Preparation of poly (arylene ether nitrile)/NzdFeB composite film with excellent thermal properties and tensile strength

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Abstract. PEN/NdFeB composite films were prepared by the solution casting method. The thermal properties, fracture morphology and tensile strength of the composite films were tested by DSC, TGA, SEM and electromechanical universal testing machine, respectively. The results reveal that the composite film has good thermal properties and tensile strength. Glass-transition temperature and decomposition temperatures at weight loss of 5% of the composite films retain at 166±1 C and 462±4 C, respectively. The composite film with 5 wt.% NdFeB has the best tensile strength value for 100.5 MPa. In addition, it was found that the NdFeB filler was well dispersed in PEN matrix by SEM analysis.

1. Introduction

Considering that polymer-based composite consists of polymer and inorganic filler has both the advantages of polymer and inorganic filler. A lot of research has been carried out [1, 2]. Meanwhile, due to the excellent comprehensive performance, polymer-based composite can be widely used in many fields. Poly (arylene ether nitrile) (PEN) is a kind of linear aromatic compounds with side chain containing cyano-group. As a new type of high performance specialty engineering plastics, PEN has excellent toughness, heat resistance, chemical stability, electrical insulation and mechanical properties [3, 4]. It has potential applications in many areas such as aerospace, electronic devices and automotive industries. It is the focus of the study on the blending modification of PEN [5, 6]. The purpose of modification is to further functionalize, enhance properties of composite and expand its applied range. Due to excellent characteristics such as high coercivity, remanence, maximum energy product [7, 8], NdFeB, as the third generation rare earth permanent magnet material, has been widely used in practical applications. In this work, PEN/NdFeB composite films were prepared by the solution casting method. The thermal properties and tensile strength of the composite films were studied and discussed.

2. Experimental section

2.1. Preparation of PEN/NdFeB composite films

PEN/NdFeB composite films were prepared by the solution casting method. PEN was added into NMP solvent and then the obtained mixture was heated to form a transparent PEN solution in a three-

necked bottle equipped with a mechanical stirrer. With that different content of NdFeB (2.5, 5.0, 7.5, 10.0 and 15.0 wt.%) were added into the PEN solution and then stirred for 1 h under the condition of ultrasonic water bath and a nitrogen atmosphere. After that the PEN/NdFeB solution was poured onto a clean horizontal glass plate and dried in an oven at 80, 120, 160 and 200 °C for 2 h, respectively. Finally, the as-prepared films were cooled to room temperature. For comparison purposes, the pure PEN film was also prepared by similar method without adding NdFeB.

2.2. Characteristic

Differential scanning calorimetric (DSC) analysis of all samples was performed using a modulated DSC-Q100 equipment (TA Instruments) at a heating rate of 10 °C min^{-1}. Thermogravimetric (TGA) and derivative thermogravimetric analysis (DTG) were carried out on a TGA-Q50 (TA Instruments) at a heating rate of 20 °C min^{-1}. The fractured surface morphology of all samples were observed by a scanning electron microscopic (SEM, JSM6490LV, Japan). The tensile strength of all samples were measured by a SANS CMT6104 series desktop electromechanical universal testing machine at room temperature and the test values were calculated as average values from five samples of each film.

3. Results and Discussion

In this work, NdFeB fillers with different particle sizes were prepared by ball milling. Fig. 1(a, b, c) show the SEM images of three NdFeB fillers of different particle sizes and three NdFeB fillers are labeled as S1, S2, and S3, respectively. From the SEM images, it can be found that the appearance of NdFeB filler appears as irregular block structure. The particle sizes of S1, S2, and S3 are about 3 μm, 10μm and 20μm. The fracture morphology of the PEN/NdFeB-S2 film and the PEN/NdFeB-S1 film are showed in Fig. 1(d, e, f). Compared with the PEN/NdFeB-S1 film, from the Fig. 1(d, f), there are large cracks in the fracture section of the PEN/NdFeB-S2 film, which is caused by large particle sizes. In addition, the PEN matrix is separated by the solid-phase network formed by NdFeB and this separation is more pronounced with the NdFeB content increases as shown in Fig. 1(d, e).

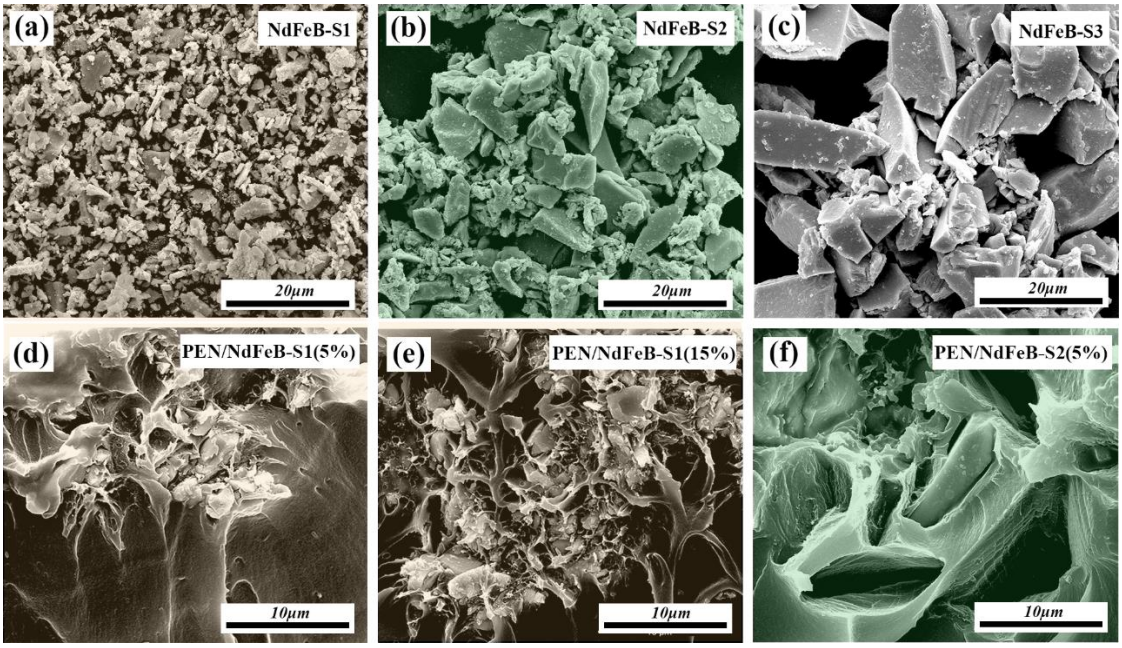

Figure 1. The SEM images of the NdFeB with different sizes (a, b, c). The fracture morphology of the PEN/NdFeB composite films (d, e, f).

The tensile strength of the composite films is showed in Fig. 2. From Fig. 2(a), it can be found that the tensile strength of the PEN/NdFeB (5 wt.%) films decreases as the NdFeB particle sizes increases.

While the tensile strength of the PEN/NdFeB-S1 films increases first and then decreases as shown in Fig. 2(b). When the NdFeB content is 5 wt.%, the tensile strength of the PEN/NdFeB-S1 film reaches up to maximum value for 100.5 MPa, which increased by 10% in comparison that of the pure PEN film (90.2 MPa). When the NdFeB content increased to 15%, the tensile strength of the PEN/NdFeB-S1film can still reach 80.0 MPa, revealing that the PEN/NdFeB-S1 film has good tensile strength.

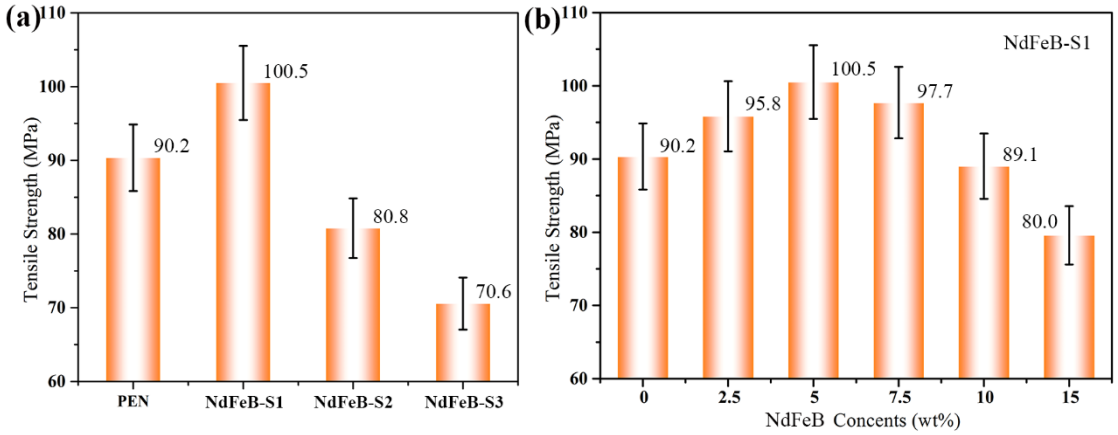

Figure 2. Tensile strength of the PEN/NdFeB composite films with different NdFeB particle sizes (a) and contents (b).

The changes of tensile strength of the composite films can be explained as follows: As a non-crosslinking point in PEN matrix, NdFeB filler with block structure has a lubricating effect on the PEN matrix, which resulted in an enhanced effect on the tensile strength of the composite film. In addition, When the composite film is subjected to external forces, filler plays the role of stress concentration and transfer energy, so that the PEN matrix has no obvious stress concentration, and the crack propagation is blocked. However, excessive filler content or larger filler particle sizes are disadvantage for improving the tensile strength. A large number of micro-cracks are easy to develop into macroscopic cracking, leading to the tensile strength decrease. The changes can be seen from the Fig. 1 (d, e, f).

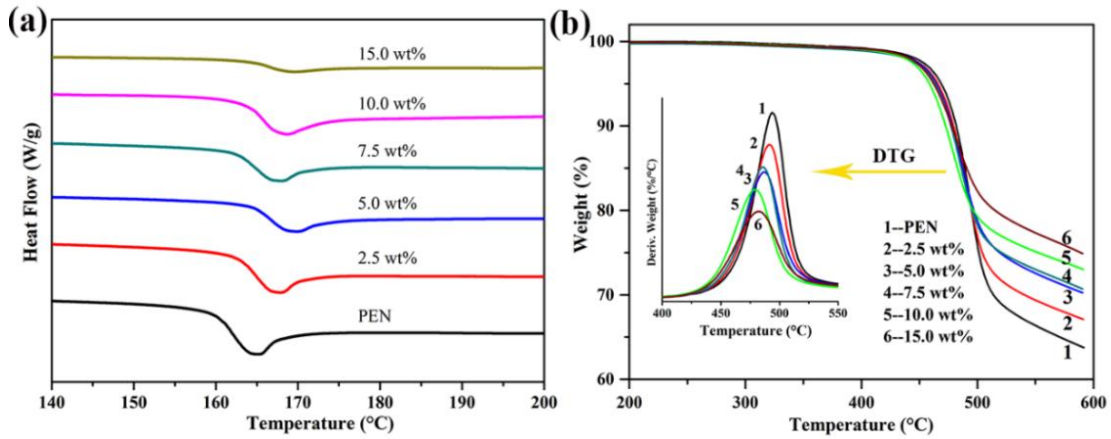

Figure 3. The DSC (a) and TGA (b) curves of the PEN/NdFeB-S1 composite films with different NdFeB contents.

Fig. 3 shows the thermal properties of the composite films. The DSC curves of the composite film are presented in Fig. 3(a). The relative glass transition temperatures (T_g) values are listed in Table 1. It

can be seen that the composite films remain stable T_g (166 ±1°C), suggesting that filler does not have significant effect on the T_g of the PEN matrix. In addition, the TGA and DTG curves of the composite films are showed in Fig. 3(b). The detailed decomposition temperatures at weight loss of 5% ($T_{5\%}$), 10% ($T_{10\%}$) and maximum decomposition temperature (T_{max}) are summarized in Table 1. It can be found that after adding NdFeB into PEN matrix, the thermal stability of the composite films has a slight decrease. While the $T_{5\%}$, $T_{10\%}$ and T_{max} of the composite films still remain at 462 ±4°C, 476 ±4°C and 485 ±6°C, respectively, revealing that the composite films show excellent thermal properties.

Table 1. The thermal properties of the PEN/NdFeB-S1 composite films.

Sample	PEN	2.5%	5.0%	7.5%	10.0%	15.0%
T_g (°C)	162.4	165.1	167.2	165.2	166.0	166.5
$T_{5\%}$ (°C)	468.9	465.9	464.4	463.4	458.7	462.3
$T_{10\%}$ (°C)	481.7	479.4	478.1	477.2	472.6	477.8
T_{max} (°C)	493.8	491.1	487.3	485.4	478.9	482.9

4. Conclusion

PEN/NdFeB composite films were prepared successfully by the solution casting method. The results show that the composite films have good tensile strength and excellent thermal properties. In addition, filler content and filler particle sizes are two significant elements for enhancing the tensile strength; the introduction of filler does not have significant effect on the T_g of the PEN matrix, while the thermal stability of the composite films has a slight decrease.

Acknowledgments

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