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Preparation and properties of montmorillonite/ organo-soluble polyimide hybrid materials prepared by a one-step approach

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Montmorillonite (MMT)/organo-soluble polyimide (PI) hybrids were prepared using a one-step approach. The organo-modified MMT was dispersed in a solution of diphenylether-3, 3′4,4′-tetracarboxylic dianhydride and 4,4′-diamino-3,3′-dimethyldiphenylmethane. The solution polycondensation followed by a direct solution imidization at 180 °C resulted in MMT/PI hybrid solutions. From wide angle X-ray diffraction (WAXD) and transmission electron microscopy (TEM) results, the MMT is basically exfoliated in the hybrid films cast from the solutions when the MMT content is below 5 wt%. Further increase in the MMT content leads to severe aggregation. The properties of a MMT/PI hybrid are significantly dependent upon the MMT content. When the MMT content is below 6 wt%, the introduction of the MMT leads to strengthening and toughening to the PI matrix at the same time. The introduction of the MMT also results in improved thermal stability, marked decrease in coefficient of thermal expansion, slight increase in glass transition temperature and increase in modulus.

1. Introduction

Polymer-clay hybrid materials have received considerable attention because of the interesting combination of the properties of both clay and polymer due to the strong interactions between them [1, 2] Clays have been extensively used in the polymer industry as reinforcement agents to improve the physico-mechanical properties of the final polymers. The efficiency of the clay to modify the properties of the polymer is primarily determined by the degree of its dispersion in the polymer matrix, which, in turn, depends on the size of clay particles. However, the hydrophilic nature of the clay surfaces impedes its homogeneous dispersion in the organic polymer phase.

Montmorillonite (MMT) is one of the most widely used clays. It is composed of silicate sheets of 1 nm thickness with adsorbed exchangeable inorganic cations [3, 4]. The organic compounds and polymers could be intercalated into the MMT layers due to the easy exchange of these inorganic cations with organic cations. This intercalation increases the spacing between the MMT sheets and even leads to the complete dissociation of the sheets to form a MMT/organic composite with a nanometer scale [1, 2]. The MMT/polymer hybrids possess some interesting features: The introduction of a very small fraction of MMT would lead to dramatic property improvement [5]; The hybrids possess excellent "barrier" property because of the two dimensional structure of MMT introduced [1, 2]. The incorporation of MMT into various types of polymers, such as poly(methyl methacrylate) [6], polystyrene [7–9], polypropylene [10–12], nylon 6 [13–15], poly(ethylene oxide) [16], polyurethane [17], poly(ethylene glycol) [18], silicone rubber [19], epoxy [20, 21], poly(ε-caprolactone) [22–24] and polyimide [25–29] has been reported.

Aromatic polyimides (PI) exhibit outstanding dielectric and mechanical properties at elevated temperature [30, 31]. However, most of the aromatic polyimides are neither soluble nor fusible. They are usually prepared by a two-step method. A solution polycondensation between diamine and dianhydride monomers forms a polyamic acid (PAA) solution. This PAA solution is then used to cast a film. The PAA film is further converted to a PI via a thermal imidization reaction at high temperature. This two-step approach has been employed to prepare clay/PI hybrids [25–29]. In our previous studies [27, 28], an organo-soluble PI is used as the matrix to prepare MMT/PI hybrids by using this two-step approach. However, the employment of the two-step approach in the preparation of clay/PI hybrids has several drawbacks. Because of the instability of a PAA solution (which undergoes partial imidization and decomposition) at room temperature, the clay/PAA solution would have a short shelf life. Furthermore, a large amount of water is released during the thermal

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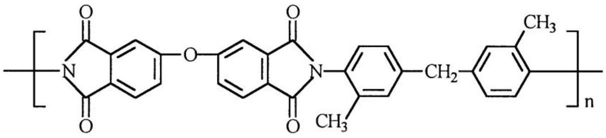
imidization. The water would be very difficult to remove from the hybrid film and its presence reduces the intactness and the performance of the film [32].

The one-step approach has been widely used to prepare organo-soluble PI solutions [33]. A PI solution is formed by a solution polycondensation between diamine and dianhydride monomers at room temperature followed by a direct imidization at an elevated temperature (such as 180 °C) as the water generated is removed from the system simultaneously. Only the removal of the solvent is required to obtain a PI film from this PI solution.

In this paper, we report the preparation of the MMT/organo-soluble PI hybrids using the one-step approach. The chemical structure of the polyimide is shown in scheme 1. Apart from the advantages mentioned, the high temperature used in the solution imidization process may also be beneficial to the dispersion of MMT. The study on the characterization and the properties of the hybrids are also presented.



Scheme 1 Chemical structure of polyimide.

2. Experimental

2.1. Materials

A sodium montmorillonite (Na-MMT) was supplied by the Institute of Chemical Metallurgy, Chinese Academy of Sciences. The particle size is 40 μm. 4,4′-Diamino-3,3′-dimethyldiphenylmethane (MMDA) was synthesized by a reaction of *o*-methyl aniline with formaldehyde [34]. Diphenylether-3,3′,4,4′-tetracarboxylic dianhydride (OPDA, industrial product, purchased from Shanghai Research Institute of Synthetic Resins) was dried at 190 °C for 2 hours before use. 1-Dodecylamine ($C_{12}H_{25}NH_2$, analytical reagent grade, purchased from Beijing Chemicals Company), 1-hexadecylamine ($C_{16}H_{33}NH_2$, lab reagent, purchased from Merck), celtyltrimethyl ammonium bromide ($C_{16}H_{33}(CH_3)_3N^+Br^-$, analytical reagent grade, purchased from Shanghai Reagent Company) and *iso*-quinoline (analytical reagent grade, purchased from Beijing Chemicals Company) were used as received. *m*-Cresol (analytical reagent grade, purchased from Shanghai Reagent Company) was dried over molecular sieves before use. Common solvents for solubility measurement such as *N*-methyl-2-pyrrolidone (NMP), *N*,*N*-dimethylacetamide (DMA), *N*,*N*-dimethylformamide (DMF), *N*,*N*-dimethylsulfoxide (DMSO), chloroform and tetrahydrofuran (THF) were used without further purification.

2.2. Preparation of organophilic-MMT

(1). MMT organo-modified with 1-dodecylamine ($C_{12}H_{25}NH_2$) and 1-hexadecylamine ($C_{16}H_{33}NH_2$): The organo-modification of MMT with 1-dodecylamine ($C_{12}H_{25}NH_2$) and 1-hexadecylamine ($C_{16}H_{33}NH_2$) has been described in our previous study [27, 28].

(2). MMT organo-modified with celtyltrimethyl ammonium bromide ($C_{16}H_{33}(CH_3)_3N^+Br^-$): A dispersion of 30 g Na-MMT in 6×10^{-4} m^3 distilled water was added to a heated (80 °C) solution of 26.02 g celtyltrimethyl ammonium bromide in 10^{-4} m^3 distilled water. The mixture was agitated vigorously for one hour. Upon filtration, white precipitate was obtained. It was then repeatedly washed with hot water until no Br^- was detected in the filtrate by silver nitrate. The organo-treated MMT was finally dried at 40 °C in vacuum.

2.3. Preparation of MMT/PI hybrids

The organophilic MMT was thoroughly dispersed to 25×10^{-6} m^3 *m*-cresol with the help of agitation and ultrasonic wave at 100 °C for 30 minutes before being cooled to room temperature. To this dispersion, 0.005 mol MMDA, 0.005 mol OPDA and 2 drops of *iso*-quinoline were dissolved, consecutively. The mixture was stirred at room temperature for 2 hours and then at 180 °C for 3 hours. Upon cooling, the obtained brownish solution was cast on a glass substrate and then heated subsequently at 70 °C for 12 hours, 120 °C for 4 hours, 200 °C for 4 hours and 270 °C for 2 hours to obtain the MMT/PI hybrid film.

2.4. Characterization and property measurements of MMT/PI hybrids

The wide angle X-ray diffraction (WAXD) patterns were recorded on a Rigaku Geiger Flex D/max-RB diffractometer using Cu K_α radiation (50 kV, 100 mA). The experiments were performed in a range of $2\theta = 1.5$–40° with a scan rate of 2°/min. The samples were 10 mm × 10 mm × 50 μm films.

The transmission eletron microscopic (TEM) analysis of the ultrathin-section of the MMT/PI hybrid films was conducted on a Philip CM120 TEM using an acceleration voltage of 80 kV. The ultrathin-section was microtomed from the epoxy block which embedded the hybrid films by using a LKB2088 ultratome.

The FT-IR spectra were recorded on a Perkin Elmer Paragon 1000 Infrared Spectrophotometer.

The intrinsic viscosity of the PI and the MMT/PI hybrids was measured with an Ubbelodhe Viscometer using NMP as the solvent at 30 °C. The standard concentration was 10 kg/m^3.

The glass transition temperatures (T_gs) were measured by differential scanning calorimetry (DSC). The DSC analysis was conducted on a Perkin Elmer Pyris I Differential Scanning Calorimeter under the protection of N_2. The heating rate was 20 °C/min.

The thermal gravimetric analysis (TGA) was conducted on a Perkin-Elmer TGA 7 Thermal Analyzer under N_2 flow. The heating rate was 20 °C/min.

The dynamic thermal analysis of the PI and the MMT/PI hybrid films was recorded on a Perkin-Elmer DMA7e Dynamic Mechanical Analyzer (DMA). The heating rate is 5 °C/min. The coefficient of thermal expansion (CTE) of the PI and the MMT/PI hybrid films was also measured on a Perkin-Elmer DMA7e

Dynamic Mechanical Analyzer with a TMA mode. The load on the film was 10 mN and the heating rate is 5 °C/min. A temperature range of 75–125 °C was selected to calculate the CTE.

The stress-strain curves of the PI and the MMT/PI hybrid films were recorded on an Instron-4465 Universal Tester at the room temperature at a drawing rate of 5 mm/min.

The solubility of the PI and the MMT/PI hybrids in various organic solvents at room temperature was also measured.

TABLE I d-Value of MMT modified with various intercalation agents

Sample	2θ (°)	d (nm)	Peak intensity
Na-MMT	6.78	1.30	S
$C_{16}H_{33}(CH_3)_3N^+$-MMT	2.20	4.01	VS
	4.42	—	S
	6.68	—	W
$C_{16}H_{33}NH_2$-MMT	2.62	3.37	VS
	5.16	—	S
	7.64	—	W
$C_{12}H_{25}NH_2$-MMT	5.04	1.75	S

(S: strong; W: weak; VS: very strong).

3. Results and discussion

3.1. Organo-modification of MMT

Because of a hydrophilic nature of the MMT, the organo-modification of the MMT is an important step in the preparation of MMT/polymer hybrids. The organo-modification could change the hydrophilicity of Na-MMT to be more hydrophobic to generate a micro chemical environment appropriate to the intercalation of a polymer or a monomer. Furthermore, the increase in the layer-to-layer spacing in the MMT is also desired. Primary aliphatic amines such as 1-dodecylamine and 1-hexadecylamine and quaternary ammonium salts have been the most effective modification reagents in the preparation of MMT/polyamide and MMT/PI hybrids [13–15, 26–29]. Firstly, amines and quaternary ammonium salts can undergo cation exchange to change the hydrophilic nature of the MMT, in cation exchange reaction the amines or quaternary ammonium salt replace Na^+ in the MMT galleries. Secondly, the structural similarity of the organo-modification reagents to the diamine monomers would lead to the easy intercalation of these monomers to the MMT. In addition, the long aliphatic groups can increase the MMT layer-to-layer spacing effectively. Fig. 1 is the WAXD patterns of the Na-MMT and the organo-modified MMT. Table I lists the basal spacings of the MMT calculated from the Bragg Equation. The basal spacing of the MMT is obviously increased by the organo-modification. It is increased from 1.30 nm for an untreated MMT to 1.75 nm for a MMT treated with 1-dodecylamine ($C_{12}H_{25}NH_2$-MMT), to 3.37 nm for a MMT treated with 1-hexadecylamine ($C_{16}H_{33}NH_2$-MMT) and further to 4.01 nm to a MMT treated with celtyltrimethyl ammonium bromide ($C_{16}H_{33}(CH_3)_3N^+$-MMT). The increased size of the alkyl group leads to a greater MMT layer-to-layer spacing. These results are similar to those obtained in our previous study [27, 28].

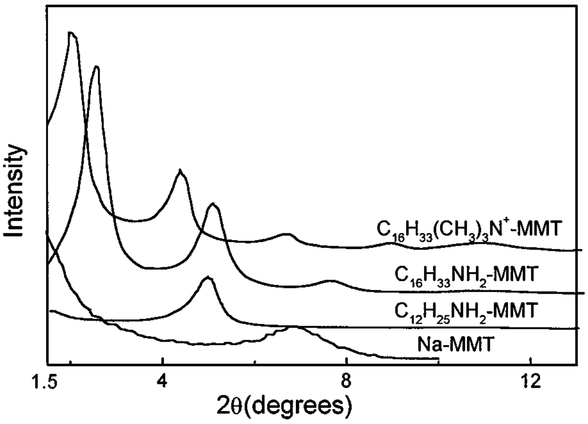


Figure 1 WAXD patterns of Na-MMT, $C_{12}H_{25}NH_2$-MMT, $C_{16}H_{33}NH_2$-MMT and $C_{16}H_{33}(CH_3)_3N^+$-MMT.

3.2. The preparation of MMT/PI hybrids by a one-step approach

The MMT/PI hybrids, in which the MMT was organo-modified with $C_{16}H_{33}(CH_3)_3N^+Br^-$, were named $C_{16}H_{33}(CH_3)_3N^+$-MMT/PI. Similarly, the two other series of MMT/PI hybrids are $C_{12}H_{25}NH_2$-MMT/PI and $C_{16}H_{33}NH_2$-MMT/PI.

Unlike the commonly used two-step method, in which a MMT/PAA hybrid is first prepared and then converted to a MMT/PI hybrid with the thermal imidization, a one-step approach is used in this study to prepare MMT/PI hybrid materials. They are obtained directly from the organo-treated MMT, diamine and dianhydride monomers by a high temperature solution intercalation/polycondensation/imidization process. The preparation is confirmed by infrared spectroscopy (Fig. 2). The absorption bands at 1038 cm^{-1} and 1090 cm^{-1} are characteristic of the Na-MMT. After the treatment with $C_{16}H_{33}(CH_3)_3N^+Br^-$, the sample exhibits the characteristic bands of C-H stretching at 2920 cm^{-1} and 2851 cm^{-1}. The characteristic absorption bands of aromatic polyimides at ~1780 cm^{-1} (C=O unsymmetrical stretching in imide groups), ~1720 cm^{-1} (C=O symmetrical stretching in imide groups) and ~1380 cm^{-1} (C-N stretching in imide groups) are observed in both the PI and the MMT/PI hybrids. Meanwhile, the characteristic absorption bands of amide groups at ~1660 cm^{-1} (C=O stretching) and ~1550 cm^{-1} (C-N stretching) are absent. The characteristic absorption band of the MMT was also observed in the MMT/PI hybrids at 1038 cm^{-1}.

$C_{12}H_{25}NH_2$-MMT/PI, $C_{16}H_{33}NH_2$-MMT/PI hybrid materials have been produced in our research work, however, they are brittle and have poor mechanical properties. this may be caused by the fact that $C_{12}H_{25}NH_2$, $C_{16}H_{33}NH_2$, $C_{12}H_{25}NH_2$-MMT and $C_{16}H_{33}NH_2$-MMT are monoamines and they may compete with the MMDA to react with OPDA and act as blockers to PI. Thus the molecule weights of PI in $C_{12}H_{25}NH_2$-MMT/PI and $C_{16}H_{33}NH_2$-MMT/PI hybrid materials are much lower than that of $C_{16}H_{33}(CH_3)_3N^+$-MMT/PI hybrid materials.

TABLE II Properties of PI and $C_{16}H_{33}(CH_3)_3N^+$-MMT/PI hybrids

MMT content (wt%)	0	2.0	3.2	5.0	10.0	20.0
$[\eta]^a$	1.09	1.01	1.07	1.03		
T_g (°)^b	286.0	286.9	289.0	290.2		
T_d (°)^c	518.0	522.0	523.6	525.0	542.0	542.0

a: Intrinsic viscosity measured at 30°C in NMP with a standard concentration of 10 kg/m^3.
b: Obtained from DSC measurements, scan rate: 20 °C/min. N_2 protection.
c: Thermal decomposition temperature (on-set) from TGA measurement, scan rate: 20 °C/min. N_2 protection.

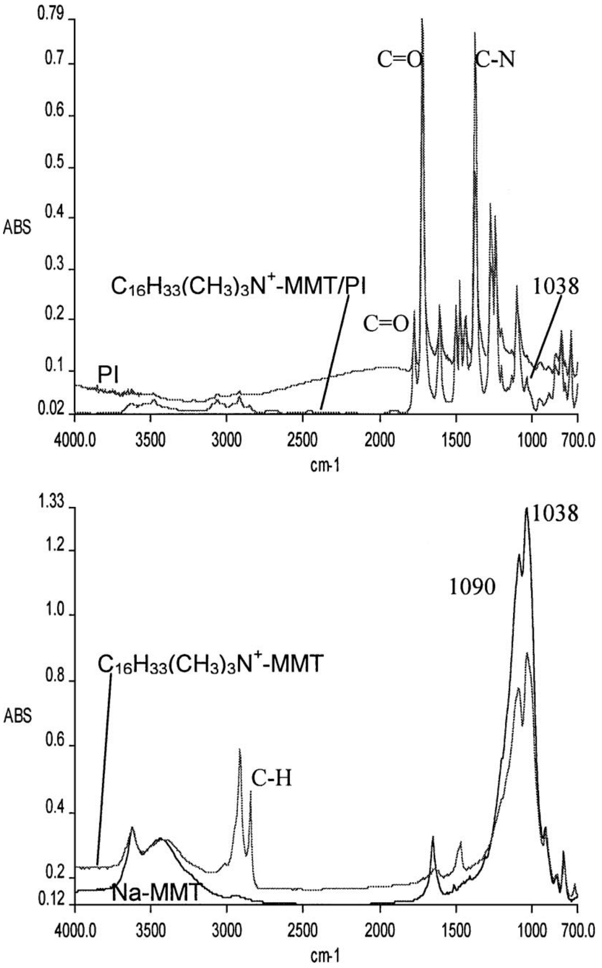


Figure 2 IR spectra of Na-MMT, $C_{16}H_{33}(CH_3)_3N^+$-MMT, PI and $C_{16}H_{33}(CH_3)_3N^+$-MMT/PI (6 wt%). PI film was cured at 70 °C for 12 h, PI/$C_{16}H_{33}(CH_3)_3N^+$-MMT hybrid film was was kept at 70 °C for 12 hours, 120 °C for 4 hours, 200 °C for 4 hours and 270 °C for 2 hours.

To reduce the influence of the molecular weight of the PI upon the properties of the PI and the MMT/PI hybrids, we prepared the PI and the MMT/PI hybrids with high and similar molecular weight. The intrinsic viscosity of some of the samples is listed in Table II.

3.3. Dispersion of MMT in PI matrix

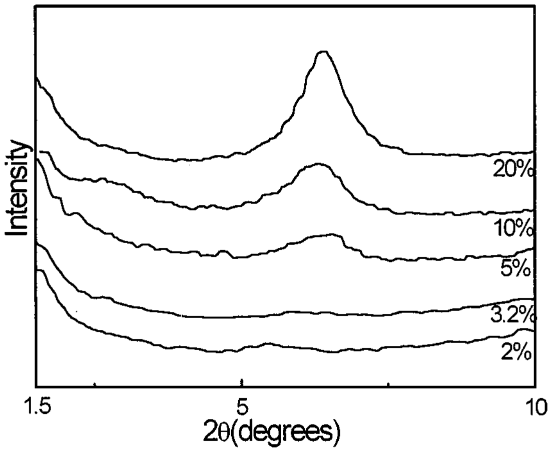


Figure 3 WAXD patterns of $C_{16}H_{33}(CH_3)_3N^+$-MMT/PI hybrid films with various MMT content.

Fig. 3 is the WAXD patterns of $C_{16}H_{33}(CH_3)_3N^+$-MMT/PI hybrid films with various MMT content. The 001 diffraction peak of the MMT in the hybrids is rarely observed when the MMT content is below 5 wt%. A small amount of MMT can be well dispersed in the PI solution with the help of ultrasonic wave and vigorous agitation at high temperature. When the MMT content is low ($\leq$6 wt%), the hybrid solution is clear, also indicating that the MMT is well dispersed. No obvious aggregation is observed even when the solvent is removed. As the MMT content is increased to 10 wt%, the aggregation happens both in solutions and in films. The aggregation of the MMT in films is well indicated in WAXD patterns as the diffraction peak intensity of the MMT is rapidly increased as the MMT content is above 5 wt%. To confirm whether the MMT is exfoliated or not in the hybrids when the MMT content is low, TEM analysis was used to study the morphology of the ultrathin-section of the $C_{16}H_{33}(CH_3)_3N^+$-MMT/PI film containing 3.2 wt% MMT (Fig. 4). The black lines in the TEM photographs represent the MMT domain while the white part represents the PI matrix. Fig. 4 shows that the MMT does not exist in layer structure and is almost orderlessly dispersed in the hybrids. The above results clearly demonstrate the feasibility of the preparation of a MMT/organo-soluble PI hybrid with this one-step approach. The good dispersibility of the MMT in the hybrid can be easily achieved. The application of the method could solve the problems raised from the water generated in the processing procedure of a MMT/PI hybrid.

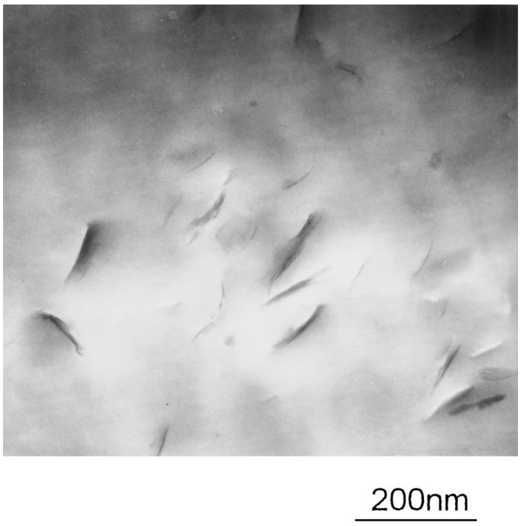


Figure 4 TEM micrographs of the ultrathin-section of $C_{16}H_{33}(CH_3)_3N^+$-MMT/PI hybrid film containing 3.2 wt% MMT.

3.4. Mechanical properties of MMT/PI hybrids

Fig. 5 is the relationship between the MMT content and the Young's modulus of the $C_{16}H_{33}(CH_3)_3N^+$-MMT/PI hybrid films. The introduction of the MMT, which has a higher modulus than the PI matrix, leads to an increase in the modulus of the hybrid film. As the MMT content is below 5 wt%, the Young's modulus is increased almost linearly to the increase of the MMT content. This linear increase may be caused by the fact that the amount of the MMT sheet is increased linearly to the MMT weight content if the MMT is exfoliated in a low MMT content. As the MMT content is further increased, the aggregation of the MMT leads to a leveling off or even a slight decrease in the modulus of the hybrid. This result is in good agreement with the WAXD results and what was observed in the preparation of the hybrids.

The influence of the MMT content upon the mechanical properties of the hybrid films is shown in Fig. 6. When the MMT content is less than 6 wt%, both the tensile strength and the elongation at break are increased with the MMT content. The tensile strength is increased from 93.1 MPa for PI to 135.0 MPa for the hybrid containing 6 wt% MMT (a 45% increase) while the elongation at break is increased from 4.6% to 7.4% (a 60% increase). The PI is strengthened and toughened simultaneously by the introduction of the well-dispersed (exfoliated) MMT. The strong interfacial interaction between the PI and the MMT and the existence of the large shear zone increases the tensile strength of the hybrid. Furthermore, the large amount of interface between the MMT sheets and the PI induces the formation of the shear zone while at the same time the layer structure of the MMT particle could also stop the development from the shear zone to cracks. This would lead to an increase in the toughness of the hybrids. When the MMT content exceeds 6 wt%, both the tensile strength and the elongation at break are decreased probably due to the aggregation of the MMT.

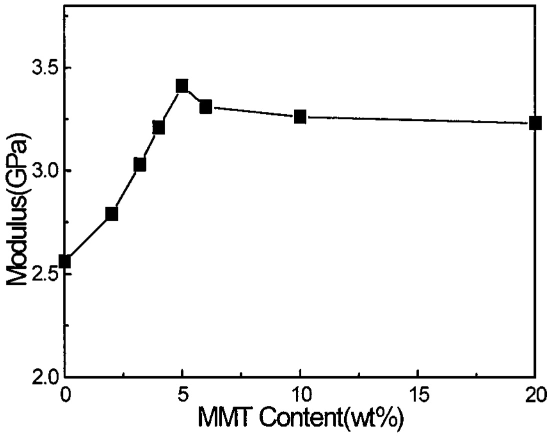


Figure 5 Relationship between the MMT content and the Young's modulus of $C_{16}H_{33}(CH_3)_3N^+$-MMT/PI hybrid films.

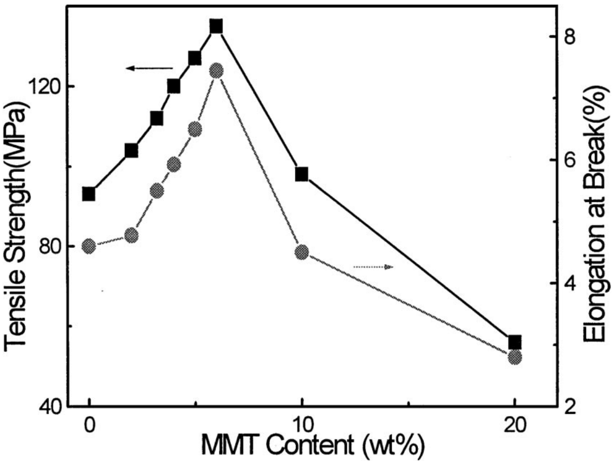


Figure 6 Relationship between the MMT content and the mechanical properties of $C_{16}H_{33}(CH_3)_3N^+$-MMT/PI hybrid films.

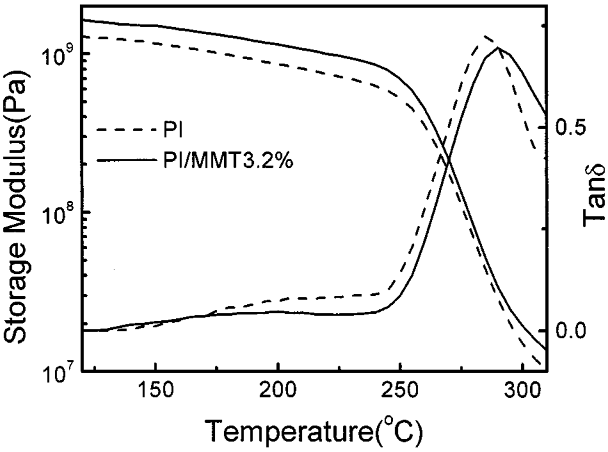


Figure 7 DMA curves of PI and $C_{16}H_{33}(CH_3)_3N^+$-MMT/PI hybrid films.

Fig. 7 shows the DMA curves of the PI film and the $C_{16}H_{33}(CH_3)_3N^+$-MMT/PI hybrid film containing 3.2 wt% MMT. The Tan δ peak of the $C_{16}H_{33}(CH_3)_3N^+$-MMT/PI hybrid film shifts to a slightly higher temperature and broadens in comparison to the PI. The hybrid exhibits a slightly higher T_g than the PI. The broadness of the Tan δ peak of the hybrid may be caused by the strong interaction between the MMT and the PI, which limits the relaxation of the PI molecular chains. The storage modulus of the PI below and above its T_g is increased when a small amount of MMT is introduced (1.49 GPa versus 1.15 GPa at 150 °C and 1.36 GPa versus 1.02 GPa at 310 °C).

3.5. Thermal properties of MMT/PI hybrids

Table II lists the thermal properties of the PI and the $C_{16}H_{33}(CH_3)_3N^+$-MMT/PI hybrids. The thermal stability of the hybrid is increased as the MMT content is increased. The initial thermal decomposition temperature (on-set temperature) accessed by TGA is increased from 518 °C for the PI to 525 °C for the hybrid containing 5 wt% MMT and further to 542 °C for the hybrid containing 10 wt% MMT. MMT possesses high thermal stability and its layer structure exhibits great barrier effect to stop the evaporation of the small molecules generated in the thermal decomposition and effectively limits the continuous decomposition of the PI.

The T_g of the hybrid accessed by DSC is also slightly increased as the MMT content is increased (see

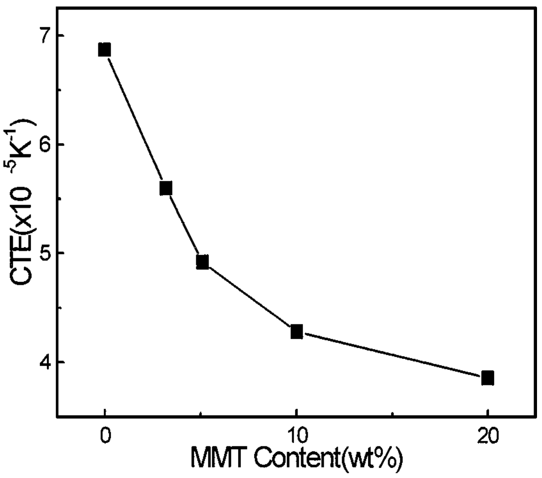


Figure 8 Relationship between the MMT content and the coefficient of thermal expansion of $C_{16}H_{33}(CH_3)_3N^+$-MMT/PI hybrid films.

Table II). This is clearly caused by the strong interaction between the MMT and the PI, which limits the segmental movement of the PI.

The reduced segmental movement of the PI would also lead to a decrease in the coefficient of thermal expansion (CTE). Fig. 8 is the relationship between the MMT content and the CTE of the $C_{16}H_{33}(CH_3)_3N^+$-MMT/PI hybrid films. It is observed that the introduction of a small amount of MMT effectively reduces the CTE. The CTE of the PI is decreased from 6.9×10^{-5} K^{-1} to 4.9×10^{-5} K^{-1} (a 29% decrease) when 5 wt% of MMT is introduced. As the MMT content is increased, the CTE decreases continuously.

3.6. Solubility of MMT/PI hybrids

Most of the aromatic polyimides based on 4,4′-diamino-3,3′-dimethyldiphenylmethane (MMDA) possess great organo-solubility due to the incorporation of the two substitute methyl groups, which leads to the increase in the free volume and the decrease of the molecular packing. The steric hindrance from the methyl groups may also lead to the distortion of the conjugation of the polyimide backbones [34].

The $C_{16}H_{33}(CH_3)_3N^+$-MMT/PI hybrid solutions prepared by the one-step method in this study were clear when the MMT content was below 5 wt%. When the hybrids were precipitated from methanol or ethanol and dried at a low temperature (i.e. 70 °C), they were still soluble in strong polar aprotic solvents such as NMP, DMA, DMF, DMSO and common low boiling point solvents such as chloroform (see Table III). This organo-soluble may be very important when the MMT/PI hybrids are applied in some special circumstances, such as to provide some photo-etching Patterns. As the hybrids were treated at a high temperature (i. e. 270 °C), the solubility was markedly reduced due to the increased chain packing and possible thermal crosslinking.

TABLE III Solubility of $C_{16}H_{33}(CH_3)_3N^+$-MMT/PI hybrids

Sample	NMP	DMA	DMF	DMSO	THF	Chloroform
PI (precipitated)	√	√	√	√	×	√
MMT/PI (precipitated)	√	√	√	√	×	√
PI film (dried at 70 °C)	√	√	√	×	×	√
MMT/PI film (dried at 70 °C)	√	√	√	×	×	√
PI film (dried at 270 °C)	×	×	×	×	×	×
MMT/PI film (dried at 270 °C)	×	×	×	×	×	×

√: soluble; ×: insoluble.

4. Conclusions

MMT/PI hybrids were successfully prepared using organo-MMT by a one-step approach. From WAXD and TEM results, the MMT silicate layers is basically exfoliated in the hybrid films cast from the solutions when the MMT content is below 5 wt%. Further increase in the MMT content leads to the severe aggregation. The MMT content influences the properties of a MMT/PI hybrid significantly. When the MMT content is below 6 wt%, the MMT/PI hybrids will be strengthened and toughened at the same time. However, when the MMT content is above 6 wt%, the mechanic properties of the MMT/PI hybrids will became poor. The introduction of MMT also results in improved thermal stability, marked decrease in coefficient of thermal expansion, slight increase in glass transition temperature and increase in modulus.

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