



of Achievements in Materials and Manufacturing Engineering

Tensile mechanical properties in PP/SEBS/Microballon composites under impact loadings

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Properties

<u>ABSTRACT</u>

Purpose: The mechanical properties of the syntactic polymer foams at the intermediate and high strain rates were not understood comprehensively. Then, this study characterizes the tensile mechanical properties of the polymer syntactic composites at high strain rates.

Design/methodology/approach: Eight kinds of syntactic foams and one neat PP/SEBS specimens are prepared at the same manufacturing process: 0, 2, 4, 8, 10, 20, 30, 40 and 50 volume percents of microballoons in the PP/SEBS blend matrix. Tensile tests are conducted at strain rates ranged from 0.3 to 100 s⁻¹. Apparent elastic modulus, yield stress and rupture strain are measured and the effects of microballoons on the mechanical properties are studied. In addition, the experimental results are compared with analytical model for closed-cell foam and the effects of the density of the PP/SEBS/microballoon composite on both apparent elastic modulus and yield stress are discussed. **Findings:** The apparent elastic moduli of PP/SEBS/microballoon syntactic composites follow the Gibson-Ashby law at the nominal strain rate of 100 s⁻¹. The yield stress of PP/SEBS/microballoon syntactic composites follow the simple rule of mixture at the relative densities larger than 0.9. The material ductility decreases drastically once the microballoons are blended in the matrix material.

Research limitations/implications: The influence of the local strain rate caused by the heterogeneous microstructure on the mechanical properties is to be further explored.

Practical implications: In the automobile applications, the thermoplastic polymer syntactic foams are believed to have many advantages because the usual commercial extruders or injection moulding machines are applicable for producing them, leading to the more light-weight polymeric components.

Originality/value: The present study investigates the effects of the strain rate and density on the tensile mechanical properties comprehensively in the polymer syntactic foams.

Keywords: Properties; Mechanical properties; Strain rate; Microballoon; Polypropylene

<u>1. Introduction</u>

Syntactic polymer foams are being used to replace the traditional polymer bulk components for weight reduction in the automobile applications. Syntactic polymer foams are composite materials consisting of spherical particles which are dispersed in a matrix polymer material. The spherical particles are usually called microballoons and microspheres. The most widely used syntactic foams are thermosetting polymers with glass microballoons as structural components [1]. For more detailed introduction to the syntactic foams, see Shutov [2], Lawrence and Pyrz [3], and Whinnery et al. [4]. In the automobile applications, the thermoplastic polymer syntactic foams are believed to have many

advantages because the usual commercial extruders or injection molding machines are applicable for producing them. The studies about mechanical properties of thermoplastic polymer syntactic foams are very few. According to the limited number of literatures describing their mechanical properties, Lawrence and Pyrz determined and compared the viscoplastic properties of polyethylene syntactic foams made from polyethylene and approximately 40 volume percent of polymer microballoons [3]. However, the mechanical properties at the intermediate and high strain rates were not understood comprehensively. The literature about the effect of strain rate on the mechanical properties of thermoplastic polymer syntactic foams is limited to the compressive mechanical properties [5-7]. Then, the effect of the strain rate on the tensile mechanical properties were characterized in syntactic foams of polypropylene (PP) blended with ethylene-propylene-rubber (EPR) and in-organic filler talc by the same author [8]. It appeared that the mechanical properties decreased drastically once the microballoons were blended at even low volume percentage in PP/EPR/talc blend. However, the previous study focused on the microballoon syntactic foams in only PP/EPR/talc blend matrix. Then, it is interesting to characterize the mechanical properties of the different type of polymer syntactic foam.

The present study attempted to characterize the tensile mechanical properties of the microballoon blend syntactic foams of PP-elastomer blend matrix at intermediate and high strain rate. The studied matrix material was PP blended with bimodal distributed styrene-ethylene-butadiene-styrene (SEBS) tri-block copolymer particles, which was developed in the previous study [9-11].

The syntactic foam in this study consists of elastically deformable microballoons in a PP/SEBS blend matrix. The microballoons expand during the manufacturing process such as injection molding because the liquid isobutene is inside the microballoon and it thermally expands during manufacturing process. Eight kinds of syntactic foams and one neat PP/SEBS specimens are prepared at same manufacturing process: 0, 2, 4, 8, 10, 20, 30, 40 and 50 volume percents of microballoons in the PP/SEBS blend matrix. Tensile tests are conducted at strain rates ranged from 0.3 to 100 s⁻¹. Apparent elastic modulus, yield stress and rupture strain are measured and the effects of microballoons on the mechanical properties are studied. In addition, fracture surfaces are observed with scanning electron microscopy (SEM) and the changes of fracture mode due to the volume percentage of microballoons are discussed. Finally, the experimental results are compared with analytical model for closed-cell foam and the effects of the density of the PP/SEBS/microballoon composite on both apparent elastic modulus and yield stress are discussed.

2. Specimen preparation and test procedures

2.1. Material

The matrix PP/SEBS matrix material was the PP (J-3003GV, Prime Polymer, Japan) blended with two types of SEBS (H1221 and H1062, Asahi Kasei Chemicals, Japan). The blend ratio of PP/SEBS blend was PP/SEBS (H1221)/SEBS (1062) = 70/15/15 vol %.

The more detail of the PP/SEBS blend can be found in the previous papers [9-11]. The microballoon was Expancel grade 950-120 (Akzo Nobel) whose nominal diameter was about 120 μ m. The material of microballoon shell is made of polymethyl methacrylate (PMMA). The volume ratios of blended mircoballoons are listed in Table 1. The microballoon was mixed with the pellet of matrix PP/SEBS blend at dry condition. Then, the mixed pellet was melted and injected into the molder. The injecting temperature was kept at 200°C during manufacturing process. The processing temperature and injecting pressure were kept constant for microballoon blend PP/SEBS and the neat PP/SEBS blend. Both materials were injection molded to the rectangular plate whose geometry was 150 mm x 150 mm x 3 mm. All tensile specimens were cut out of the plates such that the tensile direction was the same as the injection direction.

Table	1.
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Detail of PP/SEBS/microballoon composites

	Blend ratio (vol.%)	
Name	PP/SEB S	Microballoon
Neat	100	0
А	98	2
В	96	4
С	92	8
D	90	10
Е	80	20
F	70	30
G	60	40
Н	50	50

2.2.Test specimen and test apparatus

ASTM dumbbell shape (parallel portion width 4.8 mm) micro tensile test specimens were used for measuring the stress strain relationship (ASTM D1708). Figure 1 shows the geometry of the test specimen. The thickness of test specimen was 3 mm.

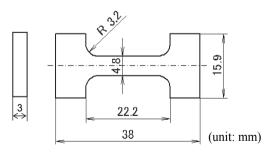


Fig. 1. Geometry of ASTM tensile test specimen

The present study used a servo-hydraulic high-speed impact test apparatus (Shimazu EHF U2H-20L: maximum tensile speed 15 m/s) to obtain mechanical characteristics under medium to high speed deformation. Strain rate was the nominal value calculated by the initial clamping distance of the test specimen which was 22.2 mm. The nominal strain rate ranged from 0.3 to 100 s⁻¹ for neat PP/SEBS blends while the nominal strain rates were 1, 10 and 100 s⁻¹ in the PP/SEBS/microballoon composites because of the limited number of syntactic foam specimens. After tensile tests, the fracture surfaces were observed with SEM (HITACHI S-4300SE/N) to identify the fracture mechanism at the nominal strain rates of 1 and 100 s⁻¹.

3. Experimental results

3.1. Tensile mechanical properties of neat PP/SEBS blends

Figure 2 shows the relationship between the apparent elastic modulus and the nominal tensile strain rate of the neat PP/SEBS blend. Tensile tests under each condition were conducted three times. The mean values calculated by three measurement data are plotted in Figure 2. It appeared that the apparent elastic modulus increased as the nominal strain rate went up in the neat PP/SEBS blend. It was found that the ductile-brittle transition would occur at the nominal strain rates between 10 and 100 s⁻¹.

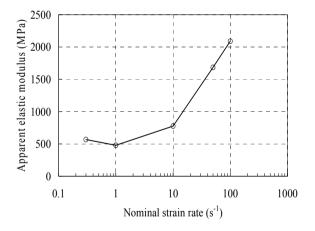


Fig. 2. Apparent elastic modulus vs. nominal strain rate of neat PP/SEBS blend

Figure 3 shows the yield stress plotted against the nominal strain rate of the neat PP/SEBS blend. The yielding of polymeric materials is very complex. The local damages such as polymer chain scission, micro crazing and so on could occur even in apparent elastic region on the stress-strain curve. Therefore, from a macroscopic point of view, the yield stress was defined as the maximum nominal stress in the stress-strain curve. In the same manner as Figure 2, the yield stress increased slightly as the nominal strain rate increased. However, the ductile-brittle transition was not observed in the case of yield stress at the nominal strain rates ranging from 0.3 to 100 s^{-1} .

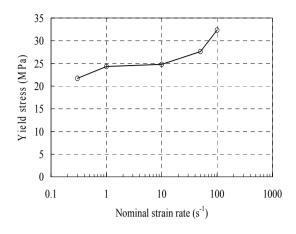


Fig. 3. Yield stress vs. nominal strain rate of neat PP/SEBS blend

3.2. Mechanical properties of PP/SEBS/microballoon composites

Figure 4 shows the typical nominal stress strain curves of PP/SEBS blend foams at the nominal strain rate of 10 s⁻¹. The flow stress decreased gradually when the blend ratio of the microballoons were larger. The interesting result here is that the rupture strain decreased drastically, once the microballoons were blended, regardless of the relative density.

Figure 5 shows the relationship between the apparent elastic modulus and the nominal tensile strain rate of various relative densities of PP/SEBS/microballoon composites. Tensile tests under each condition were conducted three times. The mean values calculated by three measurement data are plotted in Fig. 5. As shown clearly in Figure 5, the apparent elastic modulus increased as the nominal strain rate went up. Especially, at the nominal strain rate of 100 s⁻¹, the apparent elastic modulus increased drastically. This is because the ductile-brittle transition of the matrix PP/SEBS blend would occur at the strain rate between 10 and 100 s⁻¹ as inferred in Figure 2. It appeared that this ductile-brittle transition did not change among all relative densities, which indicated that the ductile-brittle transition of the syntactic composites did not depend on the relative density. Note, that the ductile-brittle transition was not dependent on the porous shape of polymer syntactic foams, reported by the same authors [12]. Based on the above discussion, it is considered that the strain rate dependency of the elastic modulus in the PP/SEBS/microballoon syntactic composites was dependent on the matrix material and it did not depend on the relative density of the syntactic composites.

In this study, the microballoon blended PP/SEBS had three phases which were pores, microballoon's shell and matrix (PP/SEBS blend). Thus, it is necessary to investigate the effect of the stiffness of the microballoon's shell on the macroscopic stiffness. When it is assumed that all blended microballoon is completely spherical shape, it is estimated that the volume percentage of the microballoon shell is only about 0.14 % at the 50 vol % of microballoons in the matrix. Note that the elastic modulus of microballoon shell was approximated as 2 GPa.

Based on the mixing rule, the effect of the elastic modulus of microballoon shell on the macroscopic elastic modulus in the current syntactic foams is negligible. Thus, it can be assumed that the microballoon blended PP/SEBS should consist of the pores and the matrix for simple calculation in this study.

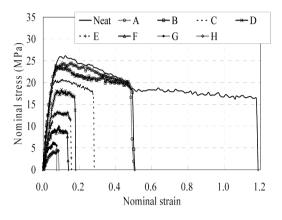


Fig. 4. Typical stress-strain curves in various PP/SEBS/microballoon composites at strain rate of 10 s⁻¹

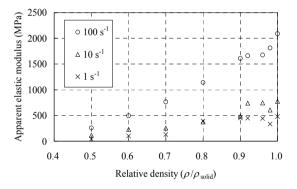


Fig. 5. Apparent elastic modulus vs. nominal strain rates in all PP/SEBS/microballoon syntactic composites

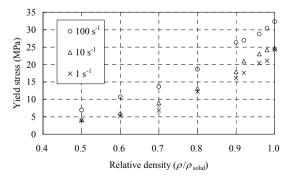


Fig. 6. Yield stress vs. nominal strain rates in all PP/SEBS/ microballoon syntactic composites

Figure 6 shows the yield stress plotted against the nominal strain rate in all the PP/SEBS/microballoon syntactic composites. In the same manner as Figure 5, the yield stress increased slightly as the nominal strain rate increased. However, the clear transition of the yield stress was not observed, indicating that the ductile-brittle transition was not observed in the case of yield stress at the nominal strain rates ranging from 1 to 100 s⁻¹. The similar trend was also obtained in the neat PP/SEBS blend as shown in Fig. 3. It is considered that the strain rate dependency of the yield stress was independent of the relative density in the PP/SEBS/microballoon syntactic composites. The strong factor for that would be the viscoplastic property of the matrix PP/SEBS blend.

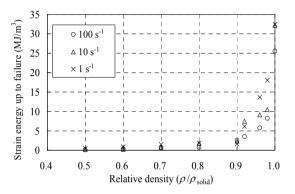


Fig. 7. Strain energy up to failure vs. nominal strain rates in all PP/SEBS/microballoon syntactic composites

Figure 7 shows the strain energy up to failure plotted against the nominal strain rate of PP/SEBS/microballoon syntactic composites. It appeared that the strain energy up to failure decreased drastically, once the microballoon was blended in the PP/SEBS blend matrix. In addition, the strain energy up to failure decreased as the nominal strain rate increased. It is considered that the fracture mechanisms would shift from ductile mode to brittle mode, leading to the small strain energy up to failure of all the syntactic composites at the high strain rate. This will be validated in the SEM fractographic analysis.

3.3. Fractographic analysis

To better understand the fracture mechanism of the present PP/SEBS syntactic foams under the low and high strain rates, the fracture surface of each tensile specimen was examined with SEM. Some of the SEM pictures are selectively presented in this paper. Figures 8 and 9 show the fracture surfaces of all the PP/SEBS syntactic foams at the nominal strain rates of 1 and 100 s⁻¹, respectively.

These SEM pictures were low magnified pictures so that several microballoons could be captured in one picture. As shown clearly, the PP/SEBS matrix between microballoons was highly elongated, leading to the ductile failure mechanism at the nominal strain rate of 1 s⁻¹. It appeared that the internal necking occurred at the PP/SEBS matrix between microballoons, where the debonding was observed

at the interface between the microballoons' shell and the matrix at the strain rate of 1 s⁻¹. On the contrary, the smooth fracture surfaces were obtained at the nominal strain rate of 100 s⁻¹ in all the relative densities. It is considered that the microballoons would work as the crack initiation site at the high strain rate in all the PP/SEBS/microballoon syntactic composites. At the low strain rate, the molecules can begin to untangle and relax, leading to the ductile fracture mechanism. However, the high strain rate loading does not give enough time for molecules to untangle and relax, leading to the brittle fracture. This microstructural analysis of ductile-brittle transition was analyzed by the molecular dynamic simulation in the lamellae model of PP [13]. This ductile brittle transition can be

found in the apparent elastic modulus as shown in Figure 5, where the apparent elastic moduli increased drastically at the strain rates between 10 and 100 s⁻¹. Based on the relationship between the apparent elastic modulus and the strain rate of the neat PP/SEBS blend as shown in Figure 2, it is expected that the ductile brittle transition nominal strain rate can range between 10 and 100 s⁻¹ in the present neat PP/SEBS blend. This indicates that the ductilebrittle transition of the syntactic composites was kept the same as that of the neat PP/SEBS blend. Therefore, it is considered that the ductile-brittle transition of the syntactic composites was dependent of the matrix material's property.

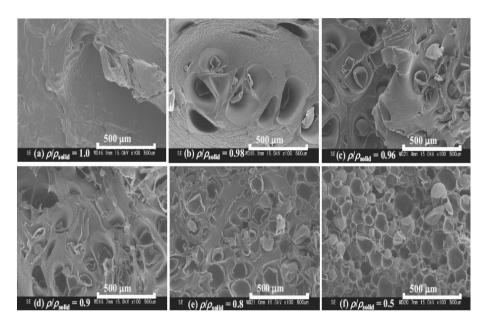


Fig. 8. Fractographs at the nominal strain rate of 1 s⁻¹

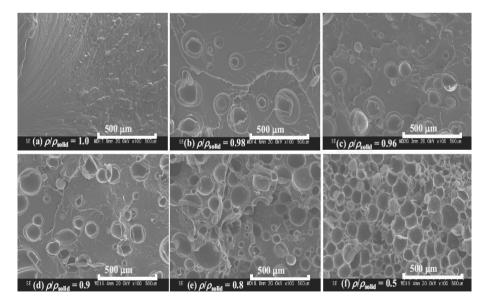


Fig. 9. Fractographs at the nominal strain rate of 100 s⁻¹

3.4. Effect of density on elastic modulus and yield stress

Currently, the quite popular theoretical model used to predict the modulus of closed-cell polymer foams is developed by Gibson-Ashby [14], which is generally related to its density by the formula;

$$E_f = \left(\rho_f / \rho_{solid}\right)^2 E_{solid} \tag{1}$$

where E_f is the modulus of the polymer foam and E_{solid} is the modulus of the matrix material. ρ_f and ρ_{solid} are the density of polymer foam and corresponding solid matrix, respectively.

The experimental results of the apparent elastic modulus of PP/SEBS/microballoon syntactic composite is plotted together with the theoretically predicted valued from Eq. (1), as shown in Figure 10. In this plot, it was assumed that the elastic moduli of the matrix (E_{solid}) in the foams were the same as that of the neat PP/SEBS blend. As shown clearly in Figs. 10, the apparent elastic moduli were close to the theoretical values at the strain rate of 100 s⁻¹. However, the apparent elastic moduli were relatively smaller than the theoretical values at the strain rates of 1 and 10 s⁻¹ At the strain rates of 1 and 10 s^{-1} , the difference between the experimental results and the theoretical ones was smaller at the relative densities more than 0.9. As shown in Figs. 8 and 9, the microballoons were separately located in the syntactic foams whose relative densities were larger than 0.9. On the contrary, it was found that the coalescence of the microballoons were observed in the syntactic foams of the relative densities smaller than 0.8, leading to the smaller elastic modulus than the theoretical values.

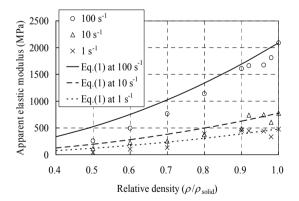


Fig. 10. Apparent elastic modulus vs. relative density with theoretical elastic modulus

In the case of the yield stress, the simple rule of mixtures gives the formula [15];

$$\sigma_{yf} = \left(\rho_f / \rho_{solid}\right) \sigma_{ysolid} \tag{2}$$

where σ_{yy} is the yield stress of the polymer foam, and σ_{ysolid} is the yield stress of the matrix material, respectively. Figure 11 shows the yield stresses plotted against the nominal strain rate in the PP/SEBS/microballoon syntacitc composites. As shown clearly in Figure 11, the yield stresses were smaller than the theoretical values calculated by Equation 2 at the relative densities smaller than 0.8 at all the strain rates in PP/SEBS foams. Therefore, it is considered that the yield stress of the syntactic composites would be strongly dependent on the microstructure of the matrix materials. Moreover, the simple mixing rule can predict the yield stress at the relative densities larger than 0.9 in the PP/SEBS/microballoon syntactic composites.

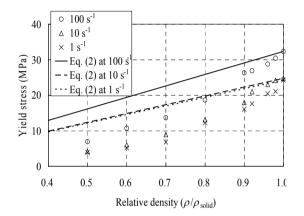


Fig. 11. Yield stress vs. relative density with theoretical yield stress

4.Conclusions

The tensile mechanical properties of the PP/SEBS/microballoon syntactic composites were characterized at the nominal strain rates of 1, 10 and 100 s⁻¹. The followings are the conclusions of the present study:

- The apparent elastic moduli of PP/SEBS/microballoon syntactic composites follow the Gibson-Ashby law at the nominal strain rate of 100 s⁻¹, which indicates that the microstructural deformation mechanisms would be correlated well with the Gibson-Ashby model when the matrix materials are the glass phase.
- The yield stress of PP/SEBS/microballoon syntactic composites follow the simple rule of mixture at the relative densities larger than 0.9.
- The material ductility decreases drastically once the microballoons are blended in the matrix material.

The polymer based syntactic composites have an advantage in light weight, low cost and good productivity for the applications not only in automobiles but also in any other field, such as airplane, mobile computers, and packaging. Especially, the relationships between the microscopic and macroscopic mechanical properties should be quantified more accurately in the future works.

Properties

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