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EVALUATION OF DYNAMIC COMPRESSIVE PROPERTIES OF PLA/PBAT POLYMER ALLOYS USING SPLIT HOPKINSON PRESSURE BAR METHOD

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Poly(lactic acid) (PLA) is a biodegradable biomass polymer. Polymer blends or polymer alloys have been used to overcome its brittleness. Because poly(butylene/adipate/terephthalate) (PBAT) is a ductile and biodegradable polymer, PLA/PBAT polymer alloys can potentially exhibit high impact strengths. Different mixing ratios of PLA/PBAT (80:20, 70:30 and 60:40) and the addition of dialkylperoxide as compatibilizing agent for each mixing ratio have been examined. The stress-strain curves of the PLA/PBAT specimens (observed using a scanning electron microscope) were measured using a split Hopkinson pressure bar (Kolsky bar) and a universal testing machine. The PBAT ratios and addition of dialkylperoxide affected the stress-strain curves. Yield stress decreased with increasing PBAT ratios. Addition of dialkylperoxide did not change the yield stress of specimens when PLA:PBAT = 80:20. At high strain rates, the addition of dialkylperoxide clearly reduced yield stress and Young's modulus when PLA: PBAT = 70:30 and 60:40.

1. Introduction

The increasing use of plastic products worldwide is causing considerable damage to the environment; therefore, biodegradable plastics (plastics that can decompose in the natural environment) and biomass plastics (plant-derived or recyclable-resource-based plastics) are being extensively investigated, and new biodegradable and biomass plastics are continuously being developed. Poly(lactic acid) (PLA) is a typical biodegradable biomass polymer (plant-derived polymer). In Japan, PLA is already being used to manufacture many industrial products such as the interior parts of cars, parts of computer cases, and cell-phone cases. Many studies have been conducted to determine other industrial products that can be manufactured using PLA. However, such applications are limited to machine parts that are subjected to low loading. Polymer blends/alloys or natural fiber reinforcing have been used to overcome the brittleness of PLA [1–4]. Because poly(butylene adipate / terephthalate) (PBAT) is a ductile and biodegradable polymer, PLA/PBAT polymer blends and alloys can potentially exhibit high impact strengths [5, 6]. In most cases, the impact resistances of biodegradable plastics and biomass plastics are based only on the experimental results of Izod/Charpy impact strength tests and Dynatup impact tests. However, the basic mechanical properties of such plastics with respect to the impact resistances remain unknown.

In the present study, the stress-strain curves of PLA/PBAT polymer alloys were measured using a universal testing machine and a split Hopkinson pressure bar (Kolsky bar) system. The effects of the mixing ratios of PLA/PBAT and the addition of dialkylperoxide as a compatibilizing agent on the Young's modulus and flow stress were also examined.

2. Experimental methods

2.1. Materials

We used PLA/PBAT alloys prepared by means of PLA from Toyota Motor Corporation (Toyota Eco-Plastic S-17) and PBAT from BASF (Ecoflex). In order to examine the effect of a compatibilizing agent, we used dialkylperoxide (NOF Corporation, PERHEXA 25B). The mixing ratios of PLA and PBAT were 80:20, 70:30, and 60:40. The mixing ratios of PLA:PBAT:dialkylperoxide were 80:20:1, 70:30:1, and 60:40:1. We prepared the polymer alloys using a twin-screw extruder (TECHNOVEL CORPORATION) at 180°C, a screw speed of 400 rpm, and a feed rate of 100 g/min. After melt mixing, the strands prepared by the twin-screw extruder were cooled rapidly, pelletized, and then dried. Next,

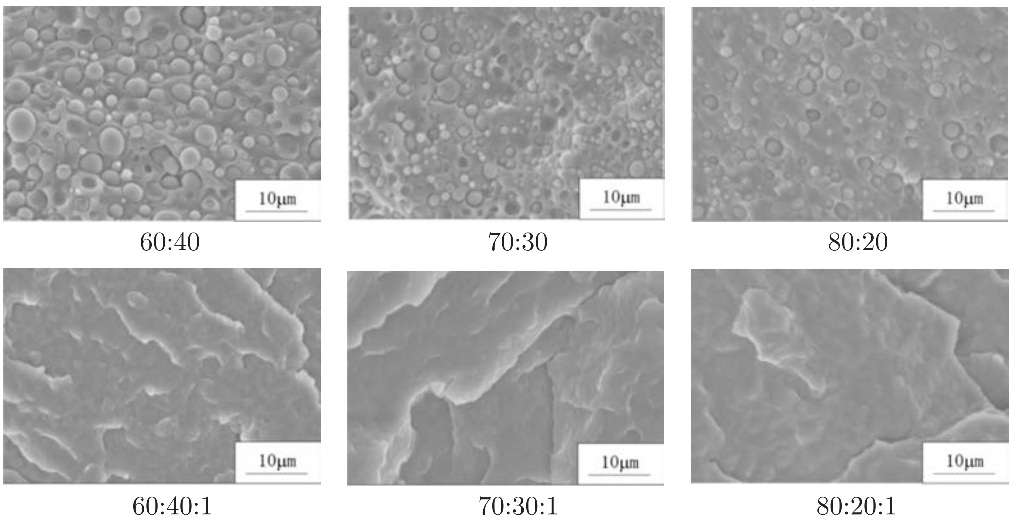

Fig. 1. Photographs of cryo-fractured surfaces (observed using a scanning electron microscope).

5-mm-thick plates were prepared using a conventional hot press at 190°C and 5 MPa for 30 min. Figure 1 shows photographs of the cryo-fractured surfaces of specimens captured using a scanning electron microscope.

In the case of the PLA/PBAT specimens, we can see a sea-island structure consisting of the PLA matrix and PBAT particles of the same size (domain). When we used the compatibilizing agent, we could not observe PBAT particles and phase separation clearly.

2.2. Izod impact tests

Izod impact test specimens were prepared using a milling machine. The specimens had an A-type notch and 63.5 × 12.7 × 5 mm dimensions. An Izod impact testing machine (Toyo Seiki Seisaku-sho, Ltd.) was used.

2.3. Compressive tests

Compressive test specimens were produced using a lathe, and their end faces were polished and parallelized. We used dynamic compressive test specimens with a diameter of approximately 15 mm in order to accurately measure the stress-strain curves using our equipment. The specimen thickness was 5 mm. The photograph of a compressive test specimen is shown in Fig. 2. In the quasi-static tests based on ASTM D695-02a, we used specimens with a diameter and thickness of 6 mm and 9 mm, respectively.

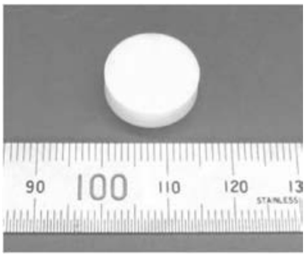

FIG. 2. Photograph of a dynamic compressive test specimen.

The quasi-static compressive tests were conducted with strain rates ranging from 10^{-4} to 10^{-2} s^{-1}, using a universal testing machine (A&D Company, Ltd., RTM-500). At high strain rates of 10^2 to 10^3 s^{-1}, compressive properties of the specimens were examined using the split Hopkinson pressure bar method (Kolsky bar), as shown in Fig. 3. The input and output bars were made of an aluminum alloy (A2024-T4), and their diameters and lengths were 28 mm and 1900 mm/1300 mm, respectively. Strain gages were applied to both sides of the input and output bars at distances of 950 mm and 300 mm from the

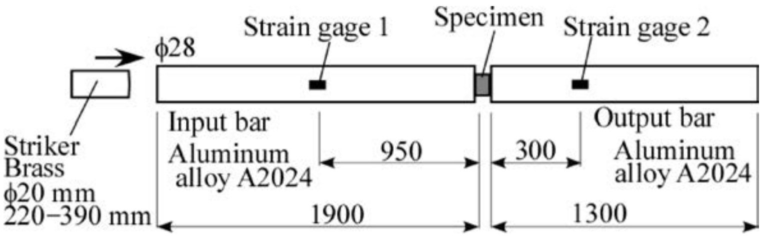

FIG. 3. Experimental setup for split Hopkinson pressure bar method (Kolsky bar).

specimen, respectively. As the stress histories were almost equal on both sides of the specimens, the strain and stress of the specimens were calculated from the strain on the bars using Eqs. (2.1) and (2.2), which are given below; in addition, the strain on the bars was measured using the strain gages [7]:

$$\varepsilon(t) = \frac{2c_3}{L} \int_0^t [\varepsilon_I(t) - \varepsilon_T(t)]\, dt, \tag{2.1}$$

$$\sigma(t) = \frac{AE}{A_S} \varepsilon_T(t). \tag{2.2}$$

Here ε_I and ε_T are the axial strains induced in the input bar by the incident wave, and in the output bar by the transmitted wave, respectively. E and c_3 are Young's modulus and elastic wave velocity, respectively, of the both the input and the output bars. L is the specimen thickness. A and A_S are the cross-sectional areas of the input/output bars and specimens, respectively. Material constants of the aluminum alloy (A2024-T4) bars used in the calculations are listed in Table 1. We used brass strikers with a diameter of 20 mm and a length of 220 to 390 mm. During the experiments, the humidity in the laboratory was 15% to 50%. Specimens were maintained at a temperature between 23°C and 25°C using silicone rubber heaters.

Table 1. Material constants of input and output bars used in calculations.

Density	Elastic wave velocity in the bar, c_3	Young's modulus E
2.77×10^3 kg/m^3	5150 m/s	73.6 GPa

3. Results and discussion

3.1. Results of Izod impact tests

The results of the Izod impact test are shown in Fig. 4. The figure shows the standard deviation of the results and their statistically significant difference. The

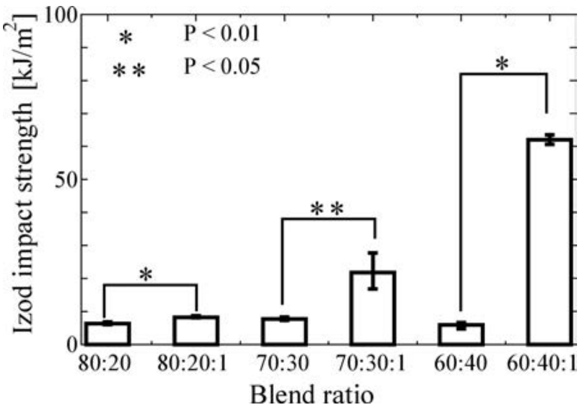

FIG. 4. Impact resistance of different PLA/PBAT alloys.

addition of dialkylperoxide increased Izod impact strength for each PBAT ratio, corroborating the results published by FUKUDA *et al.* [5]. In particular, when the mixing ratio of PLA:PBAT:dialkylperoxide was 60:40:1, the Izod impact strength was 60 kJ/m^2, comparable to polycarbonate (PC).

3.2. Results of compressive tests

The dynamic properties for PLA/PBAT alloy specimens were examined using the split Hopkinson pressure bar method. Figure 5 shows the stress-strain curves for PLA/PBAT alloy specimens, obtained from the strain history and stress history using Eqs. (2.1)–(2.2). Because the strain rate changed slightly during compression, it was determined using the averaged value of the strain rate-strain curve [8]. When PLA:PBAT = 80:20, the stress-strain curve for the specimen peaked near the elastic limit and then, the stress decreased gradually

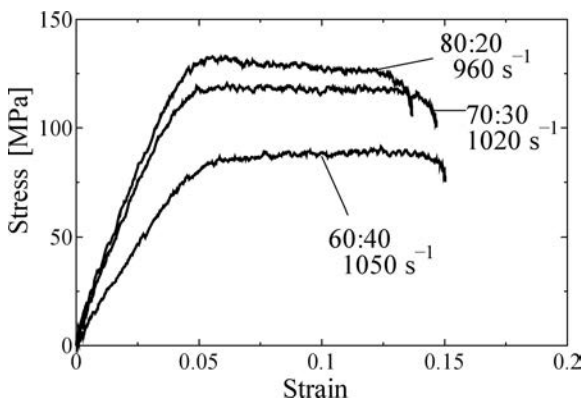

FIG. 5. Effect of PBAT content on stress–strain curves at strain rates of 960–1050 s^{-1}.

with increasing strain (softening). As the PBAT ratio increased, the peak of the stress-strain curve became smaller, and the yield stress and Young's modulus decreased. When PLA:PBAT = 70:30, the flow stress remained almost constant. When PLA:PBAT = 60:40, the flow stress increased slightly (work hardening).

Next, in order to examine the effect of dialkylperoxide addition, the stress-strain curve for PLA:PBAT = 60:40 and PLA:PBAT:dialkylperoxide = 60:40:1 was plotted, as shown in Fig. 6; the curve for PLA:PBAT = 70:30 and PLA:PBAT:dialkylperoxide = 70:30:1 was plotted, as shown in Fig. 7. The addition of dialkylperoxide reduced the yield stress and Young's modulus of the specimens and increased the work hardening.

a) 1050–1090 s^{-1} b) 560–650 s^{-1}

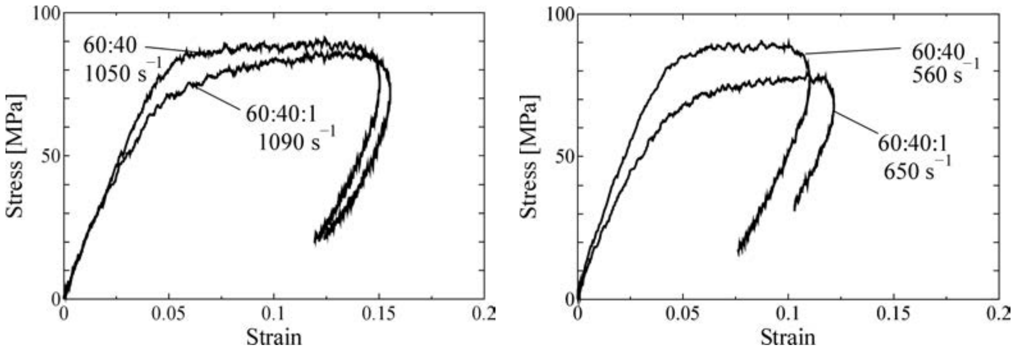

FIG. 6. Effect of dialkylperoxide addition on the stress-strain curve for PLA:PBAT = 60:40 and PLA:PBAT:dialkylperoxide = 60:40:1.

a) 990–1020 s^{-1} b) 750–790 s^{-1}

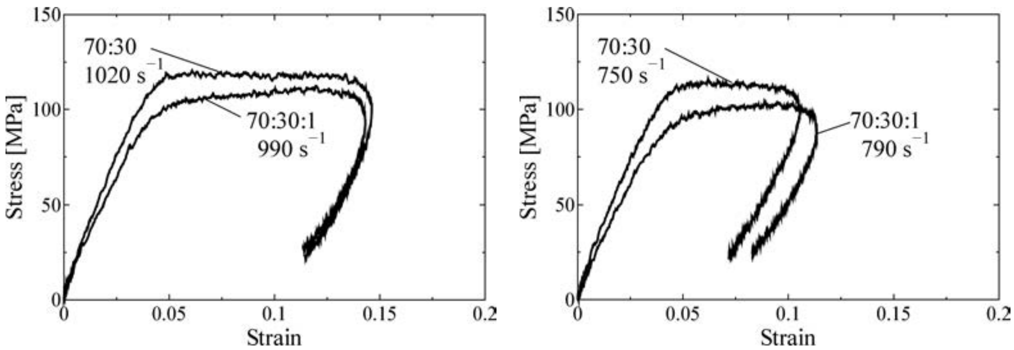

FIG. 7. Effect of dialkylperoxide addition on stress-strain curve for PLA:PBAT = 70:30 and PLA:PBAT:dialkylperoxide = 70:30:1.

Finally, Fig. 8 shows the effect of the strain rate on the yield stress. For each polymer alloy, the yield stress increased with the strain rate, what is commonly seen in most engineering plastics such as poly(methyl methacrylate) (PMMA)

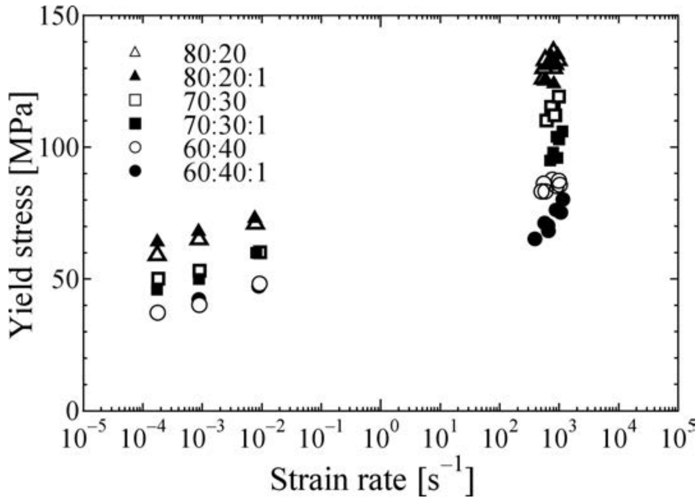

Fig. 8. Effect of strain rate on maximum stress.

and PC. As the PBAT ratio increased, the yield stress decreased at high and low strain rates. At low strain rates, we can see only a few effects of dialkylperoxide addition on the yield stress for each PBAT ratio. At high strain rates, the addition of dialkylperoxide clearly reduced the yield stress only for PLA: PBAT = 70:30 or 60:40. The strain rate dependence of the yield stress was greater when PLA:PBAT: dialkylperoxide = 70:30: 1 and 60:40:1 than when PLA:PBAT = 70:30 and 60:40.

4. Conclusions

We have examined specimens with different mixing ratios of PLA/PBAT and the effect of the addition of dialkylperoxide on them. The addition of dialkylperoxide increased the Izod impact strength. The stress-strain curves of PLA/PBAT specimens were measured using a split Hopkinson pressure bar and a universal testing machine. The yield stress and Young's modulus decreased with increasing PBAT ratios. At high strain rates, when PLA:PBAT = 70:30 and 60:40, the addition of dialkylperoxide reduced the yield stress and Young's modulus, and increased the work hardening. The strain rate dependence of the yield stress was greater when PLA:PBAT: dialkylperoxide = 70:30:1 and 60:40:1 than when PLA:PBAT = 70:30 and 60:40.

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