



Effect of trace diphenyl phosphate on mechanical and thermal performance of polyimide composite films



Shaoju Jian^{a,b}, Chenhui Ding^a, Ting Yang^a, Chunwu Zhang^a, Haoqing Hou^{a,*}

^a College of Chemistry and Chemical Engineering, Jiangxi Normal University, Nanchang 330027, PR China

^b College of Ecology and Resources Engineering, Wuyi University, Wuyishan 354300, PR China

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ABSTRACT

How to achieve polyimide (PI) with better thermal and mechanical properties is a great challenge. In this study, flame-retardant diphenyl phosphate (DPhP), was incorporated in to heterocyclic diamine contained PI films to form phosphorus-polyimide composite films (P-PIs). The structures, element distribution, thermal and mechanical properties of P-PIs were characterized by FT-IR, SEM, EDS, DMA, TGA, and tensile test. The results indicated that the incorporation of phosphorus can significantly enhance the thermal and mechanical properties. The 5% weight loss temperature in air and the glass transition temperature (T_g) of P0.6-PI film were 577 and 405 °C, which were 50 and 16 °C larger than the neat PI film, respectively. In addition, the P0.6-PI film also showed improved tensile strength, modulus and toughness by 47%, 57% and 76%, respectively, as compared to the neat PI film.

1. Introduction

Polyimides (PIs) are well-known for their superior thermal and thermo-oxidative stability, outstanding mechanical properties, as well as perfect solvent resistance, adhesives and electrical insulation properties and have been widely used in the fields of aerospace industries, membrane separation, automobile, coatings, and electronics, and so on [1–6]. Nowadays, to meet the demand of next generation spacecraft, it is highly desirable to improve their thermal and mechanical properties.

Incorporating heterocyclic units to the PI molecular chains can effectively improve the mechanical and thermal performance of PI [7–11]. Another strategy is to incorporate nanofillers to prepare PI composites. This is one of the simplest and most effective ways to enhance the mechanical and thermal performance of PIs. Carbon nanotubes [12–14], BaTiO₃ [15], short carbon nanofibers [16], graphene [17], nano-boria [18], and hybrid fillers [2,19], are mostly reported fillers to improve PI's mechanical and thermal properties.

In this study, a combination of incorporation of heterocyclic units to the PI molecular chains and organic fillers to the PI matrix was used to synergistically enhance the PI's mechanical and thermal properties. Herein, heterocyclic diamine, 2-(4-aminophenyl)-5-aminobenzimidazole (BIA), was chosen as monomer to enhance the rigidity of the PI molecular chains, and therefore enhance the mechanical properties and thermal performance of PI. On the other hand, trace flame-retardant diphenyl phosphate (DPhP) was incorporated to improve the thermal

performance. The good solubility of DPhP in precursor solution of PI can avoid the agglomeration of fillers in polymer matrix, and therefore significantly improve the thermal performance of PI.

2. Experimental

Experimental details including preparation of P-PIs and characterizations can be found in [Supporting information](#). In the following, the samples with initial loading percentage of DPhP of 0.1, 0.3, 0.4, 0.5, 0.6, 0.7 and 0.9 wt% were denoted as Px-PAA and Px-PI (x = 0.1, 0.3, 0.4, 0.5, 0.6, 0.7 and 0.9).

3. Results and discussion

3.1. Morphologies and structures of P-PIs

The PI, P0.3-PI, P0.9-PI films showed uniform and shiny surfaces, and the color of the films with similar thickness (cross-section SEM images shown in [Fig. S1](#)) became lighter with the increase of phosphorous ([Fig. 1a–c](#)). The P0.9-PI film exhibited smooth surface without any crack ([Fig. 1d](#)). Further exploration by EDS confirmed the presence of P element and the element mapping of C, N, O and P proved that the P element was uniformly dispersed in P0.9-PI film ([Fig. 1e–i](#)), which can be attributed to the good solubility of DPhP in PAA solutions. Further investigation by FT-IR also proved that the phosphorus has

* Corresponding author.

E-mail address: haoqing@jxnu.edu.cn (H. Hou).

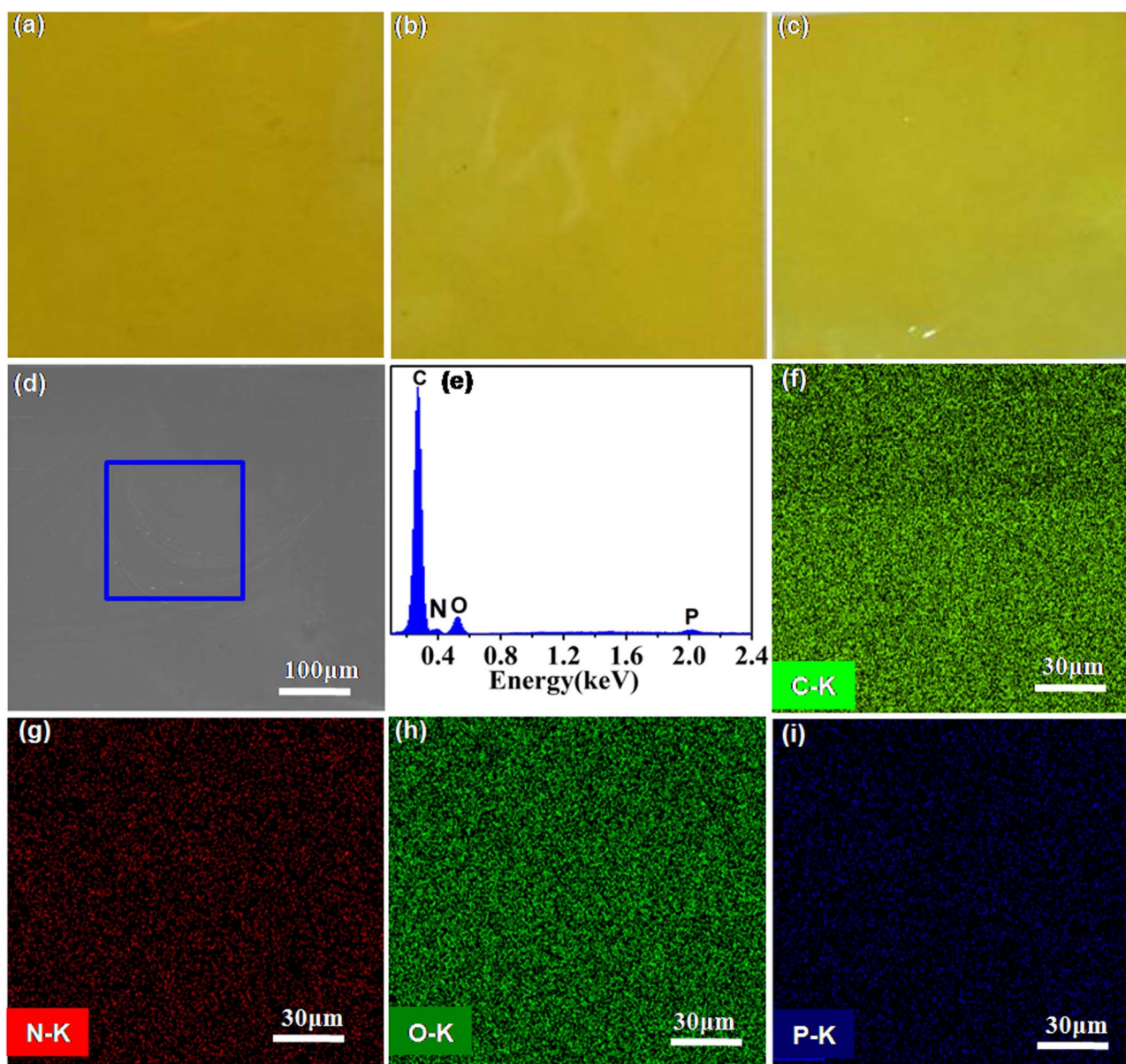


Fig. 1. Photographs of PI (a), P0.3-PI (b), P0.9-PI (c) and element mappings of P0.9-PI (d–i).

been successfully incorporated into the polyimide (Fig. S2).

3.2. Thermal properties

To evaluate the thermal properties of P-PIs, thermogravimetric analysis (TGA) and the dynamic mechanical analysis (DMA) were employed (Fig. 2). During the thermal imidization process, diphenyl phosphate may hydrolyze with water produced by the imidization under high temperature, which could promote the cyclization reaction. On the other hand, thermal imidization process of polyamide acid accompanied dissociation of polyamic acid, diphenyl phosphate and diamines may react to form phosphoramidate, and then amino end group of phosphoramidate could react with anhydride end group of BPDA or other production with anhydride end group, the recombination of terminal may embed phosphorus on the polyimide chain. Finally, organophosphorus compounds could decompose into polymetaphosphate at high temperature. DPhP was completely pyrolysed to polymetaphosphate with 31.7 wt% weight loss at 370 °C under Ar atmosphere (Fig. 2a), which was well agreed with the theoretical value (32.1 wt%). Compared with PI, the addition of phosphorus led to the of 5% ($T_{5\%}$)

and 10% ($T_{10\%}$) weight loss temperatures of P0.6-PI increased from 552 to 587 °C and from 579 to 611 °C, respectively. When the content of phosphorus was further increased, the $T_{5\%}$ and $T_{10\%}$ of P-PI (e.g., P0.7-PI and P0.9-PI) still kept constant as those of P0.6-PI. Fig. 2b showed the TGA curves under air atmosphere. The PI exhibited thermal stability with $T_{5\%}$ and $T_{10\%}$ at 527 and 557 °C, respectively. A small amount of phosphorus could significantly improve the thermal stability of P-PIs. For example, P0.1-PI possessed $T_{5\%}$ and $T_{10\%}$ of 561 and 581 °C, respectively, while P0.6-PI possessed the highest values of 577 and 593 °C, which were 50 and 36 °C higher than those of PI film. Further increasing the amount of phosphorus, the values of $T_{5\%}$ and $T_{10\%}$ did not display obviously difference.

As shown in Table 1, it can be seen that the residues of P-PIs both in argon and in air were higher than that of pure PI, suggesting that polymetaphosphate may act in the condensed phase to improve the thermal properties of PI [20–22]. The morphologies of residual char surface were shown in Fig. 2c and d. It can be seen that there were many pores on the surface of P0.1-PI, while a continuous and dense structure with fewer pores and cracks was observed on the surface of P0.9-PI. The above results indicated that polymetaphosphate can

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