

Morphological Structure and Mechanical Properties of *In Situ* Microfibrillar Composites of Modified PA66 with PP

Linghe Cheng, Wanjie Wang, Jingwu Wang, Yanxia Cao, Guixun Li, Yelong Shen

College of Materials and Engineering, Zhengzhou University, Zhengzhou 450001, China

Correspondence to: L. H. Cheng (E-mail: clh6603@126.com)

ABSTRACT: *In situ* microfibrillar composites (PP/mPA66) of modified polyamide66 (mPA66) with polypropylene (PP) were prepared by using a “post-compatibilization” technique. The mPA66 was firstly obtained by reactive extrusion of PA66 resin with a specially designed compatibilizer, which was then blended with PP through extrusion combined with a hot stretching and subsequently quenching process. The PP/mPA66 *in situ* microfibrillar composites were comparatively studied with simply blended samples of PP/PA66 that were prepared by blending PA66 and PP together with (or without) the same compatibilizer through extrusion. PA66-*g*-PP (and/or elastomers) graft copolymer formation in mPA66 was identified by dissolution test and infrared spectroscopy measurement, the compatibilizer is unevenly dispersed with large domains in PA66 as observed by scanning electron microscope (SEM). In PP/mPA66 composites, the *in situ* generated PA66 microfibrils have a rather nonuniform diameter distribution and a very rough surface. SEM observations for the fractured surface illustrated that PP/mPA66 composites have structural characteristics of stronger adhesion and moderate flexibility of the interface. Enhanced compatibilization between the PA66 microfibrils with the PP matrix resulted in improved mechanical properties of the PP/mPA66 composites. With optimized composition, the PP/mPA66 composite has notched Izod impact strength, flexural modulus, and tensile yield stress of 1.49, 1.16, and 0.99 times as those of the neat PP, respectively. Such enhanced mechanical properties balance and improved interface adhesion were not found in the simply blended samples of PP/PA66 with or without the specially designed compatibilizer. © 2012 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* 000: 000–000, 2012

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INTRODUCTION

In situ microfibrillar composites, which consist of a thermoplastic polymer as matrix and *in situ* thermoplastic polymer microfibrils as reinforcement, are attracting many attentions,^{1–8} because they provide a preferable way of achieving enhanced mechanical properties of composites. Polypropylene (PP) has such properties as low price, good chemical resistance, and so forth, and was thus studied to improve its mechanical properties by processing it with polyamide66 (PA66), in which the dispersed phase forms *in situ* reinforced microfibrils. However, the toughness of PP/PA66 composites with (or without) a compatibilizer was enhanced, and the rigidity (i.e., modulus and the tensile strength) was decreased,^{9,10} or the opposite was the case.¹¹ Even if they were all improved, the increment of tensile strength was comparatively limited.^{12–14} All these might be attributed to a weak adhesion between the reinforcing phase and the polymer matrix, an inflexibility interfacial layer, or a low aspect ratio of the generated microfibrils.

From conventional fiber-reinforced matrix, such as glass fiber reinforced plastics etc., we learnt that enhanced mechanical properties balance could be achieved with structural characteristics of stronger adhesion and moderate flexibility of the interface,^{15,16} or with profiled microfibrils.^{17,18} Thus, we attempt in this article to prepare PP/PA66 *in situ* microfibrillar composites with these structural characteristics aiming to achieve enhanced mechanical properties balance.

Considering the feature of PA66 microfibrils generated *in situ* in the targeted PP/PA66 composite and thermodynamic incompatibility between the PP matrix and the PA66 reinforcing phase, we concentrated our efforts in improving compatibility between PP and PA66, achieving profiled microfibrils formation, and attaining a stronger adhesion and moderate flexibility of the interface.

A series of specially designed compatibilizer with different composition was synthesized firstly starting from PP, ethylene-propylene, or butadiene-styrene elastomer, an epoxy compound,

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and an assisted monomer in the presence of a free radical initiator, which will appear in a separate publication. Fractional extraction and infrared spectroscopy illustrated that the prepared compatibilizers consist of unreacted PP and/or elastomers, homo- and/or copolymer of the monomer and the epoxy compound, graft copolymer of PP and elastomers, and crosslinked copolymer of PP and/or elastomers through polymer chains derived from epoxy compound and the monomer. Compatibilizers with these structural characteristics would assure the compatibility with PP matrix and would be capable to react with PA66 in the succeeding composite fabrication.

The obtained compatibilizer was allowed to react with PA66 through reactive blending yielding modified PA66 (mPA66), in which the compatibilizer is unevenly dispersed with large domains in PA66. PP/mPA66 *in situ* microfibrillar composites were finally obtained by thermal mechanical melt blending the mPA66 with PP. During the *in situ* microfibrillar formation, the compatibilizer may prefer to diffuse from mPA66 to PP matrix due to its compatibility with PP, which is determined by the decrease in interface tension energy and the increase in molecular freedom. Yet, the diffused compatibilizer may locate at the interface, which is restricted by the chemical linkages. As a result, the *in situ* generated PA66 microfibrils have profiled morphological structures, and the compatibilizer containing elastomer locates at the interface, which impart the final PP/mPA66 composites stronger adhesion and moderate flexibility of the interface. We name this as a “post-compatibilization” technique in differentiating it with conventional compatibilization techniques.

The present article reports the preparation, morphological characterization, and mechanical properties of PP/mPA66 *in situ* microfibrillar composites fabricated with this “post-compatibilization” technique. With optimized composition, the PP/mPA66 composite has enhanced mechanical properties balance as desired.

EXPERIMENTAL

Materials

Isotactic PP (F401) resin with a melt flow rate of 2.4 g/10 min (230°C, 2160 g) used for the matrix was purchased from Luoyang Petrochemical (China). PA66 (EPR27) as the microfibrillar candidate is a commercial injection grade polyamide with a relative viscosity of 2.67, which was supplied by Pingdingshan Shenma (China). Melting points of PP and PA66 were determined as 159.6 and 262.3°C, respectively by differential scanning calorimetry (DSC).

The specially designed compatibilizer was prepared according to the following procedures. PP (F401), ethylene–propylene elastomer (labeled as M), and butadiene–styrene elastomer (labeled as N) were charged into a flask in desired proportions. The mixture was stirred and heated thermostatically until homogeneous, then the epoxy compound (labeled as G) and the assisted monomer (labeled as S) were added together with a xylene solution of the radical initiator. After 160 min, the reaction was stopped and polymeric product was precipitated, filtrated, washed, and dried in a vacuum oven for 4 h. In the present

study, the specially designed compatibilizer has a composition with weight ratios of (G + S)/(PP + M + N) as 12/100, PP/(M + N) as 62/38, and M/N as 80/20, and the molar ratio of G/S is 4/6.

Preparation and Characterization of mPA66

PA66, predried in a vacuum oven at 80°C for at least 12 h, was dry-mixed with the compatibilizer in weight ratios of 95/5, 85/15, and 75/25, and then extruded with a SHJ-20 twin-screw corotating extruder equipped with a screw of D 21.7 mm and L/D 40, (Nanjing Jieente, China). The extruded filaments were immediately quenched in a water bath and then pelletized. The temperature profile from hopper to die of the extruder was 260, 275, 280, 280, and 275°C, and the screw rotation was maintained at 120 rpm.

The Molau test was performed by stirring about 0.4g of fine particle in 10 mL of formic acid and storing the test tubes for 24 h^{19,20} for PA66, PA66/(PP + M + N) (75/25), and mPA66 (with PA66/compatibilizer of 75/25). PA66/(PP + M + N) melt blend was obtained by extrusion of PA66 with PP + M + N mixture that was prepared starting from PP, ethylene–propylene and butadiene–styrene elastomer with similar procedures as that of the compatibilizer.

For Fourier transform infrared spectroscopy (FT-IR) studies, the soluble and insoluble fractions in formic acid were separated by centrifugation, and then the insoluble part was shaken with fresh formic acid for several hours to remove the residual PA66. The solid residue was washed with water, dried, weighed, and then extracted with hot xylene for 6 h to remove PP and the two elastomers. Finally, the insoluble polymer ($R_{\text{insoluble}}$) was separated by filtration, dried and weighed.

Infrared analysis was conducted with a Tensor27 FT-IR spectrometer (Bruker, Germany). The samples are thin films prepared by hot pressing at 230°C for the compatibilizer, and 270°C for $R_{\text{insoluble}}$.

Preparation of *In Situ* Microfibrillar Composites

The mPA66 pellets, predried for at least 12 h under vacuum at 80°C, were mixed with PP in a constant weight ratio of 75/25 (PP/mPA66), then extruded with the same extruder mentioned above through a roundness die (diameter of 3 mm). At the die exit, the extrudate was hot drawn by a take-up device with traction-rollers to allow the PA66 form microfibrils, while the two polymers were still in the molten state. The extrudate drawing ratio was maintained at 8 by adjusting the speed of the rolls. Subsequently, the stretched extrudate was immediately quenched in a cold water bath (about 15°C) and then pelletized. The temperature profile from hopper to die of the extruder was 170, 275, 280, 280, and 275°C, and the screw rotation was maintained at 90 rpm.

Three *in situ* microfibrillar PP/mPA66 composites were prepared with compatibilizer content of 6.25, 3.75, and 1.25, respectively. The corresponding samples are indicated as PP/mPA66(6.25), PP/mPA66(3.75), and PP/mPA66(1.25), respectively. Similarly, simply blended PP/PA66 composites were prepared by blending PA66 and PP together with (or without) the compatibilizer, and these samples are indicated as PP/PA66/C(6.25) and PP/PA66.

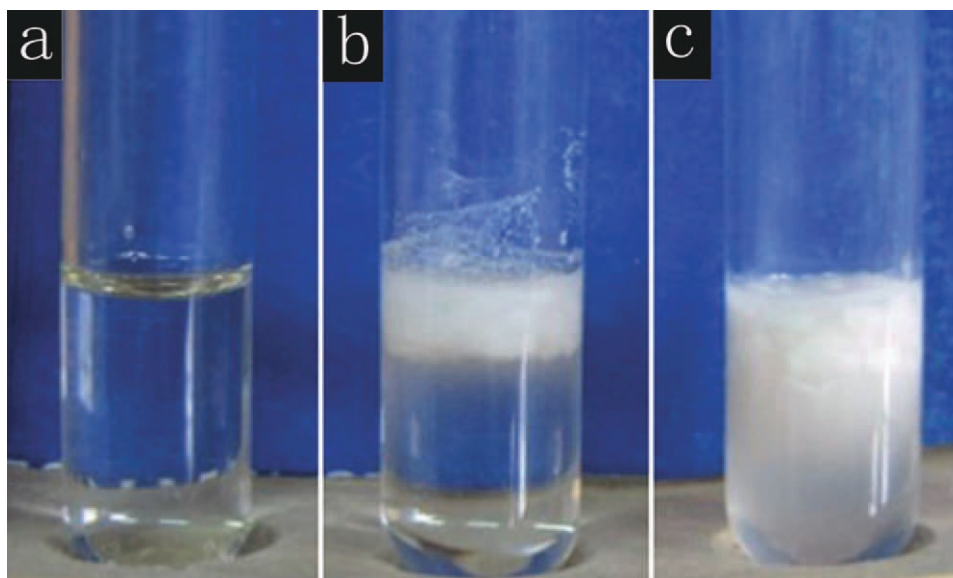


Figure 1. Dissolution of PA66 and blends in formic acid. (a) PA66, (b) PA66/(PP+M+N), and (c) mPA66. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Mechanical Properties Testing

All the composites were injection molded into specimens for testing with processing temperatures of 170, 190, 210, and 205°C, respectively, from the hopper to the nozzle, which were far below the melting point of PA66, and a mold temperature of 60°C.

Tensile yield stress (TYS) was tested on a CMT6104 universal material testing machine using dumbbell-shaped specimens at a crosshead speed of 50 mm min⁻¹ following Chinese Standard GB/T 1040.2-2006. Flexural modulus (FM) was measured on the same machine at crosshead speed of 2 mm min⁻¹ based on Chinese Standard GB/T 9341-2008. Notched Izod impact strength (NIIS) was determined on a XJU-22 pendulum impact tester with an impact rate of 3.5 m s⁻¹ following Chinese Standard GB/T 1843-2008. An average value was obtained with at least five measurements for each sample.

Morphology Observations of mPA66 and *In Situ* Microfibrillar Composites

Morphological structures were observed in a JEOL JSM-6700F scanning electron microscope (SEM). For the SEM observation of the mPA66, the extrudate was held in liquid nitrogen for 15 min, and then was broken up perpendicularly to the extrusion direction; the fracture surface was coated with a layer of gold. Samples for the observation of the *in situ* microfibrillar composites were prepared by etching the PP matrix with hot xylene at 125°C, and then the surfaces were coated with a layer of gold. The impact-ruptured surface morphology of the injection molded bars was also observed by SEM.

RESULTS AND DISCUSSION

Chemical Composition and Morphology of mPA66

Figure 1 shows dissolution of PA66, PA66/(PP+M+N), and mPA66 in formic acid. PA66 is completely soluble in formic acid forming a quite clear solution. The dissolution of PA66/(PP+M+N) yields obvious phase separation with a clear solu-

tion containing dissolved PA66 at the bottom, and the insoluble (PP+M+N) floated on the top. For mPA66, the dissolution leads to two phases, the insoluble PP and elastomers on the top, and the white turbid colloidal solution at the bottom, which is taken as an indication of graft copolymer existence. Tuzar found that a graft copolymer polyisoprene-graft-polystyrene could self-associate to form micelles in selective solution, which had central core, consisting solely of ingredient immiscible in the solvent, and an outer corona, formed of soluble composition swollen by the solvent.²¹ Therefore, we may infer that the white colloidal fraction in the case of mPA66 consists gradient distribution of PA66-*g*-(PP and/or elastomers) graft copolymers and PA66 chains from the top to the bottom.

Figure 2 shows the infrared spectra comparison of PA66, compatibilizer, and $R_{\text{insoluble}}$. The IR spectrum of $R_{\text{insoluble}}$ shows all

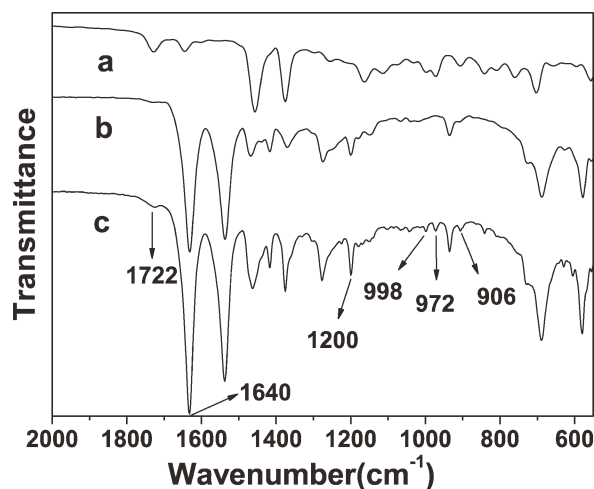


Figure 2. FTIR spectra of samples. (a) The compatibilizer, (b) PA66, and (c) $R_{\text{insoluble}}$ in mPA66.

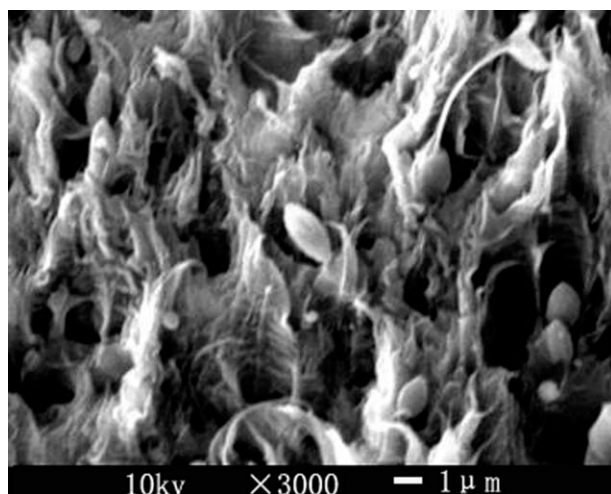


Figure 3. SEM image of the fractured surface of mPA66.

the characteristic absorptions of both the compatibilizer and PA66, in which the signals at 1722 cm^{-1} (ester group vibration of the epoxy compound), 972 cm^{-1} (aliphatic chain vibration of PP), and 906 cm^{-1} (butadiene vibration of elastomer N) are the characteristic absorptions of the compatibilizer,²² and the signals at 1640 and 1200 cm^{-1} (amide C=O stretching and N—C=O skeletal vibration) are derived from PA66.²³ These provide further proves for the formation of PA66-g-(PP and/or elastomers) graft copolymer in mPA66.

Both dissolution and IR study illustrated that the prepared mPA66 consists of unreacted PA66, 2.9% PA66-g-(PP and/or elastomers) graft copolymers, as well as unreacted PP and elastomers in the compatibilizer.

Figure 3 presents the morphology of the ruptured surface of mPA66. It can be seen that the compatibilizer particles with large domain are dispersed in the PA66 phase, which was resulted from the reactive compatibilization of PA66 with the compatibilizer. It can also be observed that the particles, although completely exposed to the ruptured surface, have an uneven distribution by major axis running parallel to the extrusion direction, which provides another support for the existence of chemical interaction between PA66 and the compatibilizer.

Microfibrils Formation in PP/mPA66(6.25) Composite

Figure 4 presents the morphologies of different microfibrillar composites in which the PP matrix has been etched away. It can be seen that PA66 microfibrils in PP/PA66 have a rather uniform diameter distribution and a very smooth surface [Figure 4(a)], which was resulted from the incompatibility of PA66 with PP. While in PP/mPA66(6.25) composite, PA66 microfibrils have a rather nonuniform diameter distribution and a very rough surface with many knots and pits [Figure 4(e)], which was originated from the influence of the compatibilizer and the “post-compatibilization” technique. At first, the embedded compatibilizer in mPA66 has a higher interfacial tension leading to larger mPA66 particles in the PP matrix during melting. During the *in situ* microfibrillar formation, the compatibilizer may prefer to diffuse from mPA66 to the PP matrix due to its compati-

bility with PP, which is determined by the decrease in the interfacial tension energy and the increase in the molecular freedom.^{24–26} In other words, the diffusion is controlled by the dual effect of enthalpy and entropy. Yet, the diffused compatibilizer may locate at the interface, which is restricted by the chemical linkages. As a result, a stronger adhesion between the PP matrix and PA66 microfibrils is constructed, which is an expected result by using the “post-compatibilization” technique. Meanwhile, the generated PA66 microfibrils have nonuniform diameters, which is resulted from heterogeneous distribution of compatibilizers in mPA66. Part of the compatibilizer aggregates with more chemical linkages can not be etched by hot xylene, resulting in knots on the surface of microfibrils, and part of them are prone to be dissolved in xylene, leading to pits. Therefore, PP and PA66 may be mutually embedded, which would further enhance the interfacial adhesion. These microfibrils may be regarded as profiled ones. With the decrease in compatibilizer amount, the corresponding diagram of the PP/mPA66 composites shows a decrease in the number of knots and pits on the surface of formed PA66 microfibrils [Figure 4(c,d)].

As discussed above, the compatibilizer can impart the final composites structural characteristics of profiled microfibrils and stronger interfacial adhesion. However, it is not totally the case for composites prepared by conventional compatibilization technique. Although with the same compatibilizer, the composite PP/PA66/C(6.25) appeared to have stronger interfacial adhesion, while PA66 microfibril formation was not observed [Figure 4(b)]. By using the conventional compatibilization technique, part of the compatibilizers would locate at the interface, the decrease of interfacial tension resulted in smaller PA66 particles during the dispersion process. Additionally, the compatibilizer coats the surface of PA66 droplets with an extremely thin layer acting as a surfactant during mixing, and prevents their aggregation during the quick extrusion and hot stretching.²⁷ Thus, the conventional compatibilization technique can improve the compatibility and form a stronger interfacial interaction between PP and PA66, but can not impart the resulting composite a reinforcement effect of microfibrils.

Fractured Surface Morphology of PP/mPA66(6.25)

The fractured surface morphology of the injection molded samples is shown in Figure 5. For the PP/PA66 sample [Figure 5(a1,a2)], it can be seen that the pulled microfibrils leave holes with generally larger diameters than those of the microfibrils; the cavitation occurs at the interface and the pulled PA66 microfibrils present relatively smooth surface. These suggest typical weak adhesion and inflexibility of the interface, which may be resulted from the inherent incompatibility of PP with PA66 and the insufficient wrapping of the PP matrix on PA66 microfibrils. An inflexible interface can constrain conformational transformation from macromolecular chains and then craze and cracking may occur in matrix when hoop tensile stress exceeds the tensile strength of PP,²⁸ thus the wrapping effect on the dispersed phase is reduced. Radial contraction stress may firstly be yielded due to the difference in thermal shrinkage of PP and PA66 during the cooling process from the injection molding temperature, and then is changed into the hoop tensile stress

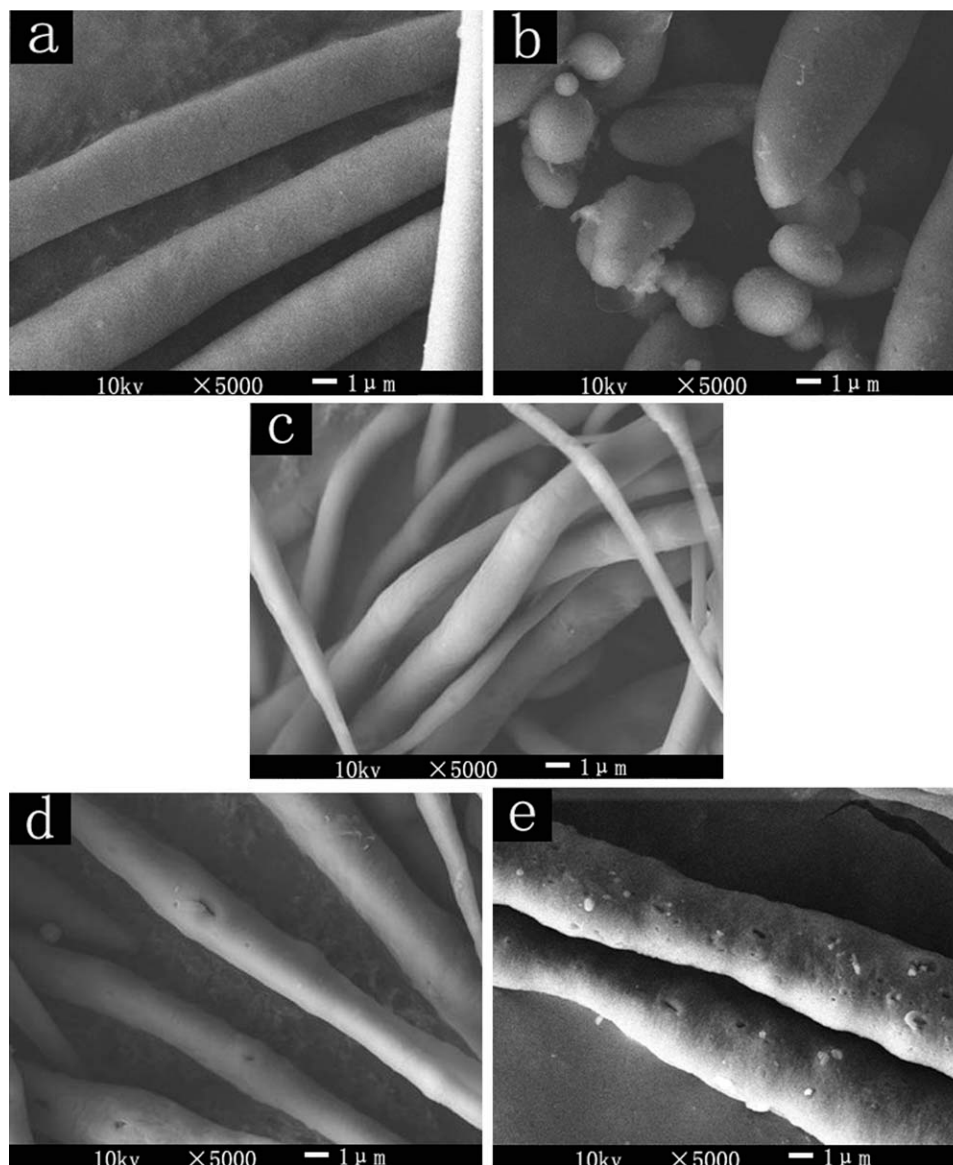


Figure 4. SEM images of PA66 microfibrils in composites. (a) PP/PA66, (b) PP/PA66/C(6.25), (c) PP/mPA66(1.25), (d) PP/mPA66(3.75), and (e) PP/mPA66(6.25).

for the supporting effect of microfibrils.²⁹ Under loading conditions, cavitations occur when stress passes through the interface, and the pulled PA66 microfibrils have smooth surface and the left holes do not shrink.

For PP/mPA66(6.25), holes with smaller diameters, stress whitening, and the pulled microfibrils with adhered resin on the surface can be obviously observed in Figure 5(c1,c2). These phenomena suggest structural characteristics of stronger adhesion and moderate flexibility of the interface. As previously discussed, the compatibilizer located at the interface and the mutually inserted two components would construct a stronger adhesion. Meanwhile, the compatibilizer containing elastomers at the interface would form a moderate interfacial flexibility, which may lead to a conformational transformation from rather coiled to relatively stretched state. The transformation accompanies a resilience force, which creates the wrapping of the PP matrix on

PA66 microfibrils.²⁸ As a result, PA66 microfibrils exhibit a distinct reinforcing effect due to the efficient stress transfer through the bonded interface under loading. When microfibrils are pulled out, the matrix at the interface shows significant plastic deformation, and then is broken up in the form of shear destruction, which is evidenced by the microfibrils surface adhered with plenty torn matrix and the holes with obvious stress whitening.^{30,31} In addition, the conformation of flexible chains returns from entropic unstable stretched state to original coiled state driven by the resilience force, which resulted in the holes shrinkage.

From Figure 5(b1,b2), it can be seen that the PP/PA66/C(6.25) composite constructs characteristics of a certain strong adhesion and moderate flexibility of the interface. While, due to the lack of microfibrils formation, a reinforcing effect could not be achieved.

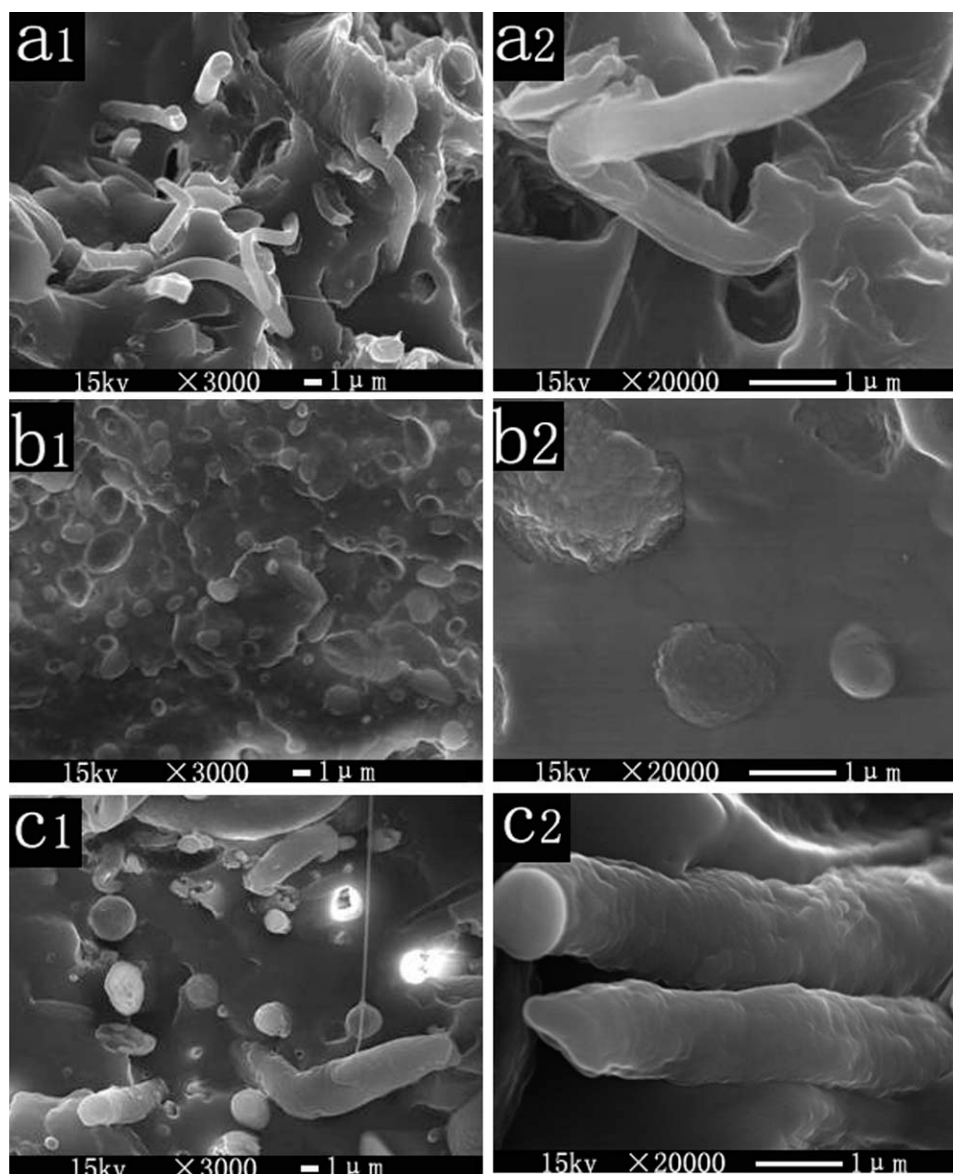


Figure 5. SEM images of the fractured surface of injection molded composites. (a1, a2) PP/PA66, (b1, b2) PP/PA66/C(6.25), and (c1, c2) PP/mPA66(6.25).

Mechanical Properties of PP/mPA66(6.25)

The mechanical properties of PP and injection molded microfibrillar composites are shown in Table I. It can be seen that TYS, NIIS of PP/PA66 sample are greatly lower than those of the neat PP, although FM is slightly higher, which is due to the weak adhesion and inflexible interface. Not only PA66 microfibrils hardly play a reinforcement role due to the inefficient shear stress transfer, but also cavitations are initiated by the high stress concentration effects at the end of microfibrils. Thus, both TYS and NIIS are greatly decreased. Conducting at a very low strain in short time, no relative slip may occur between PA66 and PP, and microfibrils can bear a part of external forces. Thus, the FM is slightly increased.³²

The addition of compatibilizer by conventional compatibilization technique makes the PP/PA66/C(6.25) sample have a greatly lowered NIIS, a similar FM and a slightly lowered TYS

as compared with those of the neat PP. The reduction in NIIS can be ascribed to three aspects. Firstly, the composite has a spherical or ellipsoidal dispersed phase, which is similar to rigid organic particle toughened plastics. Just if the plastics itself have

Table I. Comparison of Mechanical Properties^a

Material	TYS (MPa)	FM (GPa)	NIIS (kJ m ⁻²)
Neat PP	34.59	1.07	2.84
PP/PA66	26.73	1.34	2.34
PP/PA66/C (6.25)	32.17	1.09	2.27
PP/mPA66 (6.25)	34.29	1.24	4.15
PP/mPA66 (3.75)	32.17	1.19	3.21
PP/mPA66 (1.25)	30.79	1.16	3.10

^aTest temperatures: TYS and FM: 10°C; NIIS: 20°C.

higher toughness, improved NIIS could be achieved.³² However, PP matrix does not satisfy such a condition. Secondly, as large dimension and small amount of PA66 particles, interparticle distance may be larger than the critical value, which means critical stress field that satisfies yield condition of PP matrix can not be established under external force fields.³³ Finally, deformation of the PP matrix may be restricted due to interactions between PA66 and PP. During impact loading, number of crazes and shear zones can not be quickly initiated, and their propagation is inefficiently inhibited, thus NIIS is found to be greatly lowered. Equation (1) is termed the rule of mixtures,³⁴ which predicts the rigidity of filled composites,

$$M = \phi_1 M_1 + \phi_2 M_2 + \phi_3 M_3 \quad (1)$$

where M , M_1 , M_2 , and M_3 are TYS of the PP/PA66/C(6.25) sample, PP, PA66, and the compatibilizer, respectively; ϕ_1 , ϕ_2 , and ϕ_3 are volume fractions of PP, PA66, and the compatibilizer, respectively. This seems the TYS of PP/PA66/C(6.25) should be higher than that of the neat PP due to 18.7% PA66. However, the TYS of PP/PA66/C(6.25) is decreased by the incorporation of 6.25% compatibilizer as “soft” constituent and occurring of a relative slip when strain is relatively larger under tensile loading.

The PP/mPA66(6.25) composite has NIIS, FM, and TYS of 1.49, 1.16, and 0.99 times as those of the neat PP, respectively. Such enhanced mechanical properties balance is attained in the composite prepared by the “post-compatibilization” technique, which can be explained with the structural characteristics of profiled microfibrils, stronger adhesion, and moderate flexibility of the interface. It seems that the TYS of the PP/mPA66(6.25) composite should be higher than that of the neat PP for the efficient shear stress transfer from the matrix to the dispersed phase and the distinctness of the reinforcing effect by PA66 microfibrils. However, when strain is relatively large at the yield strength point, flexible interface may lead to very small relative slip, which is resulted from the discontinuous of microfibrils,³⁵ thus counteracting a part of reinforcing effects of the PA66 microfibrils. Therefore, the TYS of the PP/mPA66(6.25) composite is similar to that of the neat PP. Under bending load, as no relative slip may occur under the stronger adhesion of interface, reinforcement of microfibrils with high modulus dominates the FM. Undertaking an impact loading exercise, PA66 microfibrils can not only exhibit a distinct effect for the stronger adhesion but also inhibit number of crazes and shear zones from propagating as soon as they are initiated by stress concentration of the flexible interface.³² Furthermore, microfibrils pulled from the surrounding matrix is a probable destruction mechanism, which may be due to the length of microfibrils larger than the critical value and the higher strength. Hence, some significant plastic deformation of PP may occur.

Similar situations were also observed for the comprehensive mechanical properties of PP/mPA66(1.25) and PP/mPA66(3.75), and the fact is further substantiated and is consistent with the interpretation of the relationship between structure and mechanical properties.

CONCLUSIONS

The PP/mPA66 *in situ* microfibrillar composites prepared by using the “post-compatibilization” technique show different morphologies and mechanical properties compared with those of PP/PA66 blends. PA66-g-PP (and/or elastomers) graft copolymer formation in mPA66 was identified and the compatibilizer is unevenly dispersed with large domains in PA66. In PP/mPA66 composites, observations for the fractured surface illustrated that PP/mPA66 composites have structural characteristics of stronger adhesion and moderate flexibility of the interface. Enhanced compatibilization between the PA66 microfibrils with the PP matrix resulted in improved mechanical properties of the PP/mPA66 composites. With optimized composition, the PP/mPA66 composite has notched Izod impact strength, flexural modulus, and tensile yield stress of 1.49, 1.16, and 0.99 times as those of the neat PP, respectively. Such enhanced mechanical properties balance and improved interface adhesion were not found in the simply blended samples of PP/PA66 with or without the specially designed compatibilizer. Therefore, it can be concluded that the preparation technique plays an important role in the properties of *in situ* microfibrillar composites.

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