The toughening mechanism of rubber particles in polypropylene composite

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Abstract. Filling polypropylene materials with rubber particles can effectively increase the toughness of PP material and improve its cushioning properties. In this paper, we used the two kinds of method of the finite element analysis and experiment to study the rubber particles toughening mechanism, got the deformation process of particles when polypropylene material compressed and the yield stress of polypropylene after compression with particles filled or not.

1. Introduction
Polypropylene is commonly used as a functional material with good comprehensive mechanical properties, but its brittleness is big, which limit its application in engineering practice. So modifying the PP to improve its ability of absorbing energy is of great importance in engineering practice. There are many kinds of modification methods such as Blending modification, filling modification and composite modification, chemistry modification, surface modification. The widely used method among them is particles-filled [1-3]. Common filler particle is mainly divided into rigid filler particles and the flexible filler particles. The previous research shown that rigid filler particles like glass beads have no obvious effect on improving the performance of polymer [4]. But powdered acrylonitrile-butadiene rubber is an ideal filling material which often used in the study of polymer modification. It uses the excellent resin modifying agent and can be directly blended with a variety of resin, it also has good compatibility. Compared with block nitrile rubber, the advantage is that the small particle size and good dispersion when blended with resin and can be directly put in the extruder, making process continuous, automation, convenient, energy conservation and reducing the cost. Rubber particles as a filler particles can through own large elastic deformation and the interaction with substrate to improve material absorption ability, so we can achieve the purpose of increasing the toughness of pp matrix [5-7].
Studies show that using natural rubber and epdm rubber to toughen polypropylene respectively, with the increase of mass fraction, the impact strength and elongation at break are dramatically increased, but the tensile strength and young’s modulus decreases [8, 9]. Ravi ayyer researched the influence of rubber particle size and surface area for the structure and mechanical properties of pp matrix. AJ Kinloch used rubber to toughen epoxy resin, studied the micro structure and fracture of materials [10].

In the present article, we used powdered NBR to fill polypropylene matrix, compared to the matrix filled with glass beads of particles, combined with the finite element method and experimental methods to analyze the toughening mechanism of flexible particles.

2. Preparation and measurements

2.1. Materials
Polypropylene, Guangdong Maoming Petrochemical Factory, brand for T46F - H;
Hollow glass beads, Sinosteel Maanshan Mining Research Institute Co., LTD., types of H40;
Nitrile rubber powder, Jiangsu Jingjiang Guangsheng Pixels Materials Plant, models for GM50.

2.2. Experiment method

This experiment with micron grade nitrile rubber powder filled polypropylene, production of rubber powder/pp composites. Nitrile rubber powder and polypropylene blend with the quality ratio of 18:82. Hybrid system as a set of rubber powder and polypropylene, pure polypropylene system as the control group, two groups of materials respectively by the double screw extruder with mixing granule. Squeeze in turn set to a temperature of 205 °C, 215 °C, 215 °C, 215 °C, 215 °C, 205 °C. Speed is set to 250 r/min, melt pressure of 26 bar, melting temperature is 205 °C.

Mixing after all material system of particles with injection molding, injection molding processing of compressed sample size to 10 mm x 10 mm x 4 mm, made after the completion of the sample after cooling to finalize the design for subsequent experiments.

Normally, uniaxial compression tests were performed at room temperature by an electronic universal testing machine in accordance with GB T1041-2008. Measurement range of the machine is 10kN. The measurement accuracy is about 1%. The size of Specimens which were made for uniaxial compression tests were 10mm×10mm×4mm. The speed of crosshead was 5 mm / min [11].

The morphologies of fractured surfaces of the samples broken during impact measurement were observed through Scanning Electron Microscopy (SEM). The fractured surfaces of the samples were sputtered with gold or carbon (improve the electical conductivity) before observation by SEM [11].

2.3. Ansys method

In the process of finite element modeling, it is necessary to accurately describe the mechanical behavior of materials and establish the constitutive relation consistent with the experimental data. Powdered butadiene-acrylonitrile rubber chose the Mooney - Rivlin model that elastic materials often used in large deformation, modeling material parameters are shown in Table 1, polypropylene materials used the elastoplastic model and the elasticity parameters of the material are shown in Table 2. All the data obtained from compression experiment results.
Table 1. Modeling material parameters of Mooney-Rivlin model.

|    | C10  | C01  | D1   |
|----|------|------|------|
|    | 0.769| 0.012| 0.001|

Polypropylene’s plastic section parameters in the elastic-plastic material model need to set the yield stress and plastic strain. Due to the compression test results is engineering stress and strain curve, in the finite element simulation, we need to convert it into true stress and strain, conversion relations as follows:

\[
\varepsilon_t = \ln \left(1 + \varepsilon_n \right) \\
\sigma_t = \sigma_n \cdot (1 + \varepsilon_n)
\]

True stress and strain for finite element calculation [12, 13].

For accurately analyzing the distribution of particle and the surrounding stress in the deformation process, a single particle model is established first. This article selects the two-dimensional model, simplify the rubber powder into the spherical, model for the spherical particles embedded in the pp matrix of cuboid. The model size should be consistent with the actual material filling ratio and particle size. The actual quality of material particles and matrix than for 18:82, we can know the particle volume fraction in general by density function conversion. The processing method of interface between the filler particles and matrix is two phase idealized adhesive, namely interface nodes implement fully binding. In order to study the interaction of filler particles and their effects for yield and deformation of the matrix, this paper established a two-dimensional plane stress model of five particles.

And in order to study the shape of filler particles on the substrate and the influence on the result of the deformation, this paper established two two-dimensional models of filling particles in different shapes. Nitrile rubber as buffer energy absorption mechanism of the flexible and rigid particles, thus established the matrix particles and glass bead filled rubber particles (rigid particles) model is used in the comparison.

Table 2. The material parameters of PP.

| Material | Modulus of elasticity(MPa) | Poisson’s ratio |
|----------|---------------------------|----------------|
| PP       | 375                       | 0.43           |

3. Result

3.1. Deformation process of rubber particles with different shapes in the PP composite filled with rubber particles

To study the particle shape on the particles of different energy absorption mechanism, the influence of two irregular shape particles were taken under the condition of same compression simulation, get their compression stress nephogram, as shown in Figure 1, the particles began to deformation matrix are around the edge position is the largest tensile stress, the upper and lower edge position is one of the biggest compressive stress, similar to round particles. But uniform stress distribution around a circular
particles, and irregular shape of the particles at the edges and will produce a certain degree of stress concentration, but relatively little impact on the whole collective.

Figure 1. Deformation process of rubber particle in the PP composite filled with rubber particles (pentagon).
Table 3. The absorption of energy filled with different rubber particles.

| Different shapes | circle | Shape 1 | Shape 2 |
|------------------|--------|---------|---------|
| Absorption of energy | 33     | 32      | 31      |

3.2. Deformation process of rubber particle in the PP composite filled with rubber particles

The particles of two-dimensional plane stress model is presented in this paper, the rubber particles is set to round, pressure on the simulation process in the vertical direction to load, the stress cloud of the compression process. As shown in Figure 2, during the compression process, the left and right sides of the rubber particles produced by the tensile stress is greater than the rest of the matrix stress, and the closer distance of rubber particles produced by the stress, the greater the first stress that rubber particles to produce large deformation, has a tendency to like horizontal expansion. Rubber ball and has been a compressive stress near the loading direction, poisson is bigger, because the rubber particles under compression load deformation speed is greater than the pp matrix [14, 15]. Particles was crushed, as shown in Table 3, we find that in the event of large elastic deformation in the process of the absorbed energy, delayed the matrix of the yield process, so as to achieve the effect of buffer energy absorption. Contrast experiments of particle microstructure before and after compression, and finite element simulation results conform to the basic, rubber particles are compressed in a plane perpendicular to the direction of loading the elongated, matrix near the particles reach the yield, first occurred plastic deformation, the mechanical properties of weakened.

The compression process of rubber particles as shown in Figure 3, the rubber particles by spherical gradually became elongated elliptic, large deformation occurred. Rubber particles formed the yield zone, near the plastic deformation of the formation of yield with led matrix, and combined with the large deformation of rubber particles, and become the main cause of buffer endergonic.
Figure 2. Deformation process of rubber particle in the PP composite filled with rubber particles (circle).

Figure 3. Deformation process of rubber particles in the PP composite filled with rubber particles (five circles).

As shown in Figure 4, observation of scanning electron microscopy (SEM), sample before compression can clearly see the rubber particle and polypropylene matrix, after compression materials reach the yield limit, the rubber particle and matrix together elongated, deformation is severe, and in line with the results of finite element simulation.

Figure 4. Scanning electron microscopy (SEM) profile of the PP composite filled with rubber particles.
3.3. Effect of rubber particles on morphology and impact property of the PP composite including rubber particles

In PP matrix along the horizontal direction to take three nodes on ABC, get energy time plot as shown in Figure 5. Can be seen from the diagram, at the beginning of the curve levels did not absorb energy, over time, began to absorb energy, distance of rubber particles far point of energy absorption rate faster, that forced deformation of rubber particles absorb energy has effect on the substrate, forces acting on the matrix of smaller. Three points at the same time into the platform area, and then began to absorb energy. Beginning level relative only polypropylene matrix for a long time, the compression process at the beginning of the rubber particles absorb energy first, and the substrate to maintain current state.

![Figure 5. Curve of energy-time of the PP composite filled with rubber particles.](image)

It can also be seen from Figure 6 that the PP composite including BNRPs could absorb more energy than that of the PP under the same stress. It’s proved that filling of the rubber particles can effectively improve the toughness of PP matrix and its cushioning feature.

![Figure 6. Efficiency-stress curves for the studied materials: the PP composite including BNRPs and PP.](image)
4. Conclusions

Based on the finite element analysis and experimental results, the rubber particles filled polypropylene can through own deformation to absorb some of the impact energy, so as to improve the toughness of pp material, to achieve the effect of buffer energy absorption. When enough hours filling particles, the shape of the rubber particles on the energy absorption capacity is not big, the influence of the distribution of density can produce certain effect. Compared with glass beads filling particles, rubber particles mainly in the prophase work of polypropylene, and toughening effect than that of glass beads.

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