

Relationship of Mechanical Properties between Neat PP/SEBS Blends and Syntactic PP/SEBS Foams with Polymer Microballoons[†]

by

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The tensile mechanical properties of PP/SEBS blend syntactic foams with the relative densities from 0.5 to 1.0 are characterized at the nominal strain rates of 1, 10 and 100 s⁻¹. Moreover, the effects of the mechanical properties of the matrix materials on the macroscopic mechanical properties are investigated by comparing the experimental results of two types of the PP/SEBS syntactic foams. The apparent elastic moduli of the syntactic foams are similar without depending on the matrix's elastic moduli once the small content of the microballoons is blended. Their material ductility decreases drastically once the microballoons are blended in the matrix materials regardless of the type of the matrix. The strain rate dependency of the elastic modulus in the syntactic foams is dependent on that of the matrix material. In addition, the drastic increase of the elastic moduli in the syntactic foams is delayed by adding the polymer microballoons.

Key words : *Polymer, Tensile mechanical properties, Strain rate, Elastic modulus, Yield stress, Closed cell, Syntactic foams, Microballoon, Polypropylene, Styrene ethylene butadiene styrene*

1 Introduction

Syntactic polymer blend foams have long been considered as economical materials for many applications due to their wide variety of advantages, such as light weight, energy absorbing performance, thermal and acoustic insulation and good aging.^{1)~6)} Syntactic polymer foams consist of pores and a matrix polymer material. The spherical pores are usually made by mixing microballoons or foaming gases. For more detailed introduction to the syntactic foams, see Shutov,⁷⁾ Lawrence and Pyrz⁸⁾ and Whinnery et al.⁹⁾

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 Pyrz⁸⁾ determined and compared the viscoplastic properties of polyethylene syntactic foams made from polyethylene and approximately 40 volume percent of polymer microballoons. 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.^{10)~12)} 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 inorganic filler talc by the same author.¹³⁾ It appeared that the mechanical properties decreased drastically once the microballoons were blended at even low volume percentage in the 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 types of PP blend syntactic foams, which enables us to investigate the effects of the mechanical properties of the matrix material on the macroscopic mechanical properties of the syntactic foams.

Then, the present study attempted to investigate the effects of the tensile mechanical properties of two different PP/styrene-ethylene-butadiene-styrene (SEBS) blends matrix on the macroscopic tensile mechanical properties in PP/SEBS blend syntactic foams with polymer microballoons. In the previous studies,^{14)~16)} it was found that the small-size SEBS particles located in the fibrils were highly elongated to the tensile loading direction, enhancing the ductility of the neat PP/SEBS blend. On the contrary, PP blend with the large-size SEBS particles showed the crazing without the large plastic deformation of PP matrix, leading to less ductility than the PP blend with the small-size SEBS particles.

In this study, the experimental results are compared with analytical model for closed-cell foam and the effects of the density and matrix materials on both apparent elastic modulus and yield stress are discussed.

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2 Experiments

2.1 Materials

The syntactic foams in this study consist of elastically deformable microballoons in PP/SEBS blends. 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 specimen were prepared at same manufacturing process: 0, 2, 4, 8, 10, 20, 30, 40 and 50 volume percentages of microballoons in each PP/SEBS blend matrix. The PP/SEBS matrix materials were the PP (J-3003GV, Prime Polymer, Japan) blended with two types of SEBS (SEBS A: H1221 and SEBS B: H1062, Asahi Kasei Chemicals, Japan). The hardness (JIS K6253) of SEBS A was smaller than that of SEBS B, as listed in Table 1 with more detailed descriptions. The particle size distribution of SEBS A ranged from 80 to 140 nm and that of SEBS B ranged from 200 to 250 nm.^{14)~16)} The blend ratios of PP/SEBS blends were PP/SEBS A = 70/30 vol % and PP/SEBS B = 70/30 vol %, respectively. The more detailed descriptions of the PP/SEBS blends such as the material preparations and the morphologies can be found in the previous papers.^{14)~16)} The microballoon was Expancel grade 950 ~ 120 (Akzo Nobel) whose average diameter was about 120 μm . The material of microballoon's shell was made of polymethyl methacrylate (PMMA). The average thickness of the microballoon's shell was about 0.1 μm according to the technical data given by the manufacturer. The volume ratios of blended microballoons are listed in Table 2. 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 mold. The temperature of the injection nozzle was

kept at 200°C. The injection pressure was 75 MPa. The mold temperature was kept at 40°C. The processing temperature and injecting pressure were kept the same for the microballoon blend PP/SEBS and the neat PP/SEBS blend. Both materials were injection molded to the rectangular plate whose geometry was 150 mm \times 150 mm \times 3 mm. All tensile specimens were cut out of the plates such that the tensile direction was the same as the injection direction.

2.2 Tensile Tests

ASTM dumbbell shape (parallel portion width 4.8 mm) micro tensile test specimens were used for measuring the stress strain relationship (ASTM D1708). Fig. 1 shows the geometry of the test specimen. The width and the thickness of the test specimens were measured before tensile testing. The measured width ranged from 4.7 to 4.8 mm and the thickness variation ranged from 2.9 to 3.0 mm. Based on the measured area of the cross section, the obtained load was converted to the nominal stress in each specimen. The gauge length was kept as 22.2 mm for calculating the nominal strain. This study used a servo-hydraulic high-speed impact test apparatus (Shimazu EHF U2H-20L: maximum tensile speed 15 m/s) to obtain the mechanical characteristics under medium to high speed deformation. The 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 the neat PP/SEBS blends while the nominal strain rates were 1, 10 and 100 s^{-1} in the PP/SEBS syntactic foams because of the limited number of syntactic foam specimens. Apparent elastic modulus, yield stress and strain energy up to failure were measured and the effects of microballoons on the mechanical properties were studied. The apparent elastic modulus was defined as the slope of the stress-strain curve at the nominal strains between 1 and 2 %. 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 the 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. Tensile tests

Table 1 Detail of SEBS A and SEBS B.

	H1221 (SEBS A)	H1062 (SEBS B)
MFR (g/10 min at 230 °C)	4.5	4.5
Hardness (JIS K 6253)	42	67
Weight ratio of styrene/ethylene-butylene (wt%)	12/88	18/82

Table 2 Detail of PP/SEBS foams with microballoons.

Name	Blend ratio (vol. %)	
	PP/SEBS A or B	Microballoon
Neat SEBS A or B	100	0
A-SEBS A or B	98	2
B-SEBS A or B	96	4
C-SEBS A or B	92	8
D-SEBS A or B	90	10
E-SEBS A or B	80	20
F-SEBS A or B	70	30
G-SEBS A or B	60	40
H-SEBS A or B	50	50

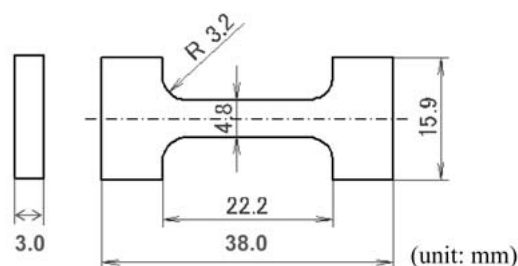


Fig. 1 Geometry of ASTM tensile test specimen.

under each condition were conducted three times. The average values calculated by three measurement data were plotted in this study. After tensile tests, the fracture surfaces were observed with the Scanning Electron Microscopy (SEM : HITACHI S-4300SE/N) to identify the fracture mechanism at the nominal strain rates of 1 and 100 s^{-1} . Moreover, the mechanical properties were compared between the two different types of PP/SEBS syntactic foams in order to investigate the effects of the mechanical properties of the matrix materials on the macroscopic mechanical properties of PP blend syntactic foams at intermediate and high strain rates.

3 Results and Discussions

3.1 Tensile Mechanical Properties of Neat PP/SEBS Blends

Fig. 2 shows the relationship between the apparent elastic modulus and the nominal tensile strain rate of the neat PP/SEBS blends. It appeared that the apparent elastic modulus increased as the nominal strain rate went up in both neat PP/SEBS A and PP/SEBS B blends. The apparent elastic modulus was larger in PP/SEBS B blend than in PP/SEBS A blend at the whole range of nominal strain rates in this study. It was observed that the apparent elastic moduli of both neat PP/SEBS blends increased drastically at the nominal strain rates between 10 and 100 s^{-1} . It is considered that these strain rate dependency of the elastic modulus would be similar phenomenon to the rubber-glass transition. The more detailed validation for that needs to be further explored. The microstructural analysis of this strain rate dependency of the apparent elastic modulus was conducted by the molecular dynamics simulation in the lamellae model of PP.¹⁸⁾

The yield stress plotted against the nominal strain rate of both neat PP/SEBS A and PP/SEBS B blends is shown in Fig. 3. It appeared that the yield stress increased slightly as the nominal strain rate increased. However, the drastic increase was not observed in the case of yield stress at the nominal strain rates ranging from 0.3 to 100 s^{-1} . In addition, it was found that the yield stress was

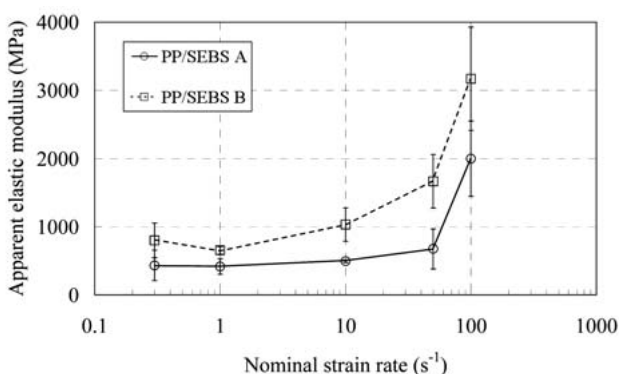


Fig. 2 Apparent elastic modulus vs. nominal strain rate of neat PP/SEBS A and PP/SEBS B blends.

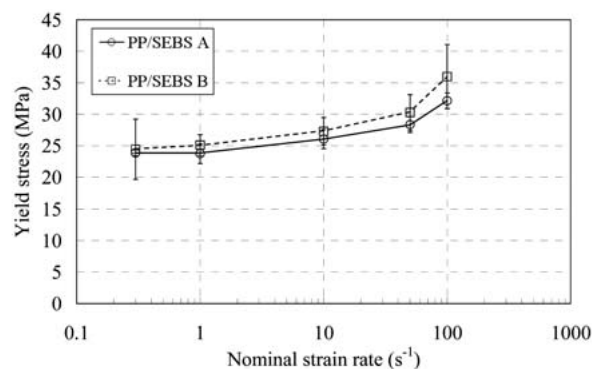


Fig. 3 Yield stress vs. nominal strain rate of neat PP/SEBS A and PP/SEBS B blends.

slightly larger in the neat PP/SEBS B blend than the neat PP/SEBS A blend at the whole range of the strain rates.

Fig. 4 shows the strain energy up to failure of the neat PP/SEBS A and B blends. As the strain rate increased, the strain energy up to failure decreased gradually in both neat PP/SEBS blends. The material ductility was larger in PP/SEBS A blend than PP/SEBS B blend at the whole range of the strain rates in this study. The ductile-brittle transition was not observed at the nominal strain rates ranged in this study. As shown in the section 3.3, the ductile fracture surface was obtained at the nominal strain rate of 100 s^{-1} in both neat PP/SEBS blends.

3.2 Tensile Mechanical Properties of PP/SEBS Syntactic Foams

The typical nominal stress strain curves of PP/SEBS A and PP/SEBS B syntactic foams at the nominal strain rate of 10 s^{-1} are shown in Fig. 5. The flow stresses and rupture strains decreased drastically once the microballoons were blended in the materials, regardless of the matrix materials (neat PP/SEBS A or neat PP/SEBS B). The PP/SEBS A syntactic foams showed the larger stress reduction than the PP/SEBS B syntactic foams after yielding. This tendency was similar to that of the neat PP/SEBS blends. It is considered that the same micro damage processes in the matrix would occur in the PP/SEBS syntactic foams as the neat PP/SEBS blends.

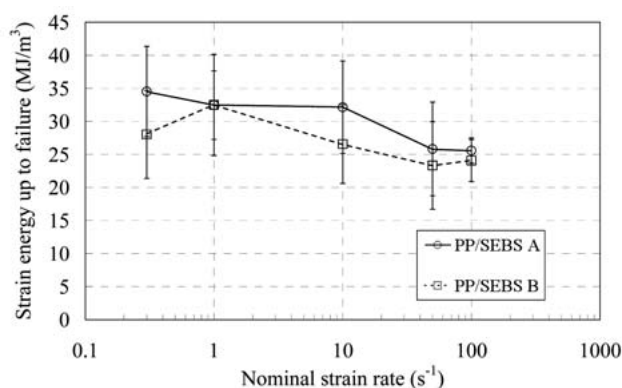


Fig. 4 Strain energy up to failure vs. nominal strain rate of neat PP/SEBS A and PP/SEBS B blends.

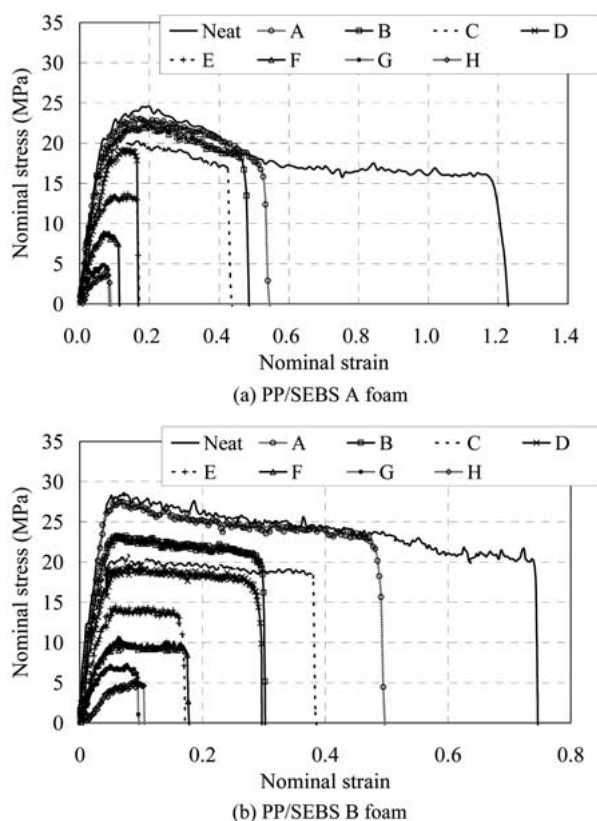


Fig. 5 Typical stress strain curves at the nominal strain rate of 10 s^{-1} .

Fig. 6 shows the relationship between the apparent elastic modulus and the nominal tensile strain rates of 1, 10 and 100 s^{-1} in various relative densities. The average values calculated by three measurement data are plotted in Fig. 6. The variations of the apparent elastic modulus, the yield stress and the strain energy up to failure were within the similar ranges to those of the neat PP/SEBS blends which are shown in Figs. 2, 3 and 4. Then, the average values of the apparent elastic modulus, the yield stress and the strain energy up to failure calculated by three measurement data are plotted in the PP/SEBS syntactic foams. As shown clearly in Fig. 6, the apparent elastic modulus increased as the nominal strain rate went up in all the PP/SEBS syntactic foams. Especially, at the nominal strain rate of 100 s^{-1} , the apparent elastic modulus increased drastically. It is considered that the similar phenomenon to the rubber-glass transition of the matrix PP/SEBS A and PP/SEBS B blends might occur at the strain rate between 10 and 100 s^{-1} . Moreover, it appeared that the drastic increase of the elastic modulus was delayed with the increasing of the microballoons. Note, that the transition from the rubber to the glass phases was not dependent on the porous shape of polymer syntactic foams, reported by the same author.¹⁷⁾ Based on the above discussion, it is considered that the strain rate dependency of the apparent elastic modulus in the PP/SEBS syntactic foams was dependent on that of the matrix

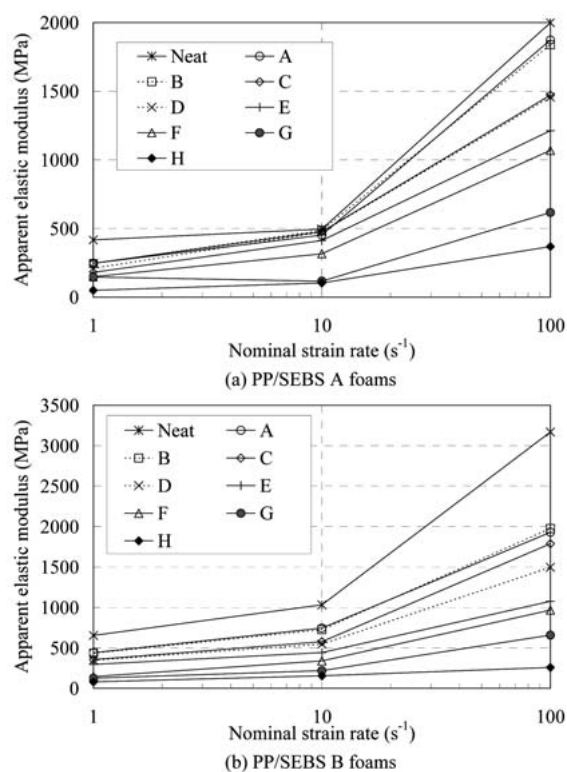


Fig. 6 Apparent elastic modulus vs. nominal strain rate.

material and it did not depend on the relative density of the syntactic foams.

Fig. 7 shows the yield stress plotted against the nominal strain rate in various relative densities. On the contrary to Fig. 6, the yield stress increased slightly as the nominal strain rate increased in both PP/SEBS A and PP/SEBS B foams. The drastic increase of the yield stress was not

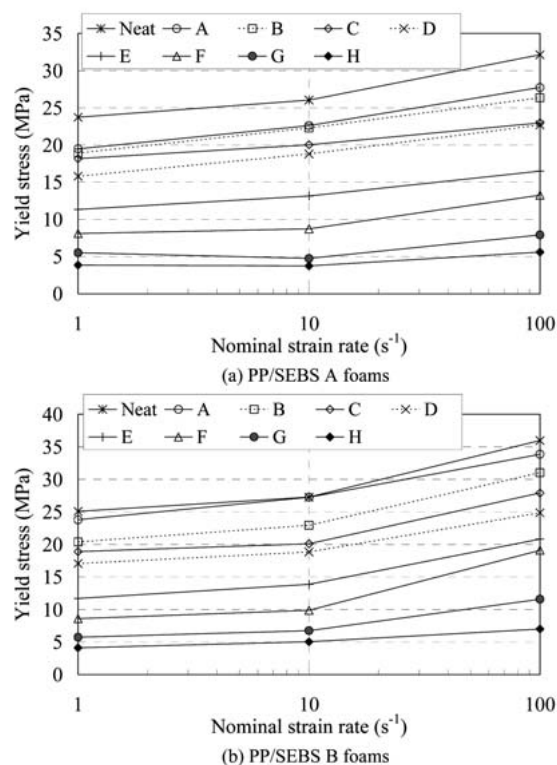


Fig. 7 Yield stress vs. nominal strain rate.

observed at the nominal strain rate ranging from 1 to 100 s^{-1} . The similar trends were also obtained in the neat PP/SEBS A and PP/SEBS B blends as shown in Fig. 3. Thus, it is considered that the strain rate dependency of the yield stress was independent of the relative density in PP/SEBS syntactic foams. The strong factor for that would be the viscoplastic property of the matrix PP/SEBS blends.

Fig. 8 shows the strain energy up to failure plotted against the nominal strain rate. The strain energy up to failure decreased as the nominal strain rate increased in both PP/SEBS A and PP/SEBS B foams. 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 foams at the high strain

rate. This will be validated in the SEM observations of the fracture surfaces. In addition, the absorbed energy decreased drastically with the decreased relative densities in both PP/SEBS foams. For instance, the strain energy up to failure decreased by about 80 % of the neat PP/SEBS blends at the relative density of 0.9 in both PP/SEBS syntactic foams. This is the ductile-brittle transition caused by adding the microballoons.

3.3 Fractography

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 specimen was examined with SEM. The typical SEM pictures are presented in this paper. Figs. 9 and 10 show the fracture surfaces of PP/SEBS A and PP/SEBS B foams at the nominal strain rates of 1 and 100 s^{-1} , respectively. There were microaloons, cavities, and elongated matrix and microballoons on the fracture surfaces obtained at the nominal strain rate of 1 s^{-1} as shown in Figs. 9 (a) and 10 (a). Moreover, the de-bonding between the microballoons and the matrix were clearly observed as shown in Figs. 9 (a-v) and 10 (a-v), for example. The microballoons consisted of the hollow shells as indicated by the white-color arrows in Figs. 9 (a-iii) and 9 (b-iii). It appeared that the matrix was highly elongated, leading to the ductile fracture mode in the relative densities from 0.8 to 1.0 at the nominal strain rate of 1 s^{-1} . At the low strain rate, the molecules of the matrix materials can begin to untangle and relax, leading to the ductile fracture mode.

On the contrary, the high strain rate loading does not give enough time for molecules to untangle and relax, leading to the brittle fracture mode. In the present materials, fibrils and matrix elongation were observed partially on the PP/SEBS matrix at the relative densities larger than 0.9 in both PP/SEBS syntactic foams at the nominal strain rate of 100 s^{-1} as shown in Figs. 9 (b) and 10 (b). However, at the relative density of 0.8, the relatively

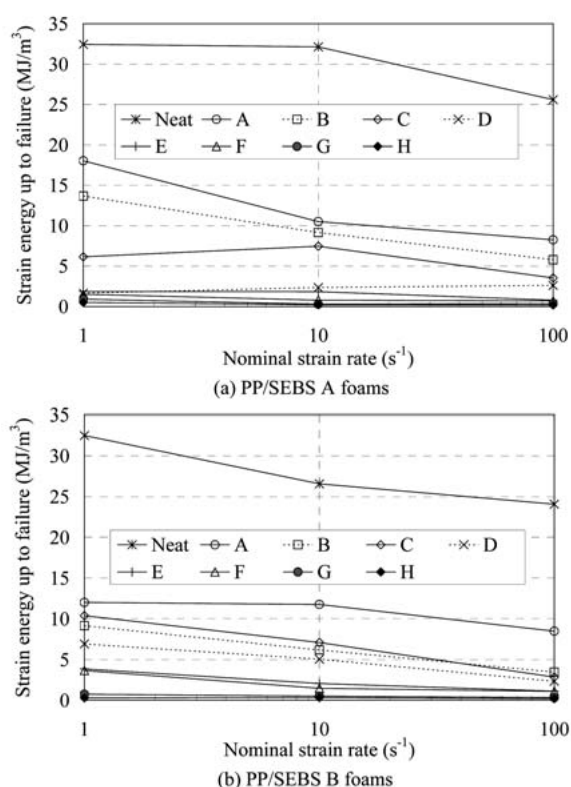


Fig. 8 Strain energy up to failure vs. nominal strain rate.

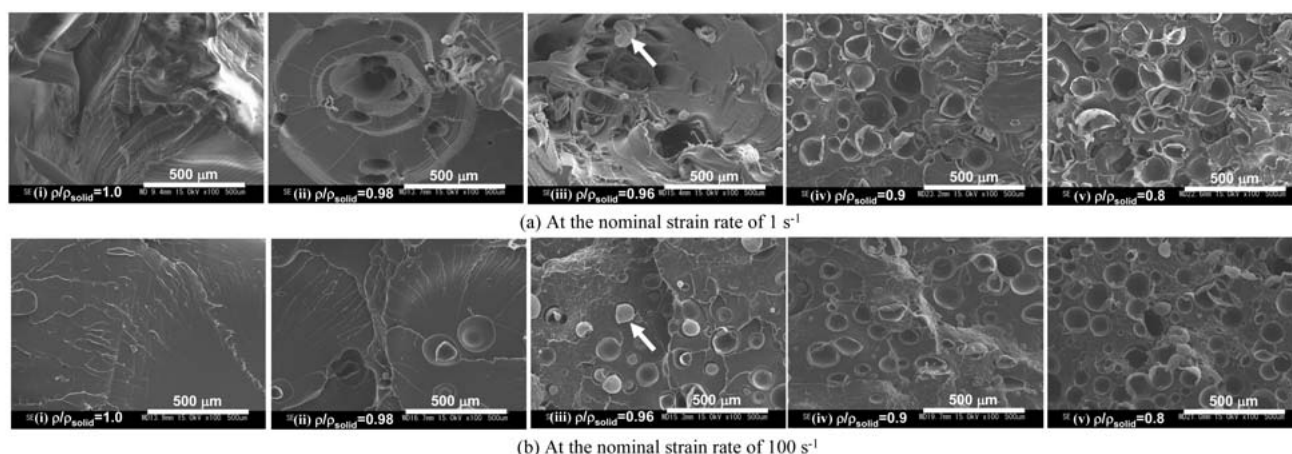


Fig. 9 SEM pictures of fracture surfaces of neat PP/SEBS A blend and PP/SEBS A syntactic foams.

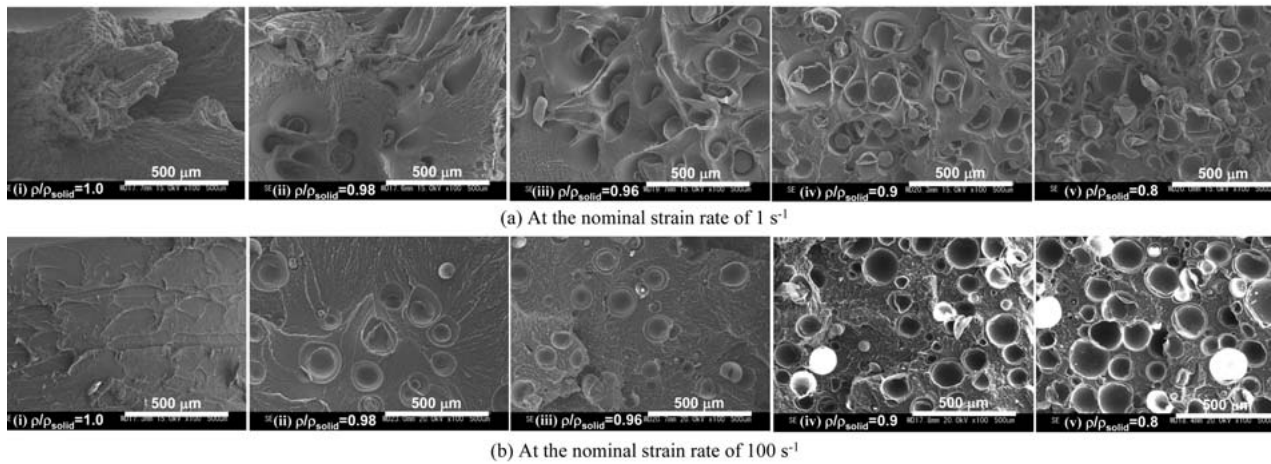


Fig. 10 SEM pictures of fracture surfaces of neat PP/SEBS B blend and PP/SEBS B syntactic foams.

smooth fracture surface was observed in the PP/SEBS matrix at the nominal strain rate of 100 s^{-1} in both syntactic foams as shown in Figs. 9 (b-v) and 10 (b-v). When the relative density was 0.98, the matrix elongation pattern would be similar to the neat PP/SEBS blends as shown in Figs. 9 (b-i) and 9 (b-ii), and, Figs. 10 (b-i) and 10 (b-ii). As the summary, the observed fracture surfaces were similar in both PP/SEBS syntactic foams at the nominal strain rates of 1 and 100 s^{-1} . Therefore, it is considered that the fracture mechanism of the syntactic foams did not depend on the matrix material's property.

3.4 Effects of Matrix's Properties on Elastic Modulus and Yield Stress of Syntactic Foams

Currently, the quite popular theoretical model used to predict the modulus of closed-cell polymer foams is developed by Gibson-Ashby,¹⁾ which is generally related to its density by the formula

$$E_f = (\rho_f / \rho_{\text{solid}})^2 E_{\text{solid}} \quad (1)$$

where E_f is the modulus of the polymer foam. ρ_f and ρ_{solid} are the densities of the polymer foam and the corresponding solid matrix, respectively. The experimental results of the apparent elastic modulus of both PP/SEBS foams were plotted together with the theoretically predicted values from Eq. (1), as shown in Fig. 11. In both plots, it was assumed that the elastic moduli of the matrix (E_{solid}) in the foams were the same as those of the neat PP/SEBS A and PP/SEBS B blends. As shown clearly in Fig. 11, the apparent elastic moduli of the PP/SEBS A foams were close to the theoretical values at the strain rate of 10 and 100 s^{-1} while the apparent elastic moduli of the PP/SEBS A foams were relatively smaller than the theoretical values at the strain rates of 1 s^{-1} in the PP/SEBS A syntactic foams. On the contrary, the apparent elastic moduli of PP/SEBS B foams were much smaller than the theoretical values at the nominal strain rates of 1, 10 and 100 s^{-1} .

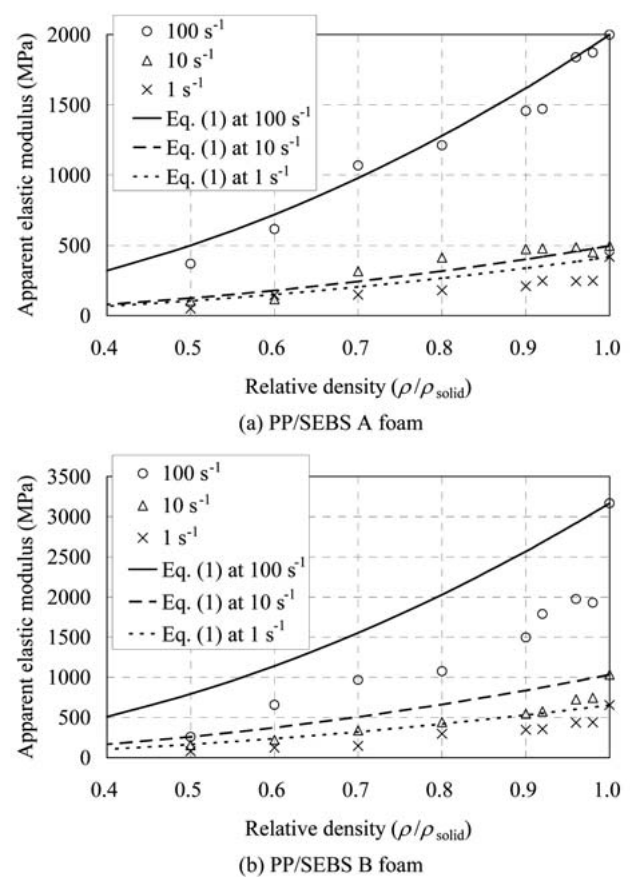


Fig. 11 Apparent elastic modulus vs. relative density with theoretical elastic modulus.

The interesting result here is that the apparent elastic modulus drastically decreased by adding the microballoon at even 2 vol % in the PP/SEBS B matrix although the PP/SEBS A foams followed Eq. (1) at the relative density of 0.98. Moreover, it was found that the apparent elastic moduli of both syntactic foams were similar values at the nominal strain rates of 1, 10 and 100 s^{-1} when the present microballoons were blended even at the small volume fraction. It is expected that the apparent elastic modulus of the PP/SEBS syntactic foam blended with

the present microballoons would have weak dependency of the matrix's apparent elastic modulus, leading to the similar elastic modulus regardless of the matrix's property. The further investigation of this mechanism is the future study.

In the case of the yield stress, the simple rule of mixtures gives the formula.¹⁹⁾

$$\sigma_{yf} / \sigma_{ysolid} = \rho_f / \rho_{solid} \quad (2)$$

where σ_{yf} is the yield stress of the polymer foam, and σ_{ysolid} is the yield stress of the matrix material, respectively. Fig. 12 shows the yield stresses plotted against the relative densities with those calculated by Eq. (2). In this plot, it was also assumed that the yield stresses of the matrix in the syntactic foams should be the same as those of the neat PP/SEBS A and PP/SEBS B blends. As shown clearly in Fig. 12, the yield stresses were smaller than the theoretical values calculated by Eq. (2) in both PP/SEBS A and PP/SEBS B syntactic foams, except that the yield stresses of PP/SEBS B syntactic foams were approximately close to the theoretical values at the relative density of 0.98 at the strain rates of 1, 10 and 100 s⁻¹. However, the whole trend of the yield stress was roughly similar between the PP/SEBS A and PP/SEBS B syntactic foams. This is due to the similar yield stresses in the neat PP/SEBS A and the neat PP/SEBS B blends, as shown in Fig. 3. The pre-

vious study indicated that the porous shape had strong effect on the yield stress of the syntactic foams.¹⁷⁾ In this study, the micro porous shape was the same between the PP/SEBS A and the PP/SEBS B foams as inferred in Figs. 9 and 10, leading to the similar yield stresses of the syntactic foams to those of the matrix materials.

4 Conclusions

The mechanical properties of two types of PP/SEBS blend syntactic foams were characterized at the nominal strain rates of 1, 10 and 100 s⁻¹. In addition, the effect of the mechanical properties of the matrix materials on the macroscopic mechanical properties of the syntactic foams were investigated by comparing the experimental results of the PP/SEBS A and PP/SEBS B syntactic foams. The followings are the conclusions of the present study :

(1) The apparent elastic modulus of the PP/SEBS A foam follows the Gibson-Ashby's law at the nominal strain rates of 10 and 100 s⁻¹, which indicates that the microstructural deformation mechanisms would be correlated well with the Gibson-Ashby's model. However, that of the PP/SEBS B foam does not follow the Gibson-Ashby's law.

(2) The apparent elastic modulus of the syntactic foams with the present microballoons becomes the similar values without depending on the matrix's apparent elastic modulus.

(3) The yield stress of PP/SEBS B foam follows the simple rule of mixture at the relative density of 0.98. However, except for that relative density, the yield stresses of PP/SEBS A and PP/SEBS B foams do not follow the simple mixing rule.

(4) The material ductility of PP/SEBS A and PP/SEBS B foams decrease drastically once the microballoons are blended in the matrix materials.

(5) The strain rate dependency of the elastic modulus in the PP/SEBS syntactic foams is dependent on that of the matrix material.

(6) The drastic increase of the elastic modulus in the PP/SEBS syntactic foams is delayed by adding the polymer microballoons.

The polymer based syntactic foams have an advantage in light weight, high energy absorbability, low cost and good productivity for the applications not only in automobile but also in any other field, such as airplane, mobile computers, packaging and so on. Therefore, more detailed studies are encouraged. Especially, the reason why the apparent elastic modulus of the syntactic foams was similar values without depending on the apparent elastic modulus of the matrix is left as future work. In addition, the effects of the local strain rates which would change with the micro porous morphologies are to be further explored in the future study.

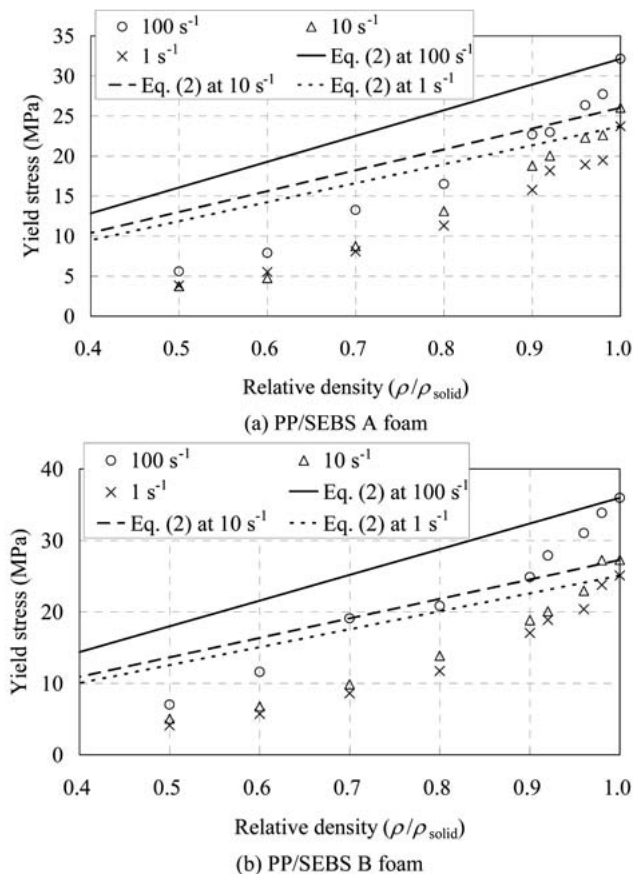


Fig. 12 Yield stress vs. relative density with theoretical yield stress.

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