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Study on Properties of PP Resin Composites Modified by Aluminum Powder

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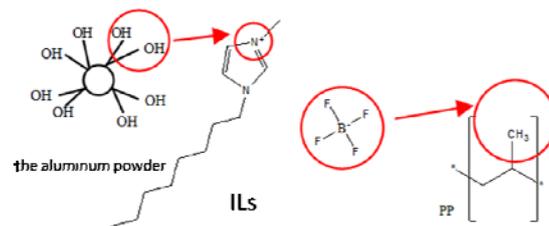
Abstract. The aluminum powder was used to blend with PP to impart a metallic luster to the PP matrix for achieving the effect of coating, and the ionic liquid (1-octyl-3-methylimidazolium tetrafluoroborate) was added into PP to improve the compatibility between aluminum powder and PP matrix. The results were analyzed by observing the different changes in thermal properties, crystallinity, rheological properties, mechanical properties, and sectional structures of the composite after filling different amount of aluminum powder. The results show that when adding 2phr aluminum powder, the crystallinity is high, the initial decomposition temperature of the composite is 295°C, the shear viscosity is 61.52Pa·s, the tensile strength is 23.15MPa, and the hardness is 55.6N/mm². In this ratio, the composite material has an outstanding performance.

1. Introduction

Polypropylene (PP) is a thermoplastic resin, usually seem like a colorless, translucent solid. The industrial polypropylene is widely used in automobile parts, pipes, and commodity due to its well-defined structure, high melting point, outstanding thermal resistance and mechanical properties. In recent years, there have been many researches on the properties of various aspects of PP, such as mechanical properties, flame retardancy, and so on [1-4]. There are also many methods for modifying PP, which are divided into physical modification and chemical modification. He et al. modified the PP/wood fiber composites with surface-treated nano-SiO₂ to improve the mechanical properties and hydrophobicity of the materials [5]. Liu Ting et al. summarized the chemical modification of different methods [6]. In order to improve the appearance of plastic products, the surface is often endowed an appointed colour and gloss by spraying. However, the method of spray is cumbersome, the paint will distribute unevenly that lead a low scrap rate and lead the environment pollution. Therefore, the spray-free process has become a hot topic in recent years [7, 8].

Ionic liquid (IL) has been applied in many domains due to its excellent properties, such as good stability, compatibilization and settability", especially for the modification of polymer materials [9-11]. The mechanism of ionic liquid used as a solubilizer between resin matrix and filled particles is as follows [12]: On the one hand, inorganic particles distributed more evenly in the matrix under the action of ionic liquids. On the other, ILs' special molecular structure contains both polar and non-polar groups, which act as the linker between organic molecules and inorganic particles; Moreover, the adding of ILs can increase the binding force through chemical reation among groups. The aluminum powder-filled PP resin blended and modified using the ionic liquid (1-octyl-3-methylimidazolium tetrafluoroborate). The molecular structure depicted in the following Scheme 1.





Scheme 1. The molecular structure of the ionic liquid.

The ionic liquid contains both polar and non-polar groups, which can respectively attract the methyl groups in the PP matrix and the hydroxyl groups on the surface of the aluminum powder, thereby combining the two phases and improving the interface environment between the PP and the aluminum particles, that boost the even-distribution and a better compatibility with the matrix. Then, the properties of the composite were studied.

2. Experiment

2.1 Materials

PP; aluminium powder: particle size of 7-8 μ m, purity 99.0%, silver white powder, Dongguan Ou Peide Chemical Pigment Co., Ltd.; ionic liquid (1-octyl-3-methylimidazolium tetrafluoroborate): relative molecular weight 282.13, purity 99%, Shanghai Chengjie Chemical Co.Ltd.

2.2 Preparation of composites

- (1). The PP pellets and aluminum powder were baked in a constant temperature drying oven at 100 °C for 12 hours to remove surface moisture and low molecular substances.
- (2). The PP particles and the aluminum powder were mixed at a mass ratio of 100:0, 100:1, 100:2, 100:3, and 100:4. Then adding 3 phr of ionic liquid and mixed well.
- (3). Five batches of the mixture in step 2 were separately mixed into an internal mixer for ten minutes, the temperature was set to 220 °C, and the rotation speed was set to 40-50 r/min. After 10 minutes of mixing, the material was discharged, cooled and crushed with a crusher, and then dry them at 100 °C for 4 hours.
- (4). The mixture was preformed by a vulcanizing press, the experimental temperature was set to 220 °C, and the load was applied at 3 MPa. After the pressure was applied for two minutes, the sheet was taken out to make samples.

2.3 Characterization

2.3.1. Scanning electron microscopy (SEM). The samples were frozen under liquid nitrogen for 24 hours and then brittle. After sections were sprayed with gold, all the samples were examined under a ZEISS EVO18 (Germany) scanning electron microscope (SEM) with an accelerating voltage of 20 kV.

2.3.2. Differential scanning calorimetry (DSC) measurement. Crystallinity of the material were measured by Diamond DSC differential scanning calorimetry tester from Perkin Elmer, USA. Scanning temperature is 30-190 °C at heating rate of 10°C/min.

2.3.3. Thermogravimetric Analyses (TGA). TGA were carried out at 10 °C /min under nitrogen flow (flow rate: 40ml/min) with a NETZSCH STA 449F3 (Germany) thermogravimetric analyzer. In each case, the samples (2-5mg) were heated from 50 °C to 800 °C at the rate of 20 °C /min.

2.3.4. Rheological characteristics analysis. The rheological properties of the samples were characterized by MLW-400B capillary rheometer from Changchun Intelligent Instrument Equipment Co.Ltd.

2.3.5. Mechanical Properties Test

Impact property. The impact strength test was carried out in accordance with GB/T1843-1996 using the JJ-2 type memory impact tester of Changchun Intelligent Instrument Co.Ltd.

Tensile property. The tensile strength test was carried out in accordance with GB1040-2006 using the WSM-5KN universal testing machine of Changchun Intelligent Instrument Equipment Co.Ltd.

Surface hardness. The hardness test was carried out according to the standard GB/T3398.1-2008 using a QYS-96B plastic ball hardness tester.

3. Result and Discussion

3.1 Microstructure of composite materials

Shown in figure 1 are the scanning electron microscope images of fracture surface with different contents aluminum powder. As shown in figure 1(a), the sample without aluminum powder is relatively smooth and indicates a completely brittle fracture. In contrast, it can be seen that the cross section of the samples after adding a certain amount of aluminum powder are irregularly rough. In general, an incomplete interface will directly affect the macroscopic performance of material, especially the reduction of mechanical properties. However, there are many factors affecting the interface structure. We can consider them mainly from the interface combination and the agglomeration of filled particles. From the picture, many holes left in the interface after the aluminum particles coarse fell off the section, the phenomenon suggesting a weak interfacial binding force between the particle and the pp matrix. When 1 phr of aluminum powder added, as the aluminum powder randomly dispersed in the resin matrix, the continuous structures of the resin matrix destroyed, resulting in irregular distribution of the cross-sectional structure. When 2 phr of aluminum powder added, the distribution of aluminum particles in the matrix was in a relatively uniform saturated state, and the stress concentration inside the matrix is small, which makes the section protrusions relatively smooth. When the aluminum powder fraction was increased to 3 phr, the aluminum powder particles were agglomerated due to supersaturation in the resin matrix to cause stress concentration, resulting in a very irregular scaly shape of the cross section. It will appear more pronounced stress concentration, resulting in a rougher profile after adding 4phr aluminum powder.

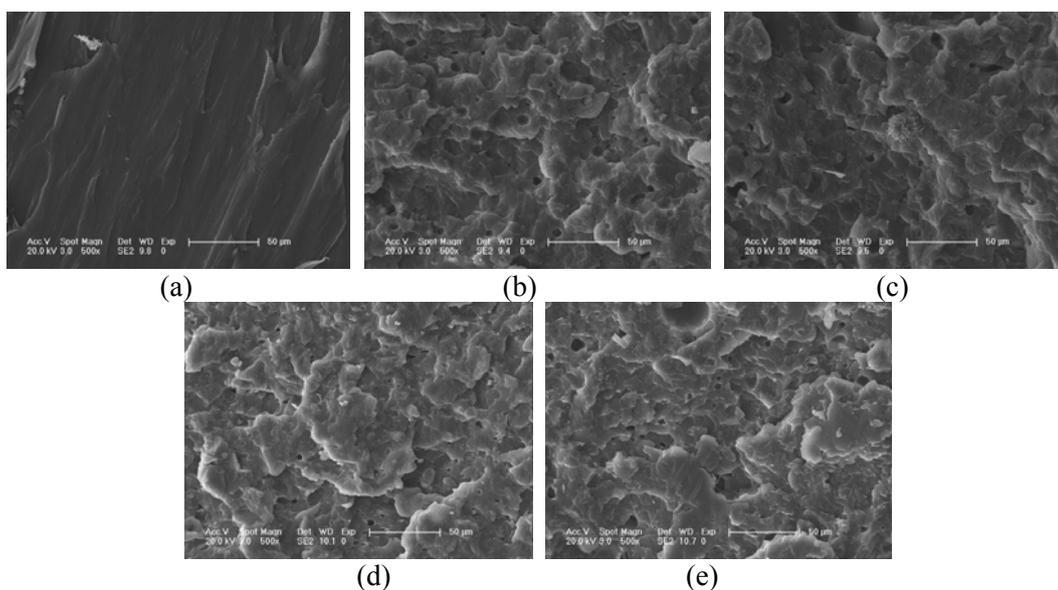


Figure 1. SEM images of composites fracture surfaces
(a) Al=0phr; (b) Al=1phr; (c) Al=2phr; (d) Al=3phr; (e) Al=4phr.

3.2 Thermal properties and crystallization properties

Figure 2(a) shows the DSC test results of composites with different aluminum powder contents. The peak area in the image can reflect the melting enthalpy (ΔH_f) of the polymer, and the crystallinity can be calculated by formula (1). ΔH_0 found to be 207J/g.

$$X_c = \frac{\Delta H_f}{\Delta H_0} \times 100\% \quad (1)$$

In the formula: X_c - the crystallinity (%) of the polymer, ΔH_f - the heat of fusion (J/g) of the polymer, ΔH_0 - the heat of fusion of the polymer at 100% crystallinity (J/g).

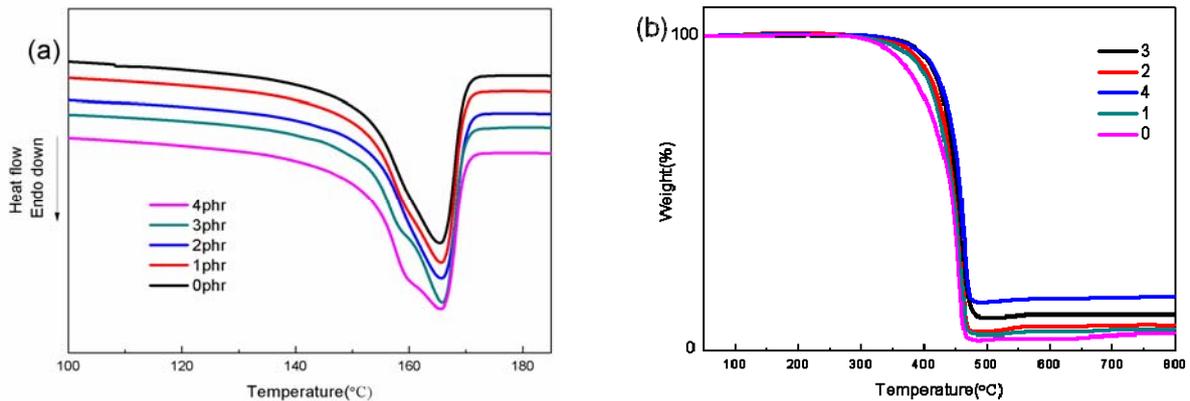


Figure 2. (a) Melting behavior of PP/Al/IL blends with different Al phr. (b) The TG curves of PP/Al/IL system.

The crystallinity of the filler shows in table 1. It can be seen from the table that the crystallinity of the composite first increases and then decreases. The addition of ionic liquid can destroy the entanglement effect between the macromolecules of the polypropylene, which causes a low crystallization of the composite. In addition, tiny aluminum particles act as heterogeneous nucleation centers, appropriate addition can promote crystallization. When 2 phr of aluminum powder added, the effect of heterogeneous nucleation was obvious, and the arrangement of the polypropylene macromolecules ordered as the uniform distribution of filling particles, two factors make the crystallinity of the composite improve. Once the aluminum powder exceeds 2 phr, the excess particles cause the entanglement effect of the polypropylene macromolecule to be inconspicuous. Moreover, if the matrix is broken, the crystallization will consume more energy, making it difficult to crystallize, so as the fraction of the aluminum powder increases, the crystallinity of the filler material begins to decrease.

Table 1. The crystallinity of samples.

phr of Al powder	0	1	2	3	4
ΔH_f (J/g)	93.09	85.46	88.78	85.54	78.32
X_c	44.97	41.28	42.89	41.32	37.84

Figure 2 (b) is the TG curve of the samples which reflects the thermal stability of the composite. As shown in the figure 2(b), when the aluminium powder content of the composite is gradually increased, the carbon residue rate is also increasing. The decomposition temperature of the samples (as shown in table 2) showed a tendency to increase after the addition of aluminum powder, which is related to the thermal conductivity of melt. On the one hand, the hindering action of filling particles can delay the depolymerization and decomposition of macromolecular chain. On the other, aluminum has good

thermal conductivity, which can make the melt heated evenly. Besides, when the aluminum powder content reaches 3-4 phr, the interface of aluminum particles contact each other, the heat conduction effect will be more obvious.

Table 2. Decomposition-temperature of samples.

phr of Al powder	0	1	2	3	4
Decomposition-temperature(°C)	312.7	275	295	281.4	279

3.3 Rheological analysis

Figure 3 shows the rheological properties of composites at a constant shear rate. It can be seen from the figure that the shear strength and shear viscosity of the composites decreased with the increase of aluminum powder content. The addition of ionic liquid improves the compatibility of the aluminum powder, and increasing the amount of aluminum powder added will lead to an increase in the thermal conductivity of the material [13], thereby lowering the viscosity of the melt. On the other hand, an increase in the aluminum powder content increases the collision probability between the aluminum particles, and the particle surface has a lubricating property, so that the shear strength of the composite material is lowered.

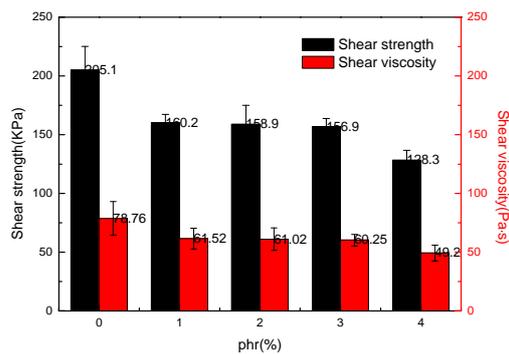


Figure 3. The result of shear strength and viscosity.

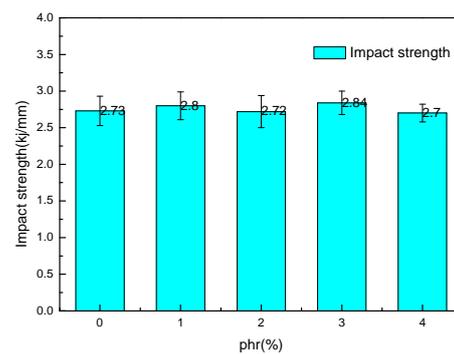


Figure 4. The impact strength of sample.

3.4 Mechanical properties analysis

Figure 4 shows the impact strength of different samples. Normally, the filling of inorganic particles will toughen the matrix [14] but the addition of aluminum powder destroys the continuous structure of the PP matrix and causes poor interface conditions, which reduces the mechanical properties of the composite. Since the aluminum particles are rigid particles, which will prevent cracks from propagating when the material was impacted [15], this is good for improving the impact strength of material. Therefore, the impact performance of the composite material does not change significantly based on correlation factor.

Figure 5 (a) shows the tensile strength of different patterns. The tensile properties of the samples after adding aluminum powder reduced to different extents compared with the pure component samples, but with the increase of the addition amount, the trend of increasing first and then decreasing was observed. This is the fact that the resin matrix blocked after the addition of the filler particles, forming a relatively continuous network structure. The mesh structure makes the tensile stress of the composite material relatively uniform and is not easy to cause stress concentration. Once the aluminum powder is excessive, the larger agglomeration formed will produce a large interface with poor affinity between the resins. When the tensile stress is applied, debonding occurs, so the tensile strength is decline.

Figure 5(b) shows the surface hardness of different patterns. As can be seen from the figure, the change in hardness is also a tendency to increase first and then decrease. There are two main factors

affecting the hardness of the filler material. Firstly, the addition of rigid particles increases the hardness of the material. Secondly, the filler particles destroy the crystallinity of the resin matrix and reduce the hardness. The amount of filler added in this experiment is relatively small, so the hardness is mainly dominated by the crystallinity of the material. The data in table 1 shows that the crystallinity of the material increases first and then decreases with the addition of aluminum powder, which is consistent with the matrix of hardness change.

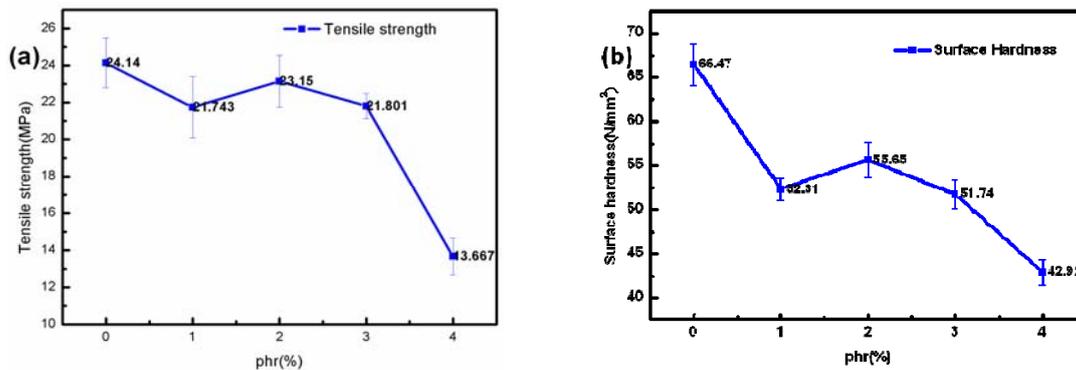


Figure 5. (a) The tensile strength of samples. (b) The surface hardness of samples.

4. Conclusions

This research investigated the variations in crystallization properties, thermal stability, rheological energy, and mechanical properties combined microstructure of the material after adding aluminum powder. The study shows that the addition of aluminum powder can improved the thermal properties of the material and change the crystallinity. Besides, the lubrication and thermal conductivity between aluminum particles reduce the viscosity of PP to a certain extent. The results of mechanical properties analysis show that the addition of aluminum powder in the PP matrix would reduce the tensile properties and hardness. When excess aluminum powder is added, there is a sharp decrease. That attributed to its damage to the matrix structure and poor interface environment. The unobvious change of impact property is the result of toughening effect of toughening effect of rigid particles and coarse interface environment

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