

# Preparation and Properties of T300 Carbon Fiber-Reinforced Thermoplastic Polyimide Composites

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**ABSTRACT:** In this study, a series of T300 carbon fiber-reinforced polyimide (CFRPI) composites were prepared by laminating premolding polyimide (PI) films with unidirectional carbon fiber (CF) layers. On the basis of PI systems design, the effect of CF volume fraction, processing conditions, and PI molecular structure on the properties of CFRPI composites was studied in detail. In addition, two kinds of nano-particles, including carbon nano-tube (CNT) and SiO<sub>2</sub> were filled into the premolding PI films with different concentrations. And the effect of nano-particles on the properties of CFRPI composites was also investigated. The surface characteristic of T300 CF was measured by X-ray photoelectron spectroscopy (XPS) and Fourier transform infrared spectroscopy (FTIR). The properties of premolding PI film and CFRPI composites were measured by dynamic mechan-

ical analysis (DMTA), SANS testing machine, scanning electron microscopy (SEM), and so forth. These experimental results showed that the properties of CFRPI composites were mainly affected by the premolding PI film and molding condition. The change of CF volume fraction from 55% to 65% took little effect on the mechanical properties of CFRPI composites. In addition, the incorporation of nano-particle SiO<sub>2</sub> could further improve the properties of CFRPI composites, but CNT hardly improved the properties of CFRPI composites. © 2006 Wiley Periodicals, Inc. *J Appl Polym Sci* 102: 646–654, 2006

**Key words:** polyimides; composites; carbon fiber-reinforced composites; nano-particles; properties

## INTRODUCTION

Polyimides (PIs) are a type of high-performance materials.<sup>1</sup> Controlled molecular weight, low melt viscosity PIs are easily processed via thermoplastic forming techniques which makes them attractive matrix materials for advanced composites applications. Because the applications of advanced resin-based composites are increasing, more and more attention of people is drawn on carbon fiber-reinforced polyimide (CFRPI) composites. To improve the toughness of advanced composites, the thermoplastic resin-based composites need to be explored further.<sup>2–4</sup>

The advanced thermoplastic composites can be prepared by directly using prepregs or combining prepregs with resin films. The latter is a flexible method to prepare the advanced composites. The properties of advanced composites are mainly decided by resin, fiber, and processing conditions. As far as the same kind of fiber is concerned, the properties of resin is based on the processing conditions and the properties of CFRPI composites. On the basis of PI molecular structure design, we synthesized eight kinds of PI

system to prepare CFRPI composites. In addition, some papers reported that the properties of PI, such as mechanical properties,  $T_g$ , and other properties, were further improved by the incorporation of nano-particles.<sup>5–9</sup> Here, the effect of nano-particles on properties of CFRPI composites was also investigated. By incorporating the SiO<sub>2</sub> and CNT nano-particles into the premolding PI films, the properties of PI film should be improved to some extent. Therefore, the premolding PI films filled with nano-particles were used to prepare CFRPI composites, and we expected the properties of CFRPI composites to be further improved.

In this study, a series of T300 CFRPI composites were prepared by a typical method of advanced thermoplastic composites, that is, CF prepregs and premolding PI resin films were combined together.<sup>10</sup> The premolding PI films were prepared in a far-infrared oven, and the accelerating effect of far-infrared radiation (FIR) on PI imidization had been reported in some papers.<sup>11–14</sup> The effect of CF volume fraction, processing conditions, and PI molecular structure on properties of CFRPI composites was investigated. In addition, two kinds of nano-particles, including SiO<sub>2</sub> and CNT, were introduced into the premolding PI films by *in situ* reaction and direct mixing. The effect of nano-particles on the properties of CFRPI composites was also discussed.

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TABLE I  
PI Systems Synthesized in This Experiment

System no.	Components	Molar ratio of co-monomers	MW (k) $\times 10^3$ g/mol
1	ODPA/3,4'-ODA/PA	–	25
2	BPDA/3,4'-ODA/PA	–	25
3	BTDA/3,4'-ODA/PA	–	30
4	BPADA/3,4'-ODA/PA	–	25
5	ODPA/BPDA/3,4'-ODA/PA	ODPA/BPDA = 90/10	25
6	BTDA/BPDA/3,4'-ODA/PA	BTDA/BPDA = 90/10	30
7	BPADA/BPDA/3,4'-ODA/PA	BPADA/BPDA = 90/10	25
8	BPDA/3,4'-ODA/4,4'-ODA/PA	3,4'-ODA/4,4'-ODA = 50/50	30

## EXPERIMENTAL

### Materials

The diamine and dianhydride used to synthesize the PI were 3,4'-oxidianiline (ODA), 4,4'-ODA, 3,3',4,4'-oxydiphthalic anhydride (ODPA), 3,3',4,4'-benzophenone tetracarboxylic dianhydride (BTDA), bisphenol A dianhydride (BPADA), and 3,3',4,4'-biphenyl dianhydride (BPDA). These starting materials were purchased from Shanghai Research Institute of Synthetic Resins. Phthalic anhydride (PA) (Shijiazhuang Bailong Chemical Co.) was used as an end-capping agent to control the PI molecular weight and improve the thermal stability in high-temperature circumstance. *N,N*-Dimethyl acetamide (DMAc) was purchased from Tianjin Second Chemical Reagent Factory and used as the solvent in the synthesis of polyamide acid (PAA). T300B (3000) high strength CF was purchased from Toray, Japan. Tetraethyl orthosilicate (TEOS) was purchased from Beijing Chemical Reagents Co. CNT was provided by Shenzhen Nanotech Port Co., whose diameter was 10–30 nm and length was 5–15  $\mu\text{m}$ .

### Preparation of premolding PI films

#### Pure premolding PI films

The diamine was first dissolved in DMAc solvent in a three-necked flask equipped with a mechanical stirrer. After the diamine dissolved completely, the dianhydride was added into the mixture group by group. To synthesize co-PI, two kinds of dianhydrides were added respectively, first one kind dianhydride was added, and then BPDA. The mixture was stirred till it became clear. Then the end-capping agent PA was added to control the molecular weight and improve the thermal stability. The total reaction time was about 6–10 h. The reaction of diamines and dianhydrides proceeded in an ice-water bath. The solid content of PAA solution was 20 wt %.

The PAA film was cast onto a clean glass, whose thickness was controlled by the diameter of the brass wire around a glass rod. The PAA film was imided in

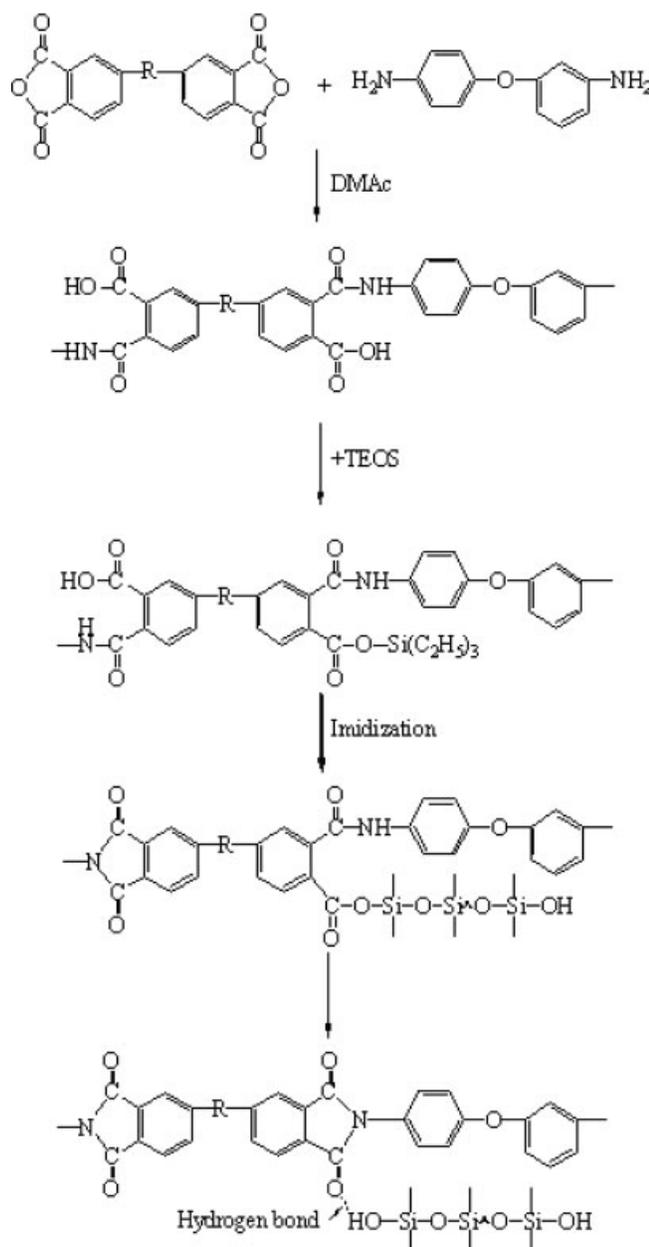
a far-infrared oven each for 1 h at 80°C, 120°C, 150 °C, and 200°C. After the temperature of oven was cooled down, the film was taken out and peeled off the glass plate by soaking in water. And then the film still needed to be dried at 100°C for 2 h in a conventional electrothermal oven. The resin systems were listed in Table I.

#### Premolding PI nano-composite films

After comparing the properties of CFRPI composites, system No.5 was selected to be used as the resin matrix in the following experiments. Two kinds of nano-particles, including SiO<sub>2</sub> and CNT, were introduced into the premolding PI films with different concentrations. The PI/SiO<sub>2</sub> nano-composites were prepared by *in situ* transition from TEOS to SiO<sub>2</sub>, and the molar quantity of TEOS was calculated according to the concentration of SiO<sub>2</sub>. That is, the synthesized PAA was mixed with TEOS for 6–8 h, and then the PAA film was imided in the far-infrared oven. The transition from TEOS to SiO<sub>2</sub> was completed in the process of imidization. However, the PI/CNT nano-composites were prepared by directly mixing the modified CNT powder in the DMAc solvent under ultrasonic for 2 h, and then the PAA was synthesized according to the method of preparation of pure premolding PI films mentioned above.<sup>15</sup> Here, the CNT powder was treated by  $\gamma$ -aminopropyltriethoxysilane (KH-560) in DMAc and dried in oven, to get the modified CNT powder. The imidization process of PAA films with nano-particles was the same with the above-mentioned. The synthesis and molecular structure of PI/SiO<sub>2</sub> and PI/CNT nano-composites were shown in Figures 1 and 2. The PI nano-composite systems were listed in Table II.

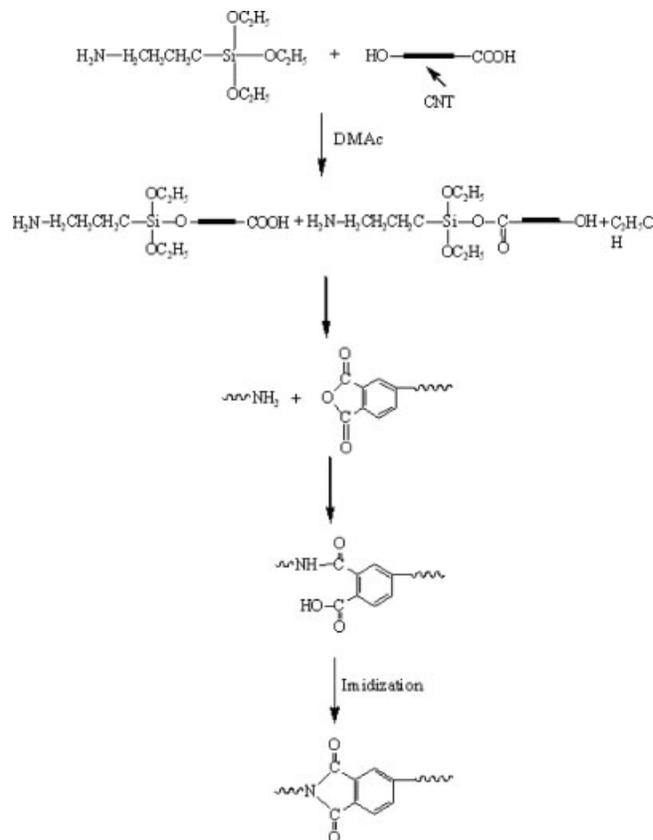
#### Preparation of CF-reinforced PI composites

The CFRPI composites were prepared by a conventional method for advanced thermoplastic composites. After diluting the PAA solution with 20% solid con-



**Figure 1** Synthesis and molecular structure of PI/SiO<sub>2</sub> nano-composites.

tent, the T300 CF was impregnated using PAA with 10% solid content to prepare CF prepreps, namely unidirectional CF fabric. The premolding PI films and CF prepreps were piled up alternately into a die to be molded. The thickness of premolding PI film was about 0.040 mm, and CF prepreps' thickness was about 0.234 mm. The laminated plate was prepared by a pressing machine, according to the following sequence of 1 h at 100°C, 1 h at 200°C, 1 h at 300°C, and 1 h at a certain final molding temperature; the pressure was controlled at 2–3 MPa; and then the CFRPI composites were cooled down under pressure. Seventeen pieces of premolding PI film and 16 pieces of CF



**Figure 2** Synthesis and molecular structure of PI/CNT nano-composites.

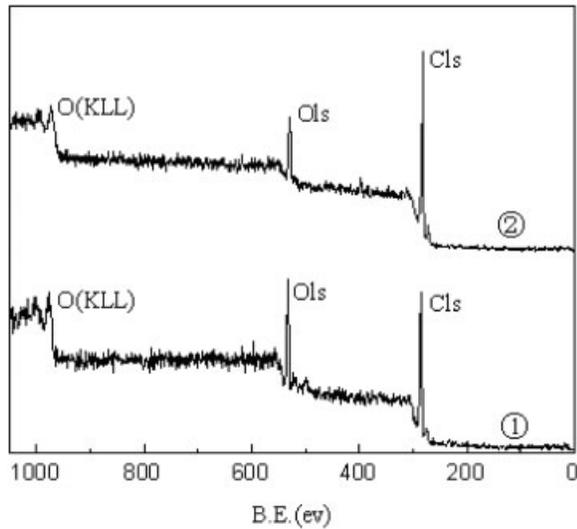
prepreg were combined into the laminated composites with about 2.100 mm thickness. The CF volume fraction was 50–60%.

### Characterization

Fourier transform infrared spectroscopy (FTIR) and X-ray photoelectron spectroscopy (XPS) were used to analyze the surface chemical properties of CF treated at 600°C for 30 min and untreated CF. The CF distribution of CFRPI composites was observed by Olym-

**TABLE II**  
System Marking for Nano-Particles Filled  
Pre-Molding PI Films

System	Nano-particles	Concentration (wt %)
A1	SiO <sub>2</sub>	0.5
A2	SiO <sub>2</sub>	1.0
A3	SiO <sub>2</sub>	3.0
A4	SiO <sub>2</sub>	5.0
B1	CNT	0.05
B2	CNT	0.1
B3	CNT	0.5
B4	CNT	1.0



**Figure 3** XPS of T300 CF ① untreated CF; ② CF treated at 600°C, 30 min.

pus BX51M microscope, and the fracture morphology was observed by S-530 scanning electron microscopy (SEM). The distribution of SiO<sub>2</sub> and CNT nano-particles on the surface of PI nano-composite films was observed by XL30 S-FEG SEM. The dynamic mechanical analysis (DMA) was applied to characterize the glass-transition temperature ( $T_g$ ). The mechanical properties of CFRPI composites were measured by SANS CMT 5504 testing machine. The rheological behavior of premolding PI films was measured by Bohlin Instruments Gemini advanced rheometer, at a heating rate of 5°C/min. The flexural testing was done according to ASTM D790. The interlaminar shearing strength (ILSS) was measured according to ASTM D2344. The fiber volume fraction was measured according to ASTM D3171. The standard deviation of mechanical properties was calculated. Here, the confidence interval for the mean loading was 95% confidence level, and the scatter range indicated was calculated by:

$$\bar{s} = \sqrt{\frac{\sum_{i=1}^n (x_i - \bar{x})^2}{n - 1}} \quad (1)$$

where  $\bar{x} = \frac{1}{n} \sum_{i=1}^n x_i$  is the average of individually measured values;  $x_i$  is value of the  $i$ th measurement; and  $n$  is the number of measurements.

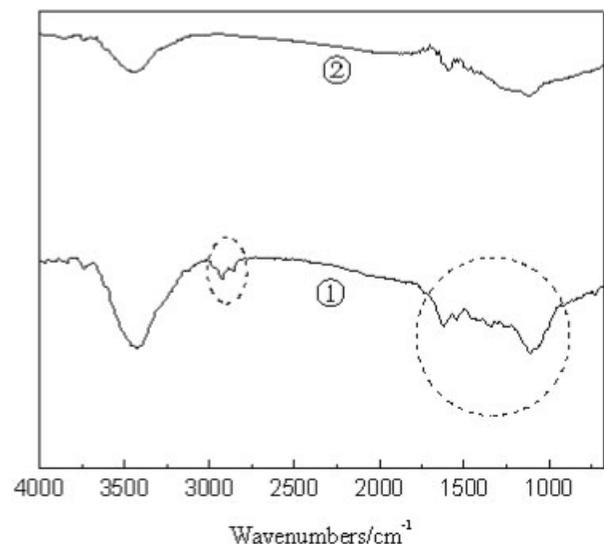
## RESULTS AND DISCUSSION

### CF surface treatment

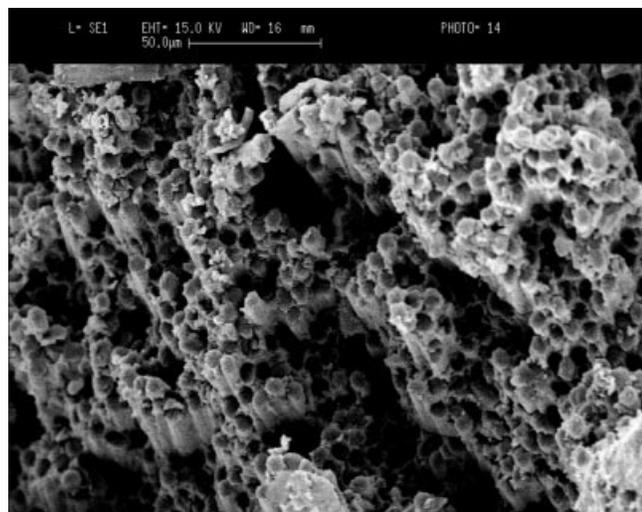
The surface characteristic of T300 CF was measured by XRD and FT-IR. After comparing the results, we se-

lected the untreated CF as the reinforcing material. In Figure 3, special peaks at 531 and 299 eV represent oxygen and carbon elements, respectively. The ratio of O element to C element of untreated CF is higher than that of CF treated at high temperature 600°C for 30 min. The decrease of active O ratio will affect the interaction between CF and PI resin, resulting in the properties of CFRPI composites decreasing. Because there was a sizing layer on the T300 CF surface, the sizing supplied some chemical activity. After the sizing was eliminated in high temperature, the chemical activity of the CF surface was weakened. Therefore, the chemical activity of treated T300 CF was lower than that of untreated T300 CF, decreasing the interaction between CF and PI resin.

In Figure 4, the active functional group decreased due to the elimination of surface sizing. The active group on the surface, mainly —OH, can act with the —COOH of PAA to increase the interfacial interaction. After the high-temperature treatment, the chemical activity of T300 CF was weakened obviously, leading to poor interface interaction. After T300 CF was treated in high temperature, the sizing was eliminated, and the surface of CF was scraggly, which improved the mechanical interaction between CF and PI resin. However, the physical action was weaker than the chemical action, such as hydrogen bond. Therefore, the T300 was used directly without any treatment in this study. Figure 5 shows the fracture morphology of CFRPI composites (system No.5), which showed that the CF bundles were wrapped by the PI resin well and that the interfacial interaction was good.



**Figure 4** FTIR of T300 CF ① untreated CF; ② CF treated at 600°C, 30 min.



**Figure 5** SEM of fracture morphology of CF-reinforced PI composites.

### Polyimide molecular structure

There are some differences in PI molecular structure, and so the properties of premolding PI film are different, and then the properties of corresponding CFRPI composites are different at the same processing conditions. The properties of premolding PI films are shown in Table III, where system Nos. 1–4 were homo-PI resins, and system Nos. 5–8 were co-PI resins. Here, ODPDA possessed —O— bond, and so its flexibility was good. BPDA was a rigid structure monomer, whose movement and adhesive ability were poor. C=O of BTDA was easy to form hydrogen bond with —O— of ODA. And BPADA exhibited good flexible and adhesive properties due to flexible bisphenol A group. The PI with rigid structure monomer exhibited high tensile strength, modulus, and  $T_g$ , and maybe its broken elongation was slightly low. And the PI with flexible monomer had a relative big broken elongation and low modulus. Moreover, the thermal resistance, adhesive properties, and processability of PI resins were mainly decided by the PI molecular

**TABLE III**  
Properties of Pre-Molding PI Films

System no.	Tensile strength (MPa)	Tensile modulus (GPa)	Elongation-at-break (%)	$T_g$ (°C)
1	93	2.0	9.4	250
2	105	2.1	6.9	282
3	107	2.0	7.9	285
4	90	1.6	9.9	218
5	99	1.8	7.0	255
6	99.5	1.8	7.7	280
7	97.5	1.8	8.7	229
8	140	2.8	8.1	270

**TABLE IV**  
Effect of Molecular Structure on the Properties of CFRPI Composites

Resin matrix no.	ILSS (MPa)	Flexural strength (MPa)	Flexural modulus (GPa)	$T_g$ (°C)
1	42.0 ± 1.4	684 ± 15.2	98.3 ± 3.2	248
2	22.6 ± 1.1	515 ± 14.1	97.6 ± 3.0	286
3	28.6 ± 1.2	536 ± 15.0	95.1 ± 3.3	276
4	66.3 ± 2.5	1512 ± 18.9	108.0 ± 3.9	220
5	115.1 ± 3.0	1791 ± 23.2	116.0 ± 4.1	251
6	25.6 ± 1.5	500 ± 14.5	90.5 ± 2.8	278
7	58.6 ± 2.3	1342 ± 25.3	101.6 ± 3.5	222
8	37.6 ± 1.4	797 ± 20.2	104.1 ± 3.5	280

structure. Therefore, the molecular structure of PI decided the processability of premolding PI films and the properties of corresponding CFRPI composites. The flexible PI exhibited low processing temperature and good ILSS, while the rigid PI needed high processing temperature.

As can be seen from Table III, system Nos. 2 and 3 exhibited high strength and modulus, while system Nos. 1 and 4 exhibited high toughness. After copolymerization, system Nos. 5–7 had similar mechanical properties, but  $T_g$  of system No. 5 was available for molding CFRPI composites. And system No. 8 needed high processing temperature.

At the selected processing condition of 370°C for 1 h, the mechanical properties of eight kinds of PI systems are compared in Table IV. The properties of CFRPI composites were decided by properties of premolding PI films. The system No. 5 exhibited better mechanical properties due to its good processability, and other systems did not. This maybe because the molding temperature was not suitable or the match of dianhydrides was not optimal. Therefore, system No. 5 was adapted to be used as resin matrix at 370°C molding temperature for 1 h. In the following study, system No. 5 was selected as the main resin matrix.

### Fiber volume fraction

Here, the effect of fiber volume fraction on properties of CFRPI composites was investigated. The fiber volume fraction changed from 55% to 65%. The corresponding mechanical properties of CFRPI composites are listed in Table V.

In Table V, the change of fiber volume fraction had little effect on the properties of CFRPI composites. In general, the mechanical properties, including ILSS, flexural strength, and flexural modulus, changed slightly. In the CFRPI composites, the PI resin acted as adhesive, and CF was the main reinforcing material. In theory, the properties of CFRPI composites should be decided by CF volume fraction. As can be seen

**TABLE V**  
Effect of Fiber Volume Fraction on Properties of CFRPI Composites

Fiber volume fraction (%)	ILSS (MPa)	Flexural strength (MPa)	Flexural modulus (GPa)
55	117.7 ± 3.1	1637 ± 14.1	118.9 ± 5.2
60	116.9 ± 2.9	1681 ± 15.6	116.0 ± 4.0
65	112.7 ± 2.6	1685 ± 18.9	116.5 ± 3.5

from Table V, when the CF volume fraction was 65%, the flexural strength was the highest, while the ILSS was the lowest due to the decrease in PI resin content. When the CF volume fraction was 55%, the content of reinforcing phase decreased, resulting in the flexural strength of CFRPI composites decreasing. When the content of PI resins increased, the ILSS of CFRPI composites was high. Because the volatile produced by premolding PI films was small, and the viscosity of PI films was good, the difference induced by CF volume fraction was little for the properties of CFRPI composites in the experiment. It showed that the CFRPI composites exhibited relatively stable mechanical properties when the CF volume fraction changed from 55% to 65%.

### Processing conditions

The processing conditions of CFRPI composites were mainly decided by the properties of premolding PI films. Taking system No. 5 as an example, the effect of molding temperature on the properties of CFRPI composites is shown in Table VI. When the molding temperature was high, the ability of premolding PI films to melt and penetrate was good. But if the molding temperature was too high, the PI films could crosslink, resulting in poor thermoplastic processability. On the contrary, if the molding temperature was too low, the ability of premolding PI films to melt and penetrate was poor, which affected PI to impregnate CF and led to form build-up resin area. From Figure 6 it can be seen that 330°C molding temperature was not enough for PI films to melt and penetrate, and 390°C molding temperature was too high to flow due to probable crosslinking. Therefore, when the molding temperature was too high or low, the PI resins build up between CF prepregs. The microstructure of CFRPI composites molded at 350°C and 370°C was better than that of CFRPI composites molded at 330°C and 390°C. However, the difference between 350°C and 370°C was little. Therefore, the optimal molding temperature was 350–370°C.

In Figure 7, the rheological behavior of premolding PI films of system No. 5 was measured. As can be seen from Figure 7, the viscosity of PI films decreased with

the temperature increasing, especially after 250°C. It indicated that the premolding PI films exhibited good thermoplastic processability.

Table VII showed the effect of molding time on properties of CFRPI composites. When the molding temperature was 370°C, the PI resin flowed and penetrated into the fiber bundles. If the molding time was short, the penetration was not enough. However, too long molding time did not improve the properties of CFRPI composites further, and it was not necessary. Therefore, the 60-min molding time was enough to prepare CFRPI composites with good properties.

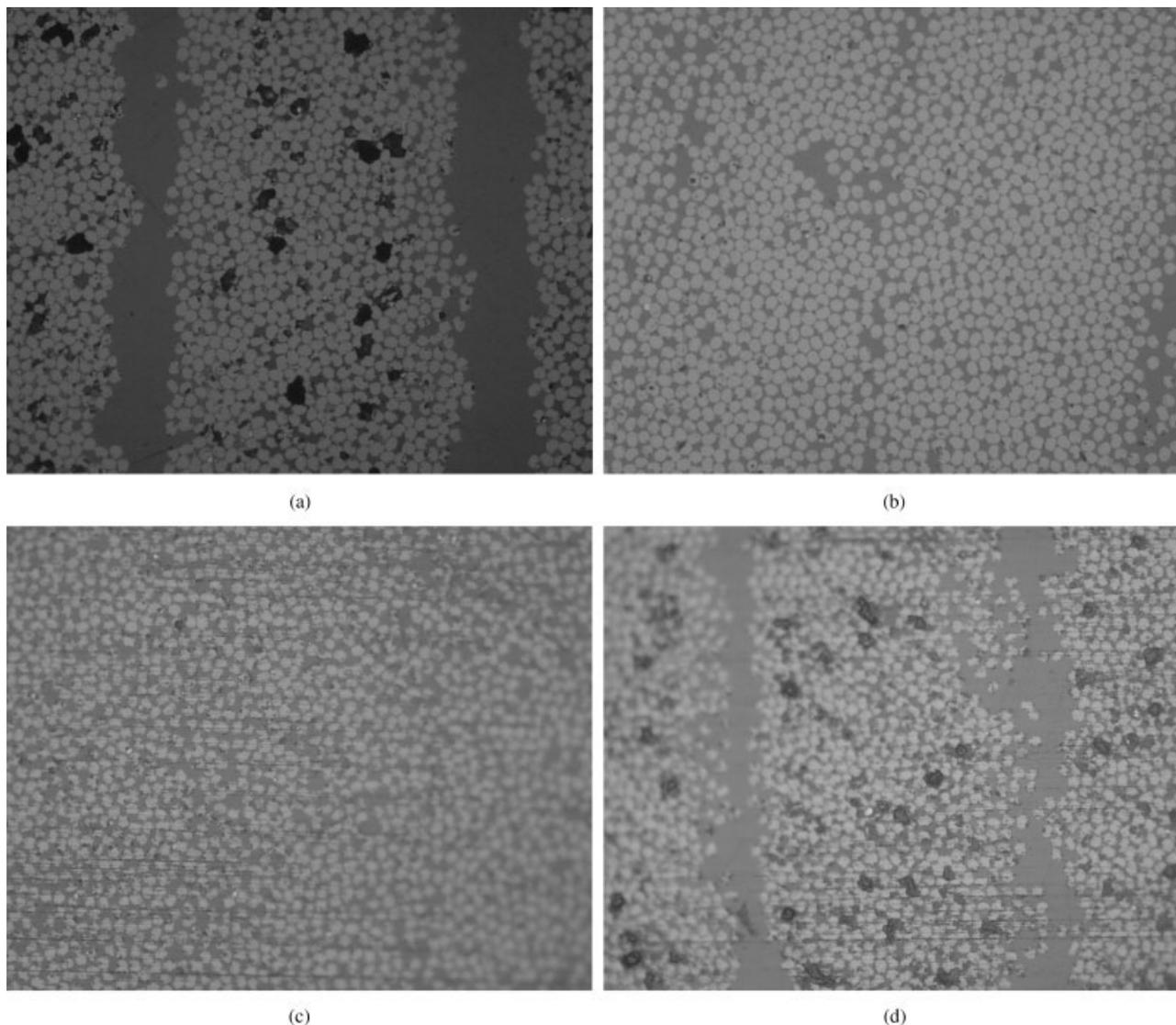
### Effect of SiO<sub>2</sub> and CNT nano-particles

Because the properties of PI were further improved due to the incorporation of inorganic nano-particles, which has been reported, the two kinds of nano-particles, SiO<sub>2</sub> and CNT, were introduced into the CFRPI composites in this study. Figure 1 indicated that the precursors of SiO<sub>2</sub> nano-particles were fixed onto the carboxyl groups (—COOH) of PAA macromolecular chain, and the SiO<sub>2</sub> nano-clusters were *in situ* produced during the following imidization process. There were a lot of silihydroxygroups (—Si—OH) with high activity on the surface of SiO<sub>2</sub> nanoclusters, which can react with carboxyl groups of the PI macromolecular chain to hydrogen bonds, so it was expected to achieve the uniform hybrid composite films. In Figure 2, the process of treating CNT with coupling agent and the reaction of CNT with PAA were shown. The couple agent improved the compatibility between inorganic and organic phase, decreasing the phenomenon of phase separation. Figures 8 and 9 show the SEM photos of PI/SiO<sub>2</sub> and PI/CNT nano-composite films, where the concentration of SiO<sub>2</sub> was 5.0 wt % and the concentration of CNT was 1.0 wt %. In Figure 8, there are some SiO<sub>2</sub> nano-clusters on the surface of the nano-composite films. In Figure 9, there are some CNT on the surface of films, and the distribution was uniform.

Because the PI/nano-particles premolding films exhibited high tensile strength, tensile modulus, and  $T_{g,r}$ , the properties of CFRPI composites were hoped to be improved further due to the incorporation of nano-

**TABLE VI**  
Effect of Molding Temperature on Properties of CFRPI Composites

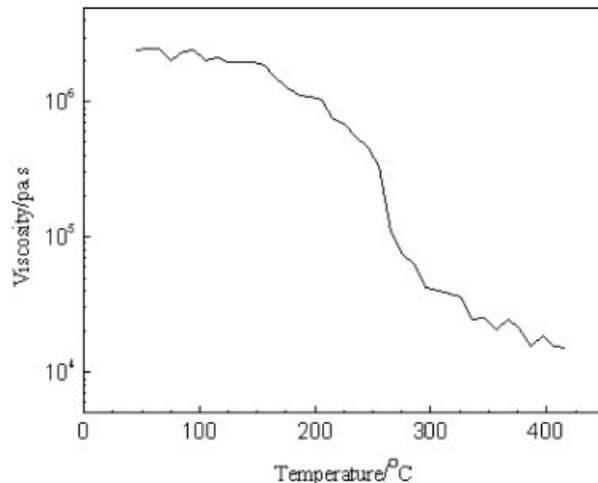
Molding temperature (°C)	ILSS (MPa)	Flexural strength (MPa)	Flexural modulus (GPa)
330	67.5 ± 1.4	536 ± 10.9	97.7 ± 3.0
350	120.3 ± 3.2	1860 ± 20.5	112.9 ± 4.4
370	115.1 ± 3.0	1791 ± 23.2	116.0 ± 4.1
390	108.2 ± 2.5	1533 ± 19.6	109.3 ± 3.7



**Figure 6** Microphotos of composites prepared at different processing temperature (magnification:  $\times 500$ ). (a) 330°C; (b) 350°C; (c) 370°C; (d) 390°C.

particles. The effect of nano-particles on properties of CFRPI composites are shown in Table VIII. For T300/PI/SiO<sub>2</sub> composites, the  $T_g$  of CFRPI composites increased from 251°C to 256°C due to the incorporation of inorganic SiO<sub>2</sub> nano-particles. Because the PI/SiO<sub>2</sub> nano-composites were prepared by *in situ* reaction of TEOS in the process of PAA imidization, the distribution of nano-particles was uniform comparatively. Therefore, the effect of SiO<sub>2</sub> nano-particles on the properties of CFRPI composites was obvious. The ILSS and flexural modulus of CFRPI composites first increased, and then decreased with the concentration of SiO<sub>2</sub> nano-particles increasing.

With the incorporation of nano-particles, the rigidity and  $T_g$  of premolding PI films increased, leading to the decrease in processability. In addition, the existence of these rigid particles in premolding films pre-



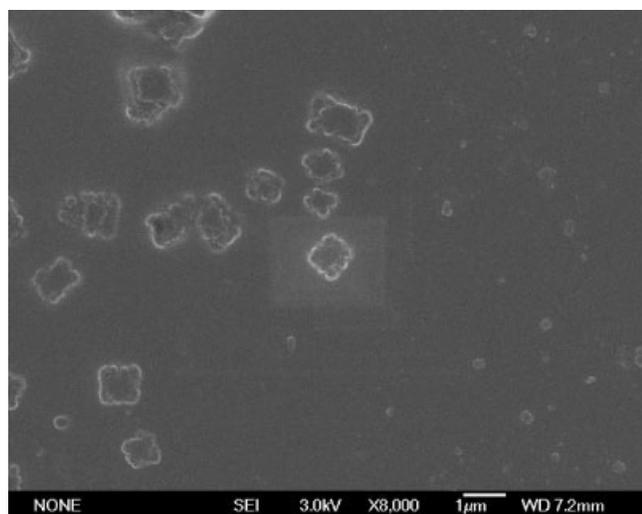
**Figure 7** Viscosity of premolding PI films changed with the temperature increasing.

**TABLE VII**  
Effect of Molding Time on Properties  
of CFRPI Composites

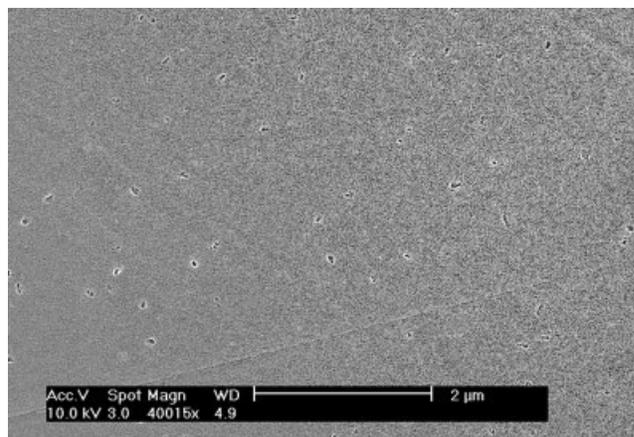
Molding temperature (°C)	Molding time (min)	ILSS (MPa)	Flexural strength (MPa)	Flexural modulus (GPa)
370	40	101.5 ± 2.8	1380 ± 15.5	93.3 ± 3.0
370	60	115.1 ± 3.0	1791 ± 23.2	116.0 ± 4.1
370	90	114.9 ± 3.3	1687 ± 14.4	106.4 ± 3.8

vented the PI resins from penetrating. Thus, the thermoplastic deformation and adhesive ability of premolding films decreased, resulting in the decrease in ILSS of CFRPI composites and the increase in modulus. However, the modulus did not increase linearly with the content of SiO<sub>2</sub> nano-particles increasing, which was related with the decrease of ILSS. When the concentration of nano-particles was low, the improvement of strength and modulus was obvious. When the concentration of nano-particles was high, the nano-particles were easy to touch each other under pressure to form stress concentration. Especially for the SiO<sub>2</sub> nano-particles on the surface, they contact with CF directly, resulting in stress concentration inside composites.

As far as T300/PI/CNT composites were concerned, the most mechanical properties of CFRPI composites decreased with the concentration of CNT increasing. The flexural modulus of CFRPI composites decreased because of the incorporation of inorganic nano-particles. At the same time,  $T_g$  of CFRPI composites was slightly increased from 251°C to 252°C. The reason for it was that even if the CNT was dispersed uniformly in the PAA, they began to reunite together in the process of imidization. And even the CNT dis-



**Figure 8** SEM of surface morphology of PI/SiO<sub>2</sub> nano-composites.



**Figure 9** SEM of surface morphology of PI/CNTs nano-composites.

persed uniformly in premolding PI film, the CNT would contact with CF under pressure. The CNT was easy to reunite because of its high surface energy, especially for the high concentration of CNT, resulting in more stress concentration. It indicated that CNT was not adapted to improve the properties of CFRPI composites. In this study, the modified CNT was used, but the further surface treatment for CNT, such as the use of other coupled agents, active monomers, surface active agents, and so forth, may improve the properties of CFRPI composites.

## CONCLUSIONS

In this study, a series of T300 CFRPI composites were prepared by a typical method of advanced thermoplastic composites. The effect of fiber volume fraction, processing conditions, molecular structure, and nano-particles on properties of CFRPI composites was investigated, and some conclusions can be drawn as follows:

**TABLE VIII**  
Effect of Nano-Particles on the Properties  
of CFRPI Composites

Resin matrix	ILSS (MPa)	Flexural strength (MPa)	Flexural modulus (GPa)	$T_g$ (°C)
pure	115.1 ± 3.0	1791 ± 23.2	116.0 ± 4.1	251
A1	118.6 ± 3.2	1667 ± 25.1	121.6 ± 3.9	252
A2	109.4 ± 2.9	1593 ± 19.5	136.6 ± 4.2	253
A3	108.4 ± 3.0	1649 ± 23.3	125.6 ± 3.9	254
A4	106.9 ± 2.8	1585 ± 27.3	121.9 ± 3.7	256
B1	113.1 ± 3.1	1640 ± 25.7	106.4 ± 3.1	251
B2	100.0 ± 2.6	1520 ± 22.8	105.8 ± 3.0	251
B3	106.2 ± 2.9	1500 ± 25.7	107.8 ± 3.5	251.5
B4	98.4 ± 2.8	1409 ± 25.0	107.6 ± 3.3	252

1. On the basis of XPS and FT-IR, the untreated T300 CF was used directly, without treating in high temperature.
2. The properties of CFRPI composites were affected by the molecular structure of PI, and system No. 5 was optimal at 370°C molding temperature.
3. The effect of CF volume fraction on the mechanical properties of CFRPI composites was little, when the CF volume fraction changed from 55% to 65%.
4. For system No. 5, 350–370°C molding temperature and 1 h molding time were optimal.
5. The SiO<sub>2</sub> nano-particles were useful to increase the  $T_g$  of CFRPI composites, the flexural modulus; and ILSS of CFRPI composites first increased, and then decreased with the SiO<sub>2</sub> concentration increasing.
6. The CNT hardly improved the properties of CFRPI composites; most mechanical properties of CFRPI composites decreased, and  $T_g$  increased slightly with the CNT concentration increasing.

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