Study of ‘one-step’ preparation of r-PET fiber-reinforced PE composites

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Abstract
In this paper, ‘one-step’ molding process was used to prepare r-PET fiber-reinforced PE composites. Effects of compatibilizer HY-3308 and fiber content on interfacial properties, thermal properties, mechanical properties, and the fiber dispersion of the composites were studied. FTIR, contact angle test, and SEM results indicated that the compatibilizer improved the interfacial combination between the PET fiber and PE matrix. DMA, DSC, mechanical properties, density, and SEM results indicated that with an increase of HY-3308 content, bonding between the fiber and matrix became tight; the phase interface became blurred, and the mechanical properties were gradually improved. Such a ‘one-step method’ preparation of the r-PET fiber-reinforced PE composites is simple, which is beneficial for the industrialization of recycling processes for r-PET fiber.

1. Introduction

The treatment of waste fibers has long been a challenge for society. As is well-known, products made of fibers are related to various aspects of human life, such as clothing, home renovation and decoration, and automobiles [1]. Normally, the service life of fiber products is short, and thus, the amount of fiber products that are disposed and the number of scraps that are generated during production are difficult to estimate. From the perspective of the annual output of various fibers, the output of chemical fibers exceeds 70% of the total output of fibers, and the total output of polyester fibers exceeds 80% of the total output of chemical fibers. In brief, waste fibers are mixed materials that have a complex composition and are mainly composed of man-made materials, which takes PET as its main component; thus, the key issue in recycling waste fibers is the reuse of PET fibers. At present, the main treatment methods for waste fibers are landfill and incineration [2]. Theformer consumes a lot of land resources and causes the pollution to soil and water resources, and the latter causes severe air pollution [3, 4]. Therefore, discovering a new method to process and recycle waste fibers as resources is urgently needed.

However, because of the presence of dyes, mixed polymers, ash, and other impurities in waste fibers, directly recycling and using waste fibers is difficult. At present, the main recycling method for the polyester fibers is to chemically degrade, purify and then polymerize them into new polyester materials for subsequent use. Polyester fibers that are recycled via this method have an excellent texture and can form a closed-loop circulation system. Recycling systems for waste polyester textiles have been established by Du Pont in the United States and Unifi in France and are representative of chemical recycling of PET fibers. Donghua University and the Fujian Huafeng Group have also jointly established a system with the same functions and have independent intellectual property rights. However, on one hand, chemical recycling of fibers uses solvents such as alcohols, alkalis, and amines [5, 6]. On the other hand, the waste includes dyes and mixed polymers, which cannot be reused, and this means that solid waste is likely to be transformed into hazardous waste of chemical treatment and to cause serious pollution. Second, this recycling method degrades PET into oligomers and even monomers, and this is conducive to refining and purifying required materials. However, this recycling method ignores the energy loss during reprocessing. Thus, this method only solves the occupation and pollution of PET on the environmental
space through high energy consumption. Because of these two reasons, the chemical recycling method has not been extensively promoted.

Other recycling methods include adding a small amount of pure polyester fiber into recycled bottle flakes and adding chain extenders or other additives to increase the molecular weight to solve the problem of decreased viscosity of PET during recycling, which affects the product properties. Then, the recycled materials are regenerated via spinning and other means. However, recycled PET fibers obtained using this method are unstable and cannot be used in high-end products [7, 8]. Moreover, because of the decreased molecular weight, the recycling process is ‘degraded regeneration’, and the product cannot be recycled again.

In addition, using recycled PET bottle flakes to prepare high-strength and high-rigidity PET-containing blend materials is a long-standing technology [9–11]. Our laboratory has also successfully prepared PET/PE blends [12], PET/POE blends [13], and so on. SEBS-g-MAH and POE-g-GMA were chosen as compatibilities, respectively. However, because the intrinsic viscosity of PET fiber is relatively low and its molecular weight is smaller than that of the bottle flakes, it is difficult to use them to prepare blended materials.

Satapathy and Nando [14] investigated mechanical properties and fracture behavior of new short PET fiber-waste PE composites. Results proved that PE-g-MAH has a good compatibilizing ability between PET fiber and PE. Zhigang Wang [15] recycled PET fibers from waste PET textiled by a PFI mill with a complex process. The composites were made by melt mixing and injection molding. Results shown that 10 wt% PE-g-MAH could be a compatibilizer in PET fiber/PE composites.

At present, there are only a few studies on the preparation of blended materials with recycled PET (r-PET) fibers. In this paper, the preparation of r-PET fiber-reinforced PE composites via a one-step method was explored using pure r-PET fibers, PE and maleic anhydride-grafted polyethylene wax (PEw-g-MAH) as the materials. The r-PET textiles only need to be cut into small pieces without further treatment. Moreover, r-PET fiber/PE composites can be produced merely through a melt mixing progress, then the composites could be mold by injection, hot-press, or any other way. In this paper, ‘one-step’ means extremely simple r-PET fiber recyle ways and composites produce progress. The effects of the amounts of compatibilizer and fibers on the properties of the composite sheets were studied.

2. Experiments

2.1. Experimental materials
r-PET fibers were selected from textile scraps and crushed materials. The composition was a single PET, and the fiber length was about 3 mm. The textiles were cut into small pieces about 3 mm in diameter without further treatment. High-density polyethylene (HDPE, Grade 00952J, ExxonMobil) with a density of 0.952 g cm$^{-3}$ and a melt index of 0.05 g/10 min (190 °C, 2.16 kg); PEw-g-MAH (HY-3308, Hangzhou Haiyi Polymer Material Co., Ltd.) with a grafting ratio of 8%. Chemically pure liquid paraffin with a density of 0.835–0.895 g cm$^{-3}$ was from Yonghua Chemical Science and Technology (Jiangsu) Co., Ltd. All of the reagents were used directly as purchased without washing or purification.

2.2. Sample preparation
2.2.1. Preparation of r-PET fiber/PE composites
Table 1 shows the specific formulations of the PET fiber/PE composites. The samples are named according to the various contents of HY-3308 and PET fibers. For example, samples composed of 1 wt.% HY-3308 and 10 wt. % PET fibers are named 1–10.

The ‘one-step’ sample preparation process was as follows: PET fiber and a flow additive were premixed. The premix, PE particles, and HY-3308 were then added in proportion and in batches into a Haake Torque

| Sample serial number | Sample name | Fiber wt.% | Polyethylene wt.% | HY-3308 wt.% | Liquid paraffin wt.% |
|----------------------|-------------|------------|-------------------|-------------|---------------------|
| 1                    | PE          | 0          | 100               | 0           | 2% of the fiber mass |
| 2                    | 0–50        | 50         | 50                | 0           |                     |
| 3                    | 1–50        | 100        | 0                 | 1           |                     |
| 4                    | 2–50        | 90         | 0                 | 2           |                     |
| 5                    | 3–50        | 80         | 0                 | 3           |                     |
| 6                    | 3–10        | 10         | 90                | 3           |                     |
| 7                    | 3–20        | 20         | 80                |             |                     |
| 8                    | 3–30        | 30         | 70                |             |                     |
2.2.2. Fiber extraction

5 g of each 0–50, 1–50, 2–50, and 3–50 samples were taken. The PE matrix was first dissolved for 20 min in boiling xylene and then filtered while hot. The PET fiber/PE mixture was filtered through a filter that was wrapped with a 200-mesh copper mesh and placed in a Soxhlet extractor [16]. Xylene was used as a solvent to extract the samples at 160 °C for 24 h. The copper mesh package was then removed, washed the fiber for 3–4 times with alcohol, and rinsed 3–4 times with deionized water. It was then dried to obtain the fiber samples.

2.3. Testing and characterizations

2.3.1. Infrared spectroscopy (FTIR) tests

A Fourier transform infrared spectrometer (Nicolet Magna-IR550, USA) was used to characterize the PET fibers that were extracted from the composites. The total reflection mode was used to test and observe variations in the surface functional groups of the fiber samples.

2.3.2. Contact angle tests

Contact angle tests were performed with deionized water on the PET fibers that were extracted from the composites (OCA20, Dataphysics Company, Germany). The relationship between variations in the fiber contact angle and the content of HY-3308 as compatibilizer was observed to indirectly determine whether or not the fibers reacted with HY-3308.

2.3.3. DMA tests

A dynamic thermomechanical property analyzer (DMA, Rheogel-E400, Japan) was used to test the storage modulus and loss modulus of the composites using its tensile mode. The sample dimensions were 40 mm × 10 mm × 3.5 mm. The vibration frequency was 11 Hz, the heating rate was 3 °C min⁻¹, and the test temperature range was from −30 °C to 100 °C.

2.3.4. DSC tests

Differential scanning calorimetry (DSC, PC200, Germany) was used to analyze the thermal properties of the composites. 5–9 mg of samples were heated from room temperature to 300 °C at a heating rate of 10 °C min⁻¹ under nitrogen atmosphere with a nitrogen flow rate of 50 ml min⁻¹.

2.3.5. Density tests

A density measurement component of a Mettler-Toledo balance was used to test the density of the composites. Water was used as the auxiliary liquid, and the density of the materials was calculated by weighing the mass of the solid measured in air (A) and in the auxiliary liquid (B) [17]. The specific formula is shown in (1):

\[
\rho = \frac{A}{A - B} (\rho_0 - \rho_L) + \rho_L
\]

where \( \rho \) denotes the density of the solid measured; A and B denote the mass of the solid measured in the air and the auxiliary liquid, respectively. \( \rho_0 \) denotes the density of the auxiliary liquid, and \( \rho_L \) denotes the density of the air. \( \rho_L = 0.0012 \text{ g cm}^{-3} \) and \( \rho_0 = 0.99704 \text{ g cm}^{-3} \).

2.3.6. Micromorphology tests

A field emission scanning electron microscope (SEM, S-4800, Hitachi, Japan) was used to analyze the liquid nitrogen-quenched section of the composites to observe the morphology of the fiber before and after modification and to observe the interfacial strength between fibers and the PE matrix. Additionally, the relationship between the surface morphology of the fiber extracted from the composites and the amount of HY-3308 added as a compatibilizer was also observed with SEM. All of the test samples were sprayed with gold, and the test acceleration voltage was 10 kV.

2.3.7. Tests of mechanical properties

According to the standards GB/T 1040–2006 and GB/T 9341–2008, the composites were cut into test pieces with the shapes required for tensile and bending tests, respectively. The tensile and bending properties were then measured using a universal tensile testing machine (GT-TCS-2000, Goodtechwill, China). The tensile rate was 10 mm min⁻¹. The bending test rate was 2 mm min⁻¹ and the bending span was 64 mm.
3. Results and discussion

3.1. Characterization of PET fibers extracted from composites

First, PET fibers extracted from the composites were analyzed using infrared spectroscopy, contact angle tests, and SEM to confirm the effect of the HY-3308 compatibilizer on the surface modification of the fibers.

Figure 1 shows FTIR spectra of PET fibers that were extracted from the composites with different compatibilizer contents. As seen in figure 1, because the monomers are terephthalic acid and ethylene glycol, the spectrum for PET fibers without compatibilizer has no methyl absorption peak at 1371 cm⁻¹ and only a weak absorption peak of methylene at 1454 cm⁻¹ [18]. While added HY-3308 as a compatibilizer, PEw-g-MAH is obtained by reacting and grafting branched polyethylene molecules with maleic anhydride; thus, there is a large amount of methylene and some methyl groups in the molecules. Therefore, the peaks of methylene and methyl groups appear in the spectra of the PET fibers that were extracted from the composites with HY-3308, and the absorption peaks became more pronounced with an increase in HY-3308 content. This observation indicates that the hydroxy functional groups on the fiber surface reacted with the maleic anhydride of HY-3308 and that the surface of the extracted fibers has PEw-g-MAH, which is connected to the fibers through chemical bonds. When the compatibilizer content was 1 wt.% and 2 wt.%, the methylene peak in the infrared spectra was small. When the compatibilizer content was 3 wt.%, the reaction between the compatibilizer and PET became pronounced in the infrared spectrum.

PET fibers are composed of polyethylene terephthalate. Although the terminal hydroxyl and terminal carboxyl groups are polar groups, the ester group, benzene ring and methylene group that constitutes the main body of the molecular chains are all hydrophobic. As a result, PET fibers are hydrophobic materials with polar groups in molecular chains. In contrast, the PE matrix is composed of a single long carbon chain, and the molecules are nonpolar; thus, its hydrophilicity is very poor. Hence, when PET and PE are blended, the difference in their molecular polarity causes a weak interfacial interaction and results in poor compatibility. Therefore, an effective means of modifying the surface of the PET fibers to improve the interfacial combination between the fibers and the matrix must be used. Table 2 shows the contact angle results of the extracted fibers. As seen from the table, when the compatibilizer was not added, the contact angle of the fibers was 115°. After the compatibilizer was added, the hydrophilicity of the PET fibers decreased. Thus, the contact angle between the fibers and water became larger, and it increased with higher compatibilizer content. When the compatibilizer contents were 1 wt.%, 2 wt.%, and 3 wt.%, the contact angles between the fibers and water reached 128°, 135°, and 139°, respectively. Because the active ingredient of HY-3308 is PEw-g-MAH, maleic anhydride can react

![Figure 1. FTIR curves of the fibers extracted from composites.](image1.png)

| Table 2. Contact angle between water and fibers extracted from composites. |
|-------------------|---|---|---|---|
| Content of the compatibilizer (wt.%) | 0 | 1 | 2 | 3 |
| Contact angle between water and fibers (°) | 115 | 128 | 135 | 139 |
with the terminal hydroxyl groups of PET, as shown in figure 2. The carbon chain part of polyethylene wax is connected to the polyester surface through the ester groups, and then it covers the fiber surface [14]. Via the effective coating modification, the surface composition of the fibers varies from PET’s ester group, terminal hydroxyl group, and terminal carboxyl group to a polyethylene wax dominated by methyl and methylene groups. Therefore, the contact angle between the fibers and water gradually increases, and thus, the hydrophobicity increases. As a result, the surface properties are more similar to those of the PE matrix, and the compatibility with the PE matrix also improves.

To more intuitively observe the surface morphologies of the r-PET fibers before and after adding the compatibilizer, SEM analysis of the extracted fibers is shown in figure 3. As seen in the figure, when no compatibilizer was added, the surface of the PET fibers was smooth and flat, and there was no adhesion on the surface of the fibers. This indicates that the combination between the fibers and the PE matrix is very weak and that the fibers have been completely pulled out. When the compatibilizer contents were 1 wt.% and 2 wt.%, the small number of HY-3308 was not enough to form a coating layer on the fiber surface but only forms a discontinuous aggregation at the position where the terminal hydroxyl groups on the fiber surface react with maleic anhydride, as shown in figures 3(b) and (c). When the HY-3308 content reaches 3 wt.%, the compatibilizer forms a coating layer that is basically continuous on the surface of the PET fibers, and so, the compatibility between the modified PET fibers and PE was greatly improved.

3.2. Properties of r-PET fiber/PE composites

Figure 4 shows variations in the storage modulus and loss modulus of the composites with respect to the content of HY-3308 when the fiber content was 50 wt.%. As seen in figure 4(a), the storage modulus of the pure PE matrix reached 1930 MPa at a temperature of −20 °C. The storage modulus of the 0–50 sample with only fiber

![Figure 2. Reaction of maleic anhydride with the terminal hydroxyl group of PET.](image2)

![Figure 3. SEM images of the fibers extracted from composites (HY-3308 content: (a) 0 wt.%, (b) 1 wt.%, (c) 2 wt.%, (d) 3 wt.%).](image3)
added and no compatibilizer decreased to 1705 MPa. When the HY-3308 content was 1 wt.%, the storage modulus of the 1–50 sample further decreased to only 1594 MPa. However, the storage modulus of the composites was significantly enhanced with the continuous addition of the compatibilizer. When the compatibilizer content was 2 wt.% and 3 wt.%, the storage modulus of the composites at −20 °C reached 2210 and 2140 MPa, respectively. With an increase in temperature, the storage modulus of the composites gradually exceeded that of pure PE. When the temperature reached 27 °C, the storage modulus of the 0–50 sample exceeded that of pure PE. When the temperature was 50 °C, the storage modulus of all the composites exceeded that of pure PE.

The main reason for the storage modulus of the 0–50 sample under low temperature was lower than that of pure PE is that when only fibers are added but no compatibilizer is added, the fibers and the matrix cannot be tightly connected; the volume change that is caused by the PE crystallization leads to gaps between the two phases. Thus, the fibers do not play a role in the ‘reinforcing phase’ and reduce the strength and modulus of the composites. After heating, the molecular chain mobility of the two phases in the 0–50 sample both increased; thus, microgaps at the phase interface gradually disappeared, and physical cross-linking and entanglement that formed between PET fibers and PE matrix slow the downward trend of the storage modulus. Therefore, the storage modulus of the 0–50 sample gradually exceeded that of pure PE. The HY-3308 content in the 1–50 sample was relatively small. From the SEM image of the extracted fibers with 1 wt.% HY-3308 shown in figure 3(b), it is found that HY-3308 locally agglomerated on the fiber surface and that a small amount of compatibilizer made the matrix and the fibers bond tightly locally, whereas the other regions are looser. Thus, this may cause more defects to be generated in the composites, which leads to a further decrease in the mechanical properties of the 1–50 sample. A comparison between the 2–50 and 3–50 samples indicates that the storage modulus of 2–50 was slightly higher than that of 3–50 when the temperature was lower than 15 °C, whereas the storage modulus of 3–50 was significantly higher than that of 2–50 when the temperature exceeded 15 °C. From the SEM images shown figures 8(c) and (d), it is found that the fibers in 2–50 and in 3–50 were binding tightly to the matrix through the compatibilizer, HY-3308. Fibers and matrix could be taken as a whole

Figure 4. Storage and loss modulus of fiber-plastic composites with different HY-3308 contents.
in both composites. Thus, the starting points of 2–50 and 3–50 storage modulus curves were close to each other. Because of more complete coating layers on fibers in 3–50, the interfacial combination between the fibers and the matrix in 3–50 was greater than that of the two phases in 2–50. With the increase of temperature, the close integration in 2–50 was affected, the load could not be transferred from the matrix to the fibers effectively. In comparison, the motion of PE molecules in 3–50 might repair structure defects between matrix and fibers, and formed a whole, finally. The load would be transferred to the fibers more efficiently. Therefore, the storage modulus of the 3–50 sample was higher than that of the 2–50 sample at high temperature.

The loss modulus curves for the composites are shown in figure 4(b). As seen in the figure, all of the curves show double peaks. Moreover, with higher HY-3308 content, the distance between the double peaks becomes larger, and the peak shape gradually becomes clear. All of these peaks are around the heat deflection temperature (HDT) of PE and the glass transition temperature of PET. This phenomenon indicates that HY-3308 increases the interfacial interaction force between the PET and PE phases. Without HY-3308, PE and PET phases show HDT and Tg in the curve, respectively. With an increase in the HY-3308 content, more PEw-g-MAH polymer covered the fiber surface, and the interfacial interaction force between PE and PET increased. Therefore, the PE matrix was gradually divided into a free PE phase and a PE phase that was tightly bonded to the PET fibers. The loss modulus of the free phase was close to HDT of pure PE. The motion of the PE phase segments that were closely bonded to the PET fibers must overcome the space resistance and interfacial friction of the PET fibers, and so, the loss modulus significantly increased and reached Tg of PET. Therefore, after adding HY-3308, the loss modulus curves of the sheets show obvious double peaks, and with higher compatibilizer content, the peak area around 80 °C became larger.

Figure 5 shows the variations of the storage modulus and loss modulus of the fiber-plastic composite sheets with respect to fiber content when the HY-3308 content was 3 wt.%. Figure 5(a) shows that the storage modulus of the fiber-plastic composites increased with an increase in the fiber content. At the beginning of the temperature increase, the storage modulus of the fiber-plastic composites decreased when the fiber content was 10 wt.%. When the content was 20 wt.%, the fiber-plastic sheet’s modulus was close to that of the pure matrix.
When the fiber content was 30 wt.%, the modulus of the sheet increased significantly. When the fiber content was 50 wt.%, the storage modulus of the sheet was the highest, and its stiffness reached a maximum. With an increase in the test temperature, the modulus of all the sheets decreased significantly, but the residual modulus of 3–50 was always higher than those of other samples. This was because when the temperature was higher than the glass transition temperature of PE, the movable segments of the PE molecular chains in 3–50 were the fewest, and the frictional resistance that the motion must overcome was large. Thus, the amount that the storage modulus decreased was the least.

As seen in figure 5(b), the loss modulus of the fiber-plastic composites gradually increased with an increase in the fiber content, and only the curve of 3–50 shows obvious double peaks. This observation indicates the following: First, the small amount fiber that was added cannot form a complete reinforcing network in the matrix, and it is tightly bonded with the PE matrix under the action of a compatibilizer, forming an overall structure dominated by the PE phase. Deformation of the composite material does not need to overcome the spatial resistance of the fiber network, and so, the loss modulus peak was a single peak that is similar to that of the pure PE matrix. Second, when the fiber content was 50 wt%, the polyester finally formed an effective reinforcing structure. Hence, deformation of the composite material must overcome the intrinsic modulus of PE and that of the fiber-reinforced structure, and this causes the loss modulus to exhibit dual peaks. Figures 4 and 5 jointly indicate that the compatibilizer guarantees tight bonding between the PE phase and PET phase, and a sufficient amount of fibers is key to the formation and function of the reinforced structure.

Figure 6 shows the DSC curves of the composites with different HY-3308 contents and a fixed fiber content of 50 wt.%. As seen from the figure, in all of the composites, the melting point of the PE phase was lower than that of the pure PE matrix, whereas the melting point of the PET phase was higher than that of pure polyester fiber. As seen from the formula table for the r-PET fibers/PE composites shown in table 1, liquid paraffin was added as a flow aid for the processing of the composites, and the added amount was 2% of the mass of the PET fibers. The liquid paraffin was a C_{16}–C_{20} alkane, which can be used as a molecular lubricant for PE to reduce interactions between the PE molecular chains and to reduce the crystallinity of PE. Therefore, after the liquid paraffin was added, the melting point of the PE phase in the composites was lower than that of the pure PE materials. The effective ingredient of HY-3308 was PEw-g-MAH. PEw is a low-molecular polyethylene that has a low molecular weight and low melting point. It can also be used as a molecular lubricant for PE. Therefore, when the content was 3 wt.%, the PE phase in the composites had the lowest melting point. In contrast to the uniform dispersion of liquid paraffin, HY-3308 remains in the composites by reacting with the terminal hydroxyl groups on the surface of PET molecules. When the compatibilizer content was insufficient, PEw-g-MAH agglomerated at the connection points that react with PET. The compatibilizer cannot be uniformly dispersed in the composites, and thus, the melting point temperatures of the PE phase in the 0–50, 1–50, and 2–50 samples were similar, and each was slightly higher than the melting point of PE in 3–50. Differences in the melting points of the PET fibers were mainly caused by variations in the crystallinity and morphology. Both the temperature of the blending process and that of the hot pressing process for the composites were maintained at 200 °C; the T_g of PET was in the range of 68 °C to 80 °C, and its melting temperature was higher than 240 °C. Therefore, the PET fibers were in a highly elastic state during processing, and the molecular segments were in a movable state. As a
result, the amorphous area of PET was constantly perfected, and the crystallinity increased; also, the melting point of the PET fibers in the composites was higher than that of the pure polyester fibers. However, as seen in the figure, the melting peak of PET in 3–50 was lower than those in other fiber-plastic composites. This is because the PET fibers were tightly bonded with the PE phase under the action of HY-3308, and the motion of the PET surface molecules was bound by the PE molecular chains. Hence, their mobility is reduced. Therefore, this also indirectly indicates that the combination between PE and PET in 3–50 was stronger than the combination of several other samples.

The curves in figure 7 show the effects of fiber content and compatibilizer content on the density of the composites. As seen in the figure, the density of the composites is relatively low when no compatibilizer was added. When the fiber content was 50 wt.% and the HY-3308 compatibilizer content increases, the combination between the fiber phase and the matrix phase increases, and the porosity of the two phases decreases; thus, the density increases significantly. The density of the composite material also indirectly shows that HY-3308 does react with the surface of PET fibers and forms effective modified coating layers. This improves the compatibility between the PET fibers and the PE phase. In the figure, it is clear that the densities of the composites with 2 wt.% and 3 wt.% compatibilizer are not much different. It was only when the fiber content was 10 wt.% that the density of the composites with 2 wt.% compatibilizer was lower than that of the composite material with 3 wt.% compatibilizer. When the fiber content was 30 wt.%, the density of the composites with 2 wt.% compatibilizer was slightly different from that of the sample with 3 wt.% compatibilizer. Thus, the effect of the compatibilizer was basically optimal when the compatibilizer content was 2 wt.%, and it was only when the content of the compatibilizer was 3 wt.% that the effect of the compatibilizer became stable.

Generally speaking, when the compatibility between the fiber and matrix is higher, the phase interface between the fiber and matrix is more blurred [19]. Therefore, the interface between the two phases in the composites was further observed using SEM, and the results are shown in figure 8. As seen in the figure, when there was no compatibilizer, obvious gaps appear between the fibers and the matrix. The fiber phase and matrix phase are only physically blended, and there is no strong interaction or connection between the PE matrix and PET fibers, that is, there are two separate phases. During cooling process, the regularity of the PE molecular chains increased, the crystallinity increased and the volume decreased although the PE melt already filled all gaps between the PET fibers during melting process, which led to the phase separation of the PE and PET phases. Hence, there is a clear gap between the fibers and the PE phase. As seen in figure 8(b), the gap between the fibers and the matrix shrunk. On one hand, a small amount of non-PET materials remained on the surface of the peeled fibers. On the other hand, most of bonding interface between the fibers and PE separated with the crystallization of PE, but there was still a small part of PET fibers that were firmly bonded to the PE matrix. The two phases in figure 8(c) were basically bonded together. The fiber phase and the matrix phase were clearly bonded; also, the cross-section fibers were not peeled off the matrix but were still embedded in the PE matrix in a tightly bonded manner, and this indicates that interfacial properties between the PET fibers and PE phase were significantly improved. In figure 8(d), the phase interface between the fibers and the matrix became blurred. The fiber with an obvious covering layer was almost perfectly integrated with the PE matrix. The fibers and the matrix were completely integrated, which indicates that the fiber and matrix were completely bonded. Therefore, in the composites, the compatibility between the polyester fibers and the PE matrix increased with an
increase in the HY-3308 content. When the HY-3308 content was 3 wt. %, a complete modified coating layer formed on the surface of the polyester fibers.

Figure 9 shows the relationship between the bending properties of the fiber-plastic composites and the contents of HY-3308 and polyester fibers. As seen in the figure, when the fiber content was less than 20 wt. %, the flexural strength and flexural modulus of the fiber-plastic composites were basically unrelated to the HY-3308 content. After the fiber content was 30 wt. %, the flexural strength of the fiber-plastic composites increased significantly with an increase in the HY-3308 content. In contrast, the flexural modulus only increased significantly when the HY-3308 content was 3 wt. %. The reason for this is that a low fiber content was not able to form an effective reinforcement network in the matrix; instead, the fibers were present in the form of agglomerates in the composites, so the combination between the fibers and the PE (that is, the compatibilizer content of HY-3308) had little effect on the flexural strength and flexural modulus of the materials. When the fiber content was 30 wt. %, a reinforcing network of fiber was constructed. Consequently, the combination between the fibers and the matrix significantly affects the mechanical properties of the composites. When the compatibility between the fiber phase and the matrix phase was higher, the rigidity of the composites was better, and the bending strength was higher. When the content of the HY-3308 compatibilizer was 3 wt. % and the fiber content was 30 wt. %, the flexural modulus of the composites increased sharply. This is because the compatibility between the fibers and PE increases and the combination between the two phases becomes larger. Therefore, the ability of the sheets to resist deformation becomes stronger, and the flexural modulus of the sheets increased significantly. During the experiments, only the bending test piece of the 3–50 sample broke, and the other test pieces did not break after the bending experiments. This is because the polyester fibers and the PE matrix were tightly bonded, and they shared the sample deformation during the bending process. When polyester was processed, its crystalline form changed and toughness severely reduced, exhibiting the state of brittle materials under the action of an external force. Therefore, the fiber is prone to break during the bending process and causes the PE wrapping around the PET fibers to break together as well. When the content of the compatibilizer was 3 wt. %, the test piece of the 3–50 composites already shows a brittle state. Thus, the research on the sample with 5 wt. % HY-3308 was not continued.
4. Conclusions

Recycled PET fiber-reinforced PE composites were prepared via internal mixing-hot pressing. This ‘one-step method’ process simplifies the treatment for r-PET fiber and is conducive to industrial use, which has pronounced practical significance for further application of r-PET fiber. The specific conclusions are as follows:

(1) FTIR, contact angle tests, and SEM results of the fiber samples extracted from the composites indicate that HY-3308 reacted with the PET fibers results in a significant improvement of the combination between the PET fiber and PE matrix phases.

(2) DMA, DSC, mechanical properties, density, and SEM analysis of the composites indicate that the higher content of HY-3308 makes bonding between the fiber and the matrix becomes stronger and the mechanical properties enhance significantly.

(3) The mechanical properties indicate that 30 wt% addition of PET fiber is the minimum amount to form a effective reinforcement network in the matrix. Combining with the results of SEM, 3 wt% of PEw-g-MAH can form effective modified coating layers on PET fibers. And then, coupling between fibers and matrix is high enough to transfer the external load to the fibers.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).
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