI (Tokyo, Japan). QL was purchased from Sinopharm Chemical Reagent Co., Ltd, NMP solvents used were obtained from Aladdin.
2.2. Preparation of PAA solution
In the case of 25 °C, nitrogen protection, 10.00 mmol of ODA was added to 32.92 g of NMP. After the ODA was completely dissolved, 10.20 mmol of PMDA was added in three portions at intervals of 0.5 h, and then the solution was thoroughly stirred for 4 h, and QL was added. Then, the PAA solution was obtained by stirring for 1.5 h. The content is shown in Table 1.

2.3. Preparation of PI films
The PAA solutions to which QL was added were spin-coated on a glass slide, and then heated in an oven for 1 h at 50 °C, 80 °C, 120 °C, 150 °C, 180 °C, 200 °C, respectively, after then cooled to room temperature.

2.4. Characterization
FTIR transmission spectra were recorded using Vertex 70 (Bruker, Germany). The spectra were recorded between 4000–600 cm⁻¹. An uncoated part of the Si wafer was used as back ground. Thermal gravimetric analyses of the polyimide films were carried out with a STD Q600 (TA Instruments, America) at a heating rate of 10 °C min⁻¹ with a nitrogen purge. Measurements of glass transition temperatures for the films were carried out using a DSC Q20 (TA Instruments, America) at a heating rate of 10 °C min⁻¹ in a nitrogen atmosphere. Mechanical properties were measured by DMA Q800 (TA Instruments, America), ranging from 1 N min⁻¹ to 18 N. Coefficients of thermal expansion (CTE) for the films were carried out using DMA Q800 (TA Instruments, America) at a heating rate of 5 °C min⁻¹ from 50 °C to 200 °C with force 0.001 N in a nitrogen atmosphere. The dielectric constant measurements for PI films were carried out in a sandwich structure of gold (Au)/polyimide/gold (Au). The top Au electrode, with an area of 19.625 mm², was deposited by thermal evaporation in a vacuum at 4 × 10⁻⁶ Pa on polyimide films. Then, these films were dried at 130 °C under vacuum for 2 days. The thicknesses of PI films are about 20 μm. The dielectric constant (εr) and dielectric loss of the films were measured on an Agilent 4294A impedance analyzer at various frequencies and ambient temperature. Prior to measurements, the surface of each film was coated with silver paint to form electrodes. εr was calculated by the following equation:

\[ ε_r = \frac{(C × d)}{(ε_o × S)} \] (1)

where C, d, and S denote the capacitance, thickness, and electrode area of the composite film, respectively, and ε_o is the permittivity of free space (8.854 × 10⁻¹² F m⁻¹).

3. Results and discussion
3.1. Infrared spectroscopy analysis of imidization
The FTIR spectra of the imidization process of PAA with QL and without QL are shown in Figure 2, and the main IR band assignments for the PAA and PI are shown in Table 2. It can be seen that for PAA, since 1660 cm⁻¹ represents the carbonyl vibration of the amide group, 1545 cm⁻¹ represents the variable angular vibration and CH stretching vibration of NH in the amide group. Therefore, as temperature rises, the two absorption bands of 1660 and 1545 cm⁻¹ gradually decrease, but the sample to which QL is added almost completely disappears at 180 °C, as shown in Figure 2(a). However, the sample without QL added 1660 and 1545 cm⁻¹ completely disappeared at 300 °C, as shown in Figure 2(b). In addition, the two absorption bands of 1776 and 1378 cm⁻¹ gradually increase with increasing temperature. In fact, they are the characteristic absorption bands of polyimide: 1776 cm⁻¹ is the in-phase (or symmetric) stretching vibration of two carbonyl groups on a five-membered imine ring, also known as imide I band; 1378 cm⁻¹ is C=N stretching vibration in polyimide [12–14], also known as imide II band, these results indicate that the imidization can be accomplished at about 200 °C by adding a low temperature curing accelerator. Additionally, the strongest absorption at 1500 cm⁻¹ is the ring stretching vibration of the benzene ring in the aryl ether, which shows almost the same intensity during the cyclization process. All these characteristic peaks and the variation in the FTIR spectra are compared with the published literatures [15–17]. Then, from the maxima of the imide bands the degree of imidization α can be calculated, which is defined [18] as:

\[ \text{Imide Index} = \frac{\text{Absorbance 1378 cm}^{-1}}{\text{Absorbance 1500 cm}^{-1}} \] (2)

Figures 2(c) and (d) show that the imidization rate of PAA with QL is 99.4% at 200 °C, while the imidization rate of PAA without the addition of QL was close to 99.3% at 350 °C. The above phenomenon shows that QL can significantly reduce the imidization temperature of PAA.

It can be seen from Figure 3 that the degree of imidization of PI with different QL dosages is greater than 90%, all of which have characteristic absorption peaks of PI, and the degree of imidization increases first with the
increase of QL dosage. Then basically remain unchanged. This may be because there are some small molecular oligomers in the PAA molecule formed by diamines and dianhydrides, in which the anhydride does not react as a terminal group, reducing the number of carboxylic acid C atoms in the molecular chain, so the actual catalysis is required. QL is lower than the theoretical value. When the ratio of the amount of the dianhydride monomer to the amount of the QL added is 1:1, the amount of the catalyst is insufficient, and the degree of imidization of PI is only 93.6%. When the ratio of the amount of the dianhydride monomer to the amount of the QL added is 1:1.5, the imidization reaction is substantially completed, and the degree of imidization is maximized to be 99.6%.

In figure 4, the catalytic mechanism of QL was proposed which served as an affinity reagent to attack the carbon atom on carboxyl group in PAA, which transfer the hydrogen atom to the carboxyl group, thereby trap
hydrogen on the amide bond, and promote cyclization dehydration of the acid group. Finally, the imidization reaction completed [19].

3.2. Contact angle
It can be seen from figures 5(a)–(f) that the contact angle [20] between water and PI is obviously reduced. The most likely cause of this phenomenon is that the degree of imidization is incomplete, that is, the carboxyl group is incompletely completely cyclized with the amino group, resulting in a large amount of carboxyl groups remaining, causing the remaining carboxyl groups and water to generate hydrogen bonds, thereby making the wetting effect change and contacting. As the degree of imidization increases, the contact angle also increases, because most of the carboxyl groups complete the cyclization reaction, and thus the contact angle becomes large showed in figures 5(d) and (e), which also indicates that PI-1.5 and PI-2 can achieve degree of imidization. This result is also consistent with the observation by infrared spectroscopy.

3.3. Electrical properties
As shown in figure 6 and table 3, the dielectric constant of PI continuously decreases with the increase of quinoline, which is mainly because of the degree of imidization. In the case of PI-0.8, PI-1 and PI-1.2, the imidization is insufficient due to the small quinoline, and there are many polyamic acids, and the polarity is large, so the dielectric constant is large. As the quinoline content increases, the degree of imidization gradually increases, the polarity becomes smaller, and the dielectric constant becomes smaller [21–24].

The dielectric loss of the composite film is gradually increased relative to the dielectric loss of the pure PI film. The reason for this phenomenon is that in the high frequency region of 1 MHz, the alternating electric field...
changes very rapidly, the polar groups and segments can not keep up with the change of the alternating electric field, there is a hysteresis delay, causing a large amount of relaxation loss, resulting in an increase in dielectric loss. As the quinoline content increases, more relaxation loss is induced, so the dielectric loss of the composite film increases as the molecular sieve content increases.

![Figure 5](image)

Figure 5. Contact angle of water and polyimide. (a) PI-0 (b) PI-0.8 (c) PI-1 (d) PI-1.2 (e) PI-1.5 (f) PI-2.

![Figure 6](image)

Figure 6. (a) Dielectric constant change of different quinoline content. (b) Dielectric loss factors change of different quinoline contents.

| Sample name | Dielectric constant (1 MHz) | Dielectric loss factors (10⁻³) (1 MHz) |
|-------------|-----------------------------|----------------------------------------|
| PI-0        | 3.21 ± 0.09                 | 1.84                                   |
| PI-0.8      | 3.64 ± 0.05                 | 4.85                                   |
| PI-1        | 3.62 ± 0.06                 | 5.06                                   |
| PI-1.2      | 3.32 ± 0.08                 | 5.41                                   |
| PI-1.5      | 3.20 ± 0.09                 | 5.98                                   |
| PI-2        | 3.22 ± 0.04                 | 6.00                                   |
Figure 7. Mechanical properties of PI coating films with different QL contents: (a) stress-strain curves of different content of QL contents. (b) Distribution of tensile strength elongation modulus with different contents of QL.

Figure 8. Thermal properties of PI coating films with different QL contents: (a) TGA curves for different QL contents (b) the distribution of weight loss 5%, Glass transition temperature and thermal expansion coefficient for different QL contents.

Table 4. Effect of different QL amounts on tensile strength, elongation at break and Young’s modulus of PI.

| Sample name | Tensile strength (MPa) | Elongation (%) | Modulus (GPa) |
|-------------|------------------------|----------------|---------------|
| PI-0        | 191.81                 | 148            | 1.68          |
| PI-0.8      | 98.39                  | 45             | 1.20          |
| PI-1        | 107.10                 | 53             | 1.23          |
| PI-1.2      | 113.35                 | 88             | 1.55          |
| PI-1.5      | 134.82                 | 145            | 1.66          |
| PI-2        | 132.98                 | 114            | 1.54          |

Table 5. Effect of different QL contents on weight loss 5%, glass transition temperature and thermal expansion coefficient.

| Sample name | $T_d$(°C) | $T_g$(°C) | CTE(20°C—100°C) (ppm/°C) |
|-------------|-----------|-----------|----------------------------|
| PI-0        | 530       | 380       | 35                         |
| PI-0.8      | 419       | 297       | 30                         |
| PI-1        | 470       | 345       | 20                         |
| PI-1.2      | 544       | 350       | 17                         |
| PI-1.5      | 545       | 376       | 24                         |
| PI-2        | 537       | 381       | 19                         |
Table 6. Recent works on literature comparison.

| References | Thermal properties | Mechanical properties | Electrical properties | Curing conditions |
|------------|---------------------|-----------------------|-----------------------|-------------------|
|            | $T_5$ (°C) | $T_g$ (°C) | CTE (20 °C–100 °C) (ppm/°C) | Tensile strength (MPa) | Elongation at break (%) | Modulus (GPa) | Dielectric constant (1 MHz) | Dielectric loss factors (10–3) (1 MHz) | $T_c$ (°C) | Curing accelerator |
| [30]       | 512      | —           | — | 9 | 12 | 18.1 | — | — | — | 280 | Et$_3$N |
| [31]       | —        | —           | — | 4 | 0.8 | 36.18 | 1.3 | — | — | 280 | Et$_3$N |
| [32]       | 487      | —           | — | — | — | — | — | — | 200 | DBU |
| [33]       | —        | —           | — | — | — | — | — | — | 200 | DBU |
| [34]       | 510      | 225         | — | — | — | — | — | — | 220 | QL |
| [35]       | —        | —           | — | 26.37 | 1.26 | 1.09 | — | — | 200 | DBU |
| [36]       | 573      | —           | — | 6.6 | 19.85 | 29.18 | 1.3 | 2 | — | Et$_3$N |
| This paper | 545      | 376         | 24 | 134.82 | 145 | 1.66 | 3.22 ± 0.04 | 6 | — | QL |

$T_5$: Weight loss 5%. $T_g$: Glass transition temperature. $T_c$: Curing temperature. CTE: Coefficient of thermal expansion. —: Not given.
QL: Quinoline. Et$_3$N: Triethylamine. DBU: 1,8-diazabicyclo[5.4.0]undec-7-ene.
3.4. Mechanical properties
As can be seen from figure 7 and table 4, the tensile strength, modulus and elongation at break of the PI film first increase and then decrease as the QL content increases. PI-0.8 has a tensile strength of 98.39 MPa, an elongation at break of 45%, an elastic modulus of 1.20 GPa, and poor mechanical properties. However, as the QL content increases, the tensile strength of PI-1.5 increases to 134.8 MPa, the elongation at break increases to 140%, and the modulus increases to 1.66 GPa, respectively, and the mechanical properties are greatly improved. Then, as the QL content continues to rise, the performance of PI-2 is slightly reduced. The possible reason for the above is that when the amount of QL added is insufficient, the imidization reaction is incomplete, resulting in poor mechanical properties of the material. As the QL increases, the imidization of the PAA gradually completes, resulting in excellent mechanical properties. However, since the boiling point of QL at room temperature is 238.05 °C and the imidization temperature is 200 °C, excessive addition of QL will result in QL residue in the PI film, destroying the interaction between PI molecules, thereby reducing the mechanical properties of the material [25, 26].

3.5. Thermal properties
As can be seen from figure 8 and table 5, PI-0.8 has a Tg of only 419 °C, a glass transition temperature (Tg) of 297 °C, a low degree of imidization of the PI film, and poor thermal stability of the material and easy decomposition [27–29]. However, as the QL content increases, the Tg of PI-1.5 rises to 545 °C, and the glass transition temperature (Tg) reaches 376 °C. As QL continues to rise, the Tg and Td of PI-2 are almost the same as PI-1.5. This phenomenon also indicates that the degree of imidization of PI is increasing. When the molar ratio of dianhydride to QL is 1:1.5, the thermal stability properties of the material are optimized, and then as the QL content increases, the thermal stability generally does not change much. It is worth observing that the CTE of PI-0 is 35 ppm °C, which is the CTE of pure PI. When QL was introduced, the CTE of PI-1.5 was reduced to 24 ppm °C, which is very close to copper (17 ppm °C) and silicon (25 ppm °C). For packaging materials, CTE matching is not important, because the difference in CTE can lead to warpage or even failure of the material, and the reduction of CTE can be instructive.

It can be seen from table 6 that for the published work, this work gives a more comprehensive property determination for the influence of low-temperature curing agent on PI material, and also shows that QL has better catalytic effect compared with triethylamine and 1, 8-diazabicyclo [5.4.0]undec-7-ene.

4. Conclusion
In this work, the introduction of QL can significantly reduce curing temperature of the polyamic acid. The PI films with superior thermal and mechanical properties were successfully obtained. Furthermore, the addition of QL can significantly reduce CTE, which will reduce residual stress in the advanced packaging applications. In addition, the tensile strength, elongation at break and modulus of elasticity of the PI film are also improved. As mentioned above, the addition of QL proved broad application prospects for low temperature curing polyimide materials.

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