Progress in Organic Coatings 124 (2018) 92–98

Contents lists available at ScienceDirect

Progress in Organic Coatings

journal homepage: www.elsevier.com/locate/porgcoat

Phosphinated polyimide hybrid films with reduced melt-flow and enhanced adhesion for flexible copper clad laminates

I-Hsiang Tseng[a], Tsung-Ta Hsieh[b], Ching-Hsuan Lin[c], Mei-Hui Tsai[b,*], Dai-Liang Ma[d], Cheng-Jung Ko[d]

[a] Department of Chemical Engineering, Feng Chia University, Taichung, 40724, Taiwan
[b] Department of Chemical and Materials Engineering, National Chin-Yi University of Technology, Taichung, 41170, Taiwan
[c] Department of Chemical Engineering, National Chung Hsing University, Taichung, 40227, Taiwan
[d] Materials and Electro-Optics Research Division, National Chung-Shan Institute of Science & Technology, Taoyuan, 32546, Taiwan

ARTICLE INFO

Keywords:
Polyimide
Phosphinate
Adhesion strength
Melt flow

ABSTRACT

Adhesion properties of phosphinate-contained polyimide (P-PI) hybrid films were studied to develop suitable heat resistant adhesive film with excellent hot-melt processability. The diamine, 1,1-bis(4-aminophenyl)-1-(6-oxido-6H-dibenz < c, e > < 1, 2 > oxaphosphorin-6-yl) ethane (i.e. DMA), was introduced to synthesize a series of P-PI films with different contents of DMA. The presence of the bulky phosphinate pendant group on PI backbone increased the adhesion strength and effectively reduced the melt flow after hot-pressing to flexible copper clad laminates (FCCL) without the usage of adhesive. The P-PI film tended to produce rougher surface that facilitated the mechanical interlocking effect between P-PI hybrid film and copper. The presence of characteristic phosphinate functional groups and the formation of polyphosphate layer on P-PI film were confirmed by FTIR and XPS results. The storage modulus and glass transition temperature of P-PI films were increased because of the enhanced rigidity by phosphinate groups. The presence of bulky phosphinate pendant group also reduced the coefficient of thermal expansion and dielectric constant of PI films. Although the introduction of phosphinate in PI reduced the initial weight loss temperature, the thermal stability of P-PI films was sufficient for the following manufacturing processes. Furthermore, another interesting observation was the enhancement of transparency at 400 nm for P-PI. Those properties make P-PI films useful in FCCL application.

1. Introduction

Polyimides (PIs) possess many outstanding properties such as good chemical resistance, low dielectric constant, excellent mechanical properties and high thermal stability. These characteristics make PIs be widely utilized in many applications. However, the intractability of PIs, such as insolubility in most organic solvents and poor processibility, still restricts their applications. In order to improve the processing properties of PI, many research efforts, for example, introduction of bulky substituents or flexible linkage, using fluoro-containing monomers and nonsymmetrical monomers have been investigated for synthesis of soluble polyimides [1–10]. In some research, phosphorus-containing diamines were utilized to synthesize PIs because of their bulky and polar groups as well as a non-coplanar structure which would increase their solubility [1,11,12]. Furthermore, the thermal oxidative stability and flame retardancy of phosphorus-containing PI were also improved [11,13–16].

Lin et al. previously synthesized novel phosphinate-functionalized aromatic diamine, 1,1-bis(4-aminophenyl)-1-(6-oxido-6H-dibenz < c,e > < 1,2 > oxaphosphorin-6-yl)ethane (DMA). The resulting PI showed flexible, high glass transition, high modulus, low coefficient of thermal expansion, moderate thermal stability and good flame-retardant property [16]. In the present work, a series of PI hybrid films with various DMA ratios were prepared and their potentials to apply for flexible copper clad laminates (FCCL) were evaluated. To meet the demand of highly miniaturized electronic devices, two-layer and direct-metallization without any adhesive layer between PI and copper are required. Earlier studies have shown that the adhesion strength of PI and copper could be enhanced by surface modification to form new chemical bonding [17–21]. Another strategy is modifying the surface roughness to utilize mechanical interlocking effect [19,22,23]. There are few references mentioned the effects of phosphinate-containing PI on adhesion strength and melt flow. Therefore, the objective here is to study the correlation between the ratios of DMA on adhesion strength,

* Corresponding author.
E-mail address: tsaimh@ncut.edu.tw (M.-H. Tsai).

https://doi.org/10.1016/j.porgcoat.2018.04.037
Received 28 November 2017; Received in revised form 29 March 2018; Accepted 29 April 2018
0300-9440/ © 2018 Elsevier B.V. All rights reserved.

melt flow amount and surface morphology. In addition, the chemical structure, optical transmittance and thermal properties of the resulting PI hybrid films were characterized and discussed in this work.

2. Experimental

2.1. Materials

4, 4′-diaminodiphenylether (ODA, purity: 97%) from Taiwan Taimide Tech Inc. was dried at 85 °C in a vacuum oven for 24 h prior to use. Phosphinate-contained aromatic diamine 1,1-bis(4-aminophenyl)-1-(6-oxido-6H-dibenz < c,e > < 1,2 > oxaphosphorin-6-yl)ethane (DMA) was obtained from Prof. Lin [16] and dried at 85 °C in a vacuum oven for 24 h prior to use. Dianhydride 5,5′-(3,3′-(propane-2,2-diyl) bis (3,1-phenylene)) bis (oxy) diisobenzofuran-1,3-dione (IDPA, purity: 97%) from Sigma-Aldrich was purified by recrystallization. Dimethylacetamide (DMAc) from TEDIA was dried over sodium aluminosilicate molecular sieves (4 Å).

2.2. Synthesis of phosphinate-contained PI

The procedures of preparing phosphinate-contained PI films were shown in Scheme 1 . Poly(amic acid) (PAA) solution were prepared by reacting equal molar amounts of diamine and dianhydride in solution (solid content w/w = 15% for DMA/ODA-IDPA in DMAc). The reaction was carried out in a three-neck flask by adding the diamine (DMA/ODA) and the dianhydride (IDPA) in solution under a nitrogen stream at 0–3 °C. After the complete dissolution of IDPA, the reaction mixture was further stirred for at least 12 h. Thus, the PAA precursor was obtained. The freestanding PI hybrid films were made by casting the PAA precursor onto a dust free glass plate with an automatic film applicator equipped with spreaders of adjustable gap and drawing speed. The wet films were then step-heated at 100 °C, 150 °C, 200 °C and 250 °C, for 1 h at each temperature, and finally at 300 °C for 2 h in an air-circulating oven. Upon cooling, the PI hybrid films were removed from the glass plate. Consequently, the PI hybrid films have an average final thickness about 25μm.

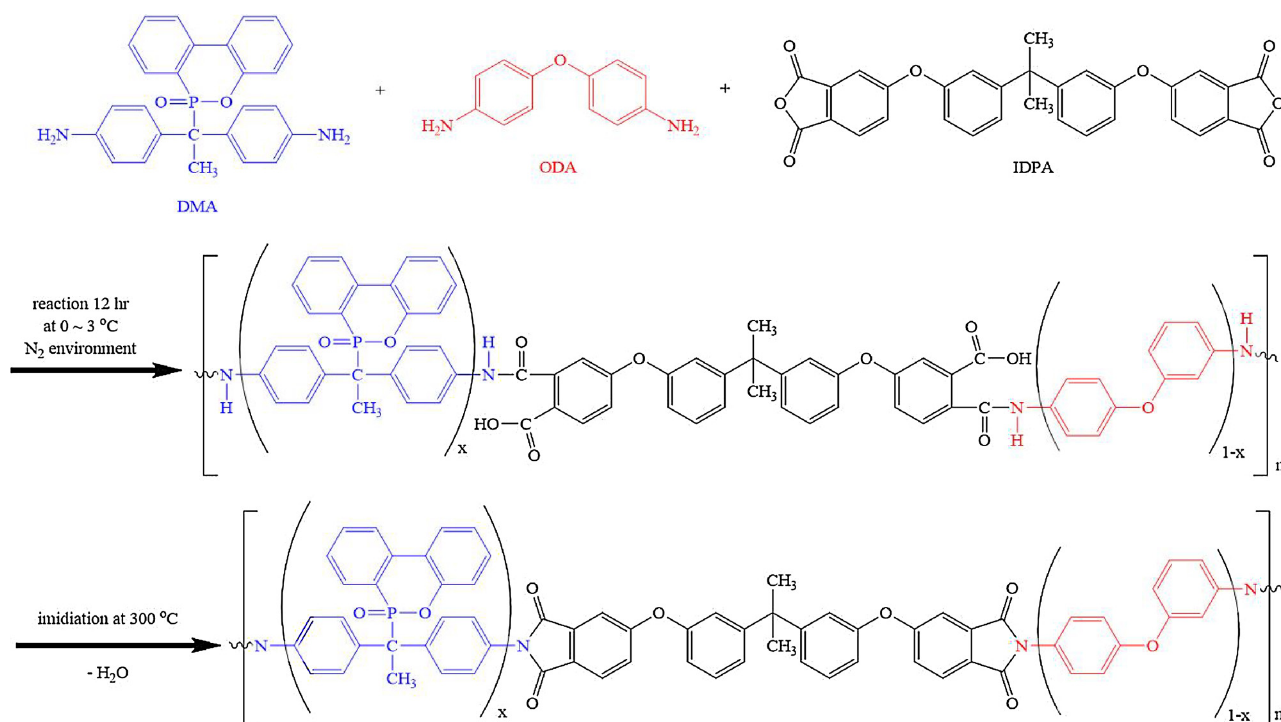
Table 1
Sample code and molar ratios between DMA and ODA.

Sample	Molar Ratio of Diamine	
	DMA mole (%)	ODA mole (%)
10D	100	0
8D2O	80	20
5D5O	50	50
2D8O	20	80
10O (pure PI)	0	100

In this study, the sample name was denoted xDyO to indicate the composition of DMA and ODA in PI films. The letters D and O were used to denote diamines of DMA and ODA, respectively, with the molar ratio of x and y. For example, 5D5O represents 50% of DMA and 50% of ODA were applied during the synthesis of PI film. Notably, the sample 10O was the pure PI synthesized by only ODA without the composition of DMA. The sample codes of the resulting PI films with the corresponding diamine molar ratios were summarized in Table 1.

2.3. Characterizations

Fourier transform infrared (FT-IR) spectra of PI films at different temperatures were obtained by an infrared spectrophotometer (Thermo Nicolet 380). Ultraviolet-visible (UV–vis) spectroscopy was recorded between 200 and 800 nm with a scanning rate of 2 nm/s. The thermal mechanical analysis (TMA, TA-Q400) was performed at a tensile force of 0.1 N and a heating rate of 10 °C/min from 30 to 300 °C. The values of coefficient of thermal expansion (CTE) were obtained in the temperature range between 100 and 200 °C. The thermogravimetric analysis (TGA, TA Q500) was conducted at a heating rate of 20 °C/min from 60 to 800 °C under nitrogen. The dynamic mechanical analysis (DMA, TA-D2980) was carried out from 30 to 300 °C at a frequency of 1 Hz and a heating rate of 3 °C/min. The specimen (length 5 cm and width 4.5 mm) for DMA was prepared by cutting the film along the coating direction. The dielectric constant of each film was measured by a precision impedance analyzer (Agilent 4294 A) at 1 MHz frequency



Scheme 1. Synthesis procedures of phosphinate-contained PI.

Download English Version:

https://daneshyari.com/en/article/7105238

Download Persian Version:

https://daneshyari.com/article/7105238

Daneshyari.com