Polyimide nanocomposites with reduced graphene oxide for enhanced thermal conductivity and tensile strength

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Abstract
Graphene oxide (GO) is reduced in dimethylacetamide by solvothermal reduction to prepare reduced graphene oxide (rGO). The thermal conductivity of rGO has been extremely improved compared with GO. The preparation method of poly amic acid (PAA) solution with rGO is in situ polymerization, then the uniformly dispersed solution of rGO/PAA is acquired. Polyimide (PI) nanocomposite films are prepared by the imidization of rGO/PAA nanocomposites. Transmission Electron Microscope (TEM) clearly shows the bonding of graphene and PI. The temperature of 7% weight loss of nanocomposite films exhibits a considerable increase (∼27 °C) by adding rGO. The tensile strength of nanocomposite films with 2 wt% rGO increases by 413.67% and the thermal conductivity coefficient increases by 1051.6% in comparison with pure PI films. The alignment of rGO is along the horizontal direction of films with rGO which can improve thermal decomposition temperature and thermal conductivity. Due to the tight bond of PI films and rGO, pulling out rGO needs more work during the stretching process, then the tensile properties of PI films are extremely improved.

1. Introduction

Polyimide (PI) is an excellent-performance material on account of its superior properties, such as high thermal resistance, mechanical strength and splendid chemical resistance. PI are widely applied in many aspects, including microelectronic devices and aerospace [1–5]. However, the pure PI films have certain limitations, for instance, their insulativity could cause the electrostatic accumulation on the surface of material [6, 7]. Besides, with the development of aircraft industry, the performance of PI films should be strengthened to meet the requirements. The structure of PI is variable, and the PI can obtain the outstanding optical and electrical properties through the selection and modification of monomers. Graphene-based nanocomposites have some excellent properties, such as high thermal conductivity and good transmittance [8, 9]. The essential application of graphene is to prepare graphene-based nanocomposites. Especially, the graphene shows the typical two-dimensional planar structure, which will have directional enhancement effect on the conductive property and mechanical property of PI films.

Due to its excellent properties and potential applications, graphene has attracted much attention [10–12]. Chemical exfoliation method to make graphene have been used in large-scale assembly and applied in nanocomposites [13, 14]. Also, graphene can induce PI films to some extent. The lamellar structure of graphene can lead to the orientation in PI films, thus improving the structure regularity of films. The existence of hydroxyl and carboxyl on the graphene oxide (GO) surface result in the thermal conductivity reduced. The oxygen groups on the reduced graphene oxide (rGO) surface decrease, and the thermal conductivity is improved compared to that of GO. Therefore, the GO should be reduced to rGO before in situ polymerization [15, 16]. Due to the reduction process of GO needs strong reductant or high-temperature condition, it not only consumes energy source but also...
2.4. Characterization
Then fully solidified carbon (crystallographic structure of the samples, with Ni carboxyl groups also exist in the rGO. Therefore, the rGO is prepared by solvothermal reduction without impurity, and the purity of graphene is high. Some hydroxyl groups and transmission electron microscopy operating at 200 kV is used to analyze morphology and interior structure of films.

3. Results and discussion

3.1. Structure and morphology analysis of GO and rGO
X-ray diffraction (XRD, figure 1(a)) spectrum of GO powder exhibits a larger interlayer spacing (GO, dominant peak at 11.6°, corresponding to an interlayer spacing of 0.76 nm), because of oxygen functional groups and the water molecules of GO existing in GO [20, 21]. In figure 1(b), the dominant peak of rGO is at about 24°. The interlayer spacing of rGO is approximately 0.367 nm, which is smaller in comparison to GO and is corresponding to the (002) graphite peak. During the reduction process, p–p interactions parts of the basilar part near the edge was reduced along with snapped, thus reducing the interlayer distance [22–24]. Figure 1(b) shows that the characteristic peak of the C–OH stretching vibration is 1630 cm⁻¹. The peak of C=O stretching vibration is 1720 cm⁻¹, and the vibration absorption peak of C–O–C is shown at 1010 cm⁻¹. Compared with GO, the intensity peak of rGO is obviously smaller than that of GO. After the heat treatment, the oxygen pollutes the environment. Therefore, the rGO is prepared by the solvothermal method [17–19]. The GO is reduced by solvothermal reduction without impurity, and the purity of graphene is high. Some hydroxyl groups and carboxyl groups also exist in the rGO. Therefore, the rGO/PI nanocomposite films can be obtained by in situ polymerization. During the subsequent carbonization and graphitization process, on one hand, the defects of carbon films and graphite films have been greatly reduced, on the other hand, the carbon yield has also been greatly improved. The fracture tensile and thermal conductivity of the films can be improved obviously.

2. Experimental

2.1. Materials
Graphite powder was bought from Nanjing XFNano Materials Tech Co. Ltd, China. Sulfuric acid (H₂SO₄, 98%), nitrate natrium (NaNO₃), hydrochloric acid (HCl 37%) and hydrogen peroxide (H₂O₂) were bought from Kelong reagent Co. Ltd, China. N, N-Dimethylacetamide (DMAC) was bought from Kelong reagent Co. Ltd, China. 1,2,4,5-Benzenetetracarboxylic anhydride (PMDA) and dianhydride (ODA) were bought from Kelong reagent Co. Ltd, China.

2.2. Preparation of rGO
The modified hummers method is used to prepared GO. GO (15 mg) was dissolved in DMAC (30 ml). The GO solution was put into a polytetrafluoroethylene reactor for 24 h under the condition of 180 °C. Then the reactor was naturally cooled to 25 °C. The in situ polymerization method is used to prepare the poly (amic acid) (PAA).

2.3. The process of preparation rGO/PI nanocomposite films
The solution of DMAC with different amount of reduced graphene was added to the clean reactor with adjusting the proper stirring rate by using N₂ as the protective atmosphere. A certain amount of ODA monomer was added to the reaction system. After completely dissolve, the proper amount of PMDA monomer was added in batches, and the mechanical stirring rate is 1200 r min⁻¹. A viscous rGO/PAA precursor solution was formed by the mixture stirred for 24 h. The prepared rGO/PAA solution was evenly coated on the clean glass slide and kept horizontally in the vacuum curing furnace at 80 °C for 3 h. Then according to the curing system, the temperature of tube furnace should be adjusted to 80 °C, 120 °C, 150 °C and 180 °C for 0.5 h and 250 °C for 1 h, then fully solidified. The rGO/PI nanocomposite films can be acquired by cooling to room temperature.

2.4. Characterization
RIGA-KUD/max-2500/PC x-ray Diffractometer (Rigaku Corporation, JAPAN) was used to determine crystallographic structure of the samples, with Ni filter, Cu as a target for K α radiation source (λ = 0.15046 nm), an acceleration voltage of 40 V, the current intensity of 100 mA, scanning speed of 5° min⁻¹, scanning interval of 0.02° and 2θ scanning range of 5° to 60°. Fourier transform infrared spectroscopy (FTIR) with total reflectance in the wave-number region of 4000 to 500 cm⁻¹ is conducted. The SEM (JSM-6701F, Japanese electronic optical, LTD, JAPAN) is used to observe the surface morphology and cross-section. Aside from above, an emission current of 2 μA and voltage from 1.0 kV to 5.0 kV were set. The high-resolution transmission electron microscopy operating at 200 kV is used to analyze morphology and interior structure of rGO/PI. X-ray photoelectron spectroscopy (XPS) (AXIS SUPRA) was performed to measure the change of groups. The tensile strength was measured by universal drawing machine (CMT4204) and the rate of extension was 5 mm min⁻¹. The thermal decomposition was measured by thermogravimetric analysis (TGA, LABSYSEVO). The thermal conductivity instrument (DRL-III) which was treated under N₂ atmosphere is used to measure the thermal conductivity of pure PI film and nanocomposite films.
functional groups are decreased, which indicates that the degradation reaction of functional groups happens in the process, and the thermal conductivity could be greatly improved by the decrease of functional groups.

In figure 2, TEM diagrams of rGO treated by the ultrasonic treatment at concentration of 1 mg ml$^{-1}$ in ethyl alcohol exhibit uniformly thick sheets. TEM diagrams show that the rGO is composed of some thin and crumpled sheets and form a disordered solid [25]. The diameter of GO and rGO lamellar is more than 1 μm, and the distance between GO layers is 0.793 nm. The ideal graphene lamellar thickness is 0.334 nm [26–28], and the graphene thickness observed in the experiment is basically 0.7–1.32 nm. The Van der Waals force of carbon and the adsorbent on surface lead to the deviation of the thickness. The GO contains a large number of hydroxyl, carboxyl epoxy and carbonyl functional groups. The π-π van der Waals force gets rid of the presence of these oxygen-containing functional groups which causes layers of GO separating and forms a larger spacing. After the reduction of GO, the oxygen-containing functional groups partially disappear, so the interlayer spacing between rGO layers decreases to 0.369 nm which is similar to the ideal graphene.

3.2. Analysis of XPS of rGO/PI nanocomposite films
The interaction between the films and nanoparticles in rGO/PI nanocomposites was studied by XPS. Figure 3 exhibits the C1s spectrum of pure PI films and 2 wt% rGO/PI. The C1s spectrum can be separated into three
components C–C (284.6 eV), C–O–C (286.3 eV) and O–C–NH (288.2 eV), respectively. The XPS pattern shows that the acylamino (O=C–NH) in rGO/PI nanocomposite films has been certainly promoted in comparison to the pure PI films, which confirms that in addition to the polymerization with PMDA, the ODA polymerize with the carboxyl groups on the graphene surface. The PI films and rGO are connected by the covalent bonding instead of simple mechanical interlocking. According to the previous research [29–32], the addition of nanoparticles can enhance the properties of composite materials. Because of the addition of rGO, PI macromolecular chain is no longer a random arrangement. It will extend outward on the plane of graphene, so it can improve its order and possess a certain orientation.

The ODA, PMDA and rGO are used to prepare PI. The rGO/PI nanocomposite films are synthesized by the two-step method. The reaction process shows in figure 4. After the solvothermal reduction, the carboxyl and hydroxyl groups on GO surface partially disappeared. In the dispersed solvent based on graphene, the carboxyl groups on graphene surface react with the amino groups on the monomer of ODA, so the bond between graphene and PI is the covalent bonding. The structure of graphene is lamellar, and the PI chain links between the layers of graphene, which can promote the tensile properties, thermal conductivity and thermal decomposition temperature of the nanocomposite films.
3.3. XRD analysis of rGO/PI nanocomposite films

In figure 5, the central diffraction peak of the films is about 18°. The structure of PI films is semi-crystalline with an amorphous structure. The crystal plane spacing can be calculated according to the Bragg formula: \(2d\sin \theta = n\lambda\) [33], and the distance between crystal planes is about 0.444~0.553 nm. However, with the content of rGO increasing, the diffraction angle moves to a high angle, and the surface crystal plane space decreases further. It can be seen that the addition of rGO has a significant impact on the crystallization of PI films. With the increase of rGO content, the preferred orientation peak becomes sharper. The formation of a relatively complete crystal structure indicates that rGO can effectively promote the grain formation and nucleation of PI. The addition of rGO destroyed the morphology of PI and formed a new co-crystalline structure of rGO/PI nanocomposite films [34-36]. The order of oriented structure in films increases.

3.4. Cross-section micrographs of rGO/PI nanocomposite films

In figure 6, it shows that the pure PI films are relatively smooth when compared to the section of the films which added rGO. After the addition of rGO, the films have some obvious protrusions and gullies. Moreover, with the content of rGO increasing, the gully size of the films becomes narrower, and the orientation of the films becomes better. The rGO lamellae mainly distribute along the plane of the films, and the rGO induces PI molecules to grow along a certain direction. Because of the planar inducement of rGO, the PI molecules have obvious orientation on the surface of rGO, thus improving the regularity of the structure of the PI molecules. When a certain amount of rGO adds, the structure of strong interaction forms owing to the strong coupling of \(\pi-\pi\) and cross-linking between molecules [37, 38].

3.5. Tensile fracture cross-section of rGO/PI nanocomposite films

In figure 7, the diagram shows the fracture morphology of rGO/PI nanocomposite films with different content of rGO. The tensile fracture cross-section surface of pure PI films is like a knife-shaped ripping fracture which is brittle, the films is rather smooth in comparison to rGO/PI nanocomposite films. The bright dots and lines reveal that the PI films are stretched out [39, 40]. The tensile fracture cross-section surface of the rGO/PI nanocomposite films is rougher in comparison to pure PI films. As the red arrows indicating, the rough surface was caused by the interface interaction between the rGO and the PI films. With the addition of rGO increasing, there are some obvious irregular tensile traces by pulling out rGO of the films. The tensile trail is more obvious, and the fracture surface is rougher with more content of rGO. The addition of rGO can improve the toughness and the plastic deformation ability of the films because of the combination with graphene. Because the graphene needs to do work in the tensile process, the tensile strength and the breaking elongation are promoted. With the addition of rGO increasing, the fracture formation transforms the simple brittle fracture into the ductile fracture, which is caused by pulling out of rGO. There is a brittle fracture with obvious plastic range, which makes the toughness increased greatly. With the increase of rGO, the dislocation easily forms during tensile deformation, which increases the toughness of the nanocomposite films [41]. The intermolecular interaction can be produced, which can significantly strengthen the mechanical properties of the nanocomposite films.
3.6. TEM analysis of rGO/PI nanocomposite films

In figure 8, the rGO nanoparticles with parallel orientation are surrounded by PI, and there are no defects such as voids or cracks at the interface, which exhibits that the good molecular bond is formed. The rGO and PI are no longer a single individual but join together by the covalent bonds to form an entirety. The results are consistent with the XPS test results. The rGO can guide the orientation of PI molecules. Most of the graphene layers distribute along the plane direction of the films. It can be inferred that the transverse flow of rGO/PI solution is caused by the film orientation along with the glass plate.
3.7. Thermal decomposition stability of PI films and rGO/PI nanocomposite films

Figure 9 shows TGA of the pure PI films and rGO/PI nanocomposite films. The temperature of 7% weightlessness of the pure PI films is 506 °C and the thermal decomposition of 7% weightlessness the rGO/PI nanocomposite films is 533 °C, which shows that the temperature of 7% weightlessness of the nanocomposite films achieves a considerable rising (∼27 °C) by adding rGO. The rGO with large length-diameter ratio produces the so-called ‘tortuous path’ effect, which is taken for barriers to hold back the permeability of oxygen and the runaway from volatile pyrolytic products. Therefore, the enhancement in thermal decomposition stability of nanocomposite films achieves [42].

3.8. Thermal conductivity and tensile strength of rGO/PI nanocomposite films

Figure 10 shows tensile strength and thermal conductivity of the pure PI films and rGO/PI nanocomposite films. In figure 10(a), the tensile strength of rGO/PI nanocomposite films was exhibited. The tensile strength of pure PI films is 66 MPa. When the content of rGO increases to 1 wt%, the tensile strength of rGO/PI nanocomposite films is 143 MPa, which shows an obvious improvement (∼216%) in comparison to that of the pure PI films. With the increase of rGO, the tensile strength increases gradually. When the rGO adds to 2 wt%, the tensile fracture strength of rGO/PI nanocomposite films is 273 MPa, which gives a visible improvement. The strength effect between rGO/PI molecules will lead to the increase of mechanical strength. With the increase of rGO content, the crosslinking effect between rGO and PI is enhanced, which makes it more difficult to pull out, and the tensile strength of the films rises further.

In figure 10(b), the thermal conductivity of nanocomposite films with 0.5 wt% rGO is 0.7249 W mK⁻¹, which increases by 214%, in comparison to the pure PI films (0.2304 W mK⁻¹). The rGO/PI nanocomposite films show the excellent coefficient of heat conductivity. The thermal conductivity of nanocomposite films with
2 wt% rGO is 2.423 W mK$^{-1}$ and increases by 1051.6% when compared to pure PI films. The highly thermal conductivity of rGO/PI nanocomposites can be imputed to the particularly stacking of rGO lamellar structure in the PI films [43–47]. From the previous analysis, it can be observed that the rGO and PI macromolecules form a network structure which improves the thermal conductivity of nanocomposite films.

4. Conclusion

In summary, there is an efficient way to produce high performance rGO/PI nanocomposite films. The rGO and PI films can make up the interface covalent bonding through the self-reaction of cross-linkable groups, thus enhancing the interface interaction. The interfaces and strengthened performances of the rGO/PI nanocomposite films lead to certain increase in thermal decomposition stability, fracture tensile properties and thermal conductivity. The improvement of tensile strength coefficient of nanocomposite films with 2 wt% rGO is 413.67%, and the improvement of thermal conductivity coefficient of nanocomposite films with 2 wt% rGO is 1051.6% when compared to the pure PI films. The temperature of 7% weightlessness of the nanocomposite films shows a reasonable increase ($\sim 27 ^\circ C$) by the addition of rGO while compared to the pure PI films. This work shows a wide range of applications of nanocomposite films with rGO.

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Conflicts of interest

There are no conflicts to declare.

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