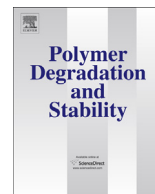




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Dielectric property and charge evolution behavior in thermally aged polyimide films

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ABSTRACT

Polyimide (PI) materials are widely used in modern industry due to their excellent flexibility, outstanding performance, and resistance to heat and chemicals. However, thermal degradation of PI materials has become a great challenge while its mechanism and effect on the macroscopic performance still remain uncertain. Here, we comprehensively address this issue by thermally aging PI (TA-PI) films at 300 °C in air for different periods and carrying out microstructure, thermal, dielectric, and space charge characterizations to obtain a deep insight. It was found that oxidation in the aromatic rings, charge transfer complex, and microvoids formed during thermal aging were responsible for the color deepening and opacity. Thermogravimetric analysis revealed that both low-mass volatile and non-volatile pyrolysis products were generated with the prolongation of thermal aging, thus resulting into increased relative permittivity and dielectric loss, decreased breakdown strength (36.6% lower), suppressed space charge, and restricted internal field distortion ($\leq 6\%$). Low-mass polar molecules are the key to deteriorating the dielectric property of PI materials. The obtained results could provide helpful support for the design and development of PI materials for future engineering applications.

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1. Introduction

Due to its outstanding properties, such as high thermal and chemical stability, low dielectric loss, excellent mechanical performance, and ease of fabrication, polyimide (PI) is used in a wide variety of industrial applications since it was put on the market in 1965 [1,2]. The demand of electrical, electronic, and aerospace engineering applications for polymers which could be suitable and stable at elevated temperatures even higher than 300 °C (572 F) significantly grows nowadays, which, generally exceeds the limit of thermal stability of most polymers but could be addressed using PI materials [3–5]. In practice, PI materials, however, still encounter

an ongoing decrease of macroscopic performance due to long-term degradation processes, such as thermal, irradiated, and electrical degradations [6–9]. Among these, thermal degradation is the most common but challenging for PI materials, the relationship between which and macroscopic performance has been an important question in past decades.

Plentiful investigations have been performed to study the influence of long-term thermal degradation on the morphology, microstructure, and thermal degradation kinetics of PI materials. It is well-appreciated that thermal exposure of PI films at elevated temperatures in the early stage was dominated by oxidative degradation, and PI films became more brittle, darker, and thicker with longer exposure times or higher heating temperatures due to the formation of oxidized layers on the surface [7,10]. Moreover, thermal degradation kinetics were comprehensively studied and several mechanisms were proposed. To name a few, Li et al. [11] and Meng et al. [12] proved that all four kinds of (maleic anhydride + hexafluoroisopropylidene 2,2-bis (phthalic

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anhydride) + 4,4'-oxydianiline) PIs and PI containing 2, 6-benzobisoxazole units obeyed the nucleation and growth models. Tiptipakorn et al. proposed the thermal degradation mechanism of polybenzoxazine and silicon-containing PI blends to be random nucleation with one nucleus on the individual particle [13]. Ferreira et al. pointed out that roughcast-like appearance was acquired and became more pronounced with the thermal treatment time due to the weight loss from CO₂ gas evolution [14]. Khazaka et al. reported the decrease of average thickness of 0.5 nm/h under thermal treatment at 300 °C, suggesting that thermal degradation was related to a surface degradation process [15].

Although abundant studies have been carried out on the dielectric properties of PI materials [5,16,17], there are few focusing on the role of thermal aging on dielectric properties, which are still uncertain and sometimes even controversial. Oxidative reaction produced free radicals and polar groups such as carboxylic acid ketones and aldehydes, which led to the increase of dielectric constant ϵ_1 and dissipation factor $\tan\delta$ [18,19]. When dielectric constant ϵ_1 increased, the volume resistivity would greatly decrease. The change of dielectric constant ϵ_1 , hence, impacted on other dielectric properties of PI samples [20]. Dielectric breakdown strength of PI films greatly decreased with the rise of thermal aging time or temperature (450–480 °C) of thermal exposure due to the pyrolysis of imide groups [21]. However, it was also reported that dielectric breakdown strength increased from 370 MV/m to 470 MV/m via thermal aging at 300 °C between 0 and 100 h [22]. Similar results of enhanced dielectric breakdown strength due to thermal aging at 350 °C in air were also reported previously [23]. As for the pre-breakdown current, it is suggested that there was not any increase after thermal aging at 300 °C in 1.5 μm -thick PI films at room temperature [15]. While this knowledge exists, the consequence of thermal aging on the electrical transport and charge evolution in PI materials and the numerous parameters that control them remain unexplored at present.

The scope of the present work critically focuses on the relationship between thermal aging and dielectric property of PI films, especially including charge evolution behavior. Thermally aged PI (TA-PI) films at 300 °C in air with different aging periods were prepared. Based on the analysis of morphology, thermal, mechanical, and dielectric characteristics in this work, we attempt to obtain a deep insight into the thermal aging process and its effects on the electrical transport and charge evolution behavior in TA-PI films. Finally, a possible process of thermal aging of PI films is proposed.

2. Experimental

2.1. Materials

In this work, commercially available PI films (Jinlvye Corp., Shenzhen, China), a linear polymer comprising heterocyclic rings linked together by one or more covalent bonds, were investigated. The molecular structure of PI films provided by the company is shown in Fig. 1, which is the same with that of Dupont Kapton PI films. The thickness and the width of PI films are (208 \pm 2 μm) and 50 mm, respectively. The relative permittivity is about 3.6.

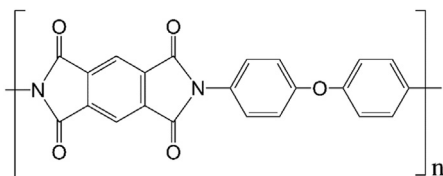


Fig. 1. Molecular structure of PI films.

2.2. Thermal aging of PI films

PI films were thermally aged at 300 °C within an oven in air for different times, i.e., 10 days, 20 days, 30 days, and up to 40 days. For convenience, abbreviation symbols were adopted for the samples, namely PI_0d, PI_10d, PI_20d, PI_30d, and PI_40d for pristine PI films, 10-day, 20-day, 30-day, and 40-day TA-PI films, respectively.

2.3. Methodology

2.3.1. Field emission scanning electron microscopy (FESEM)

FESEM was performed using a JSM-6335 coupled with an energy dispersive X-ray (EDX) module to observe the microstructure and element composition profiles of the surface and fractured faces of PI film samples. In order to obtain fractured faces, PI films were firstly immersed in liquid nitrogen and subsequently broken up. All samples were coated with platinum for 10 min. During FESEM observation and EDX analysis, the voltage was set as 15 kV. The magnification times were set as 1000 \times and 5000 \times for each observation spot.

2.3.2. Atomic force microscope (AFM)

AFM images were acquired in tapping mode using a silicon probe via a Bruker microscope (model Dimension Icon) in air with a scan rate of 1 Hz. The spring constant is about 40 N/m and the oscillating frequency is 320 kHz. The AFM images, used to determine the surface roughness average (R_a) and skewness (R_q) of PI film samples, cover small areas of 5 μm \times 5 μm in order to reflect the overall roughness conditions [24].

2.3.3. Fourier transformed infrared (FTIR) spectroscopy

FTIR spectra were obtained via a Thermo Nicolet NEXUS 650 in reflection mode. The resolution was set as 2 cm^{-1} and each acquired spectrum was the average of 16 scans. The background spectrum of the atmosphere was measured and automatically subtracted from sample spectra. Spectra ranging from 650 cm^{-1} to 4000 cm^{-1} were focused and analyzed considering the severe distortion of FTIR spectra below 650 cm^{-1} brought by the ZnSe window tablet.

2.3.4. X-ray photoelectron spectroscopy (XPS)

X-ray photoelectron spectroscopy (XPS, Thermo Scientific Escalab 250Xi) was used to analyze chemical bonding states of both initial and thermally aged PI surfaces. The XPS tool used conventional Al K α X-ray excitation, while the C1s peak of graphite (at 284.6 eV) was used as internal standard during measurements. The obtained spectra were curve-fitted with Gaussian peaks after subtracting the Shirley background.

2.3.5. Thermal property measurement

2.3.5.1. Thermogravimetric analysis (TGA). Thermogravimetric analysis of PI film samples were obtained via a TA Q50 TGA system. PI film samples, weighing in the range of 5–10 mg, were heated under a 90 mL/min N₂ flow from 20 °C to 900 °C to monitor the weight change. The heating rate was set as 15 °C/min.

2.3.6. Tensile property measurement

Tensile properties of PI film samples were tested at room temperature (25 \pm 1 °C) according to American Society for Testing Material (ASTM) D638. The reported tensile strength is the average value of at least five specimens for each group. PI films were cut into stripes with the width and the length of 15 mm and 200 mm, respectively.

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