

## Rolling of biodegradable poly(butylene succinate) PBS sheets

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### INTRODUCTION

The objective of the study was to investigate the properties improvement of sheets from biodegradable polyesters by solid-phase deformation methods. The aliphatic polyesters, poly (butylene succinate), PBS, poly (L-lactic acid), PLLA and poly (hexano-6-lactone), PCL, have been received much attention due to their biodegradability. To obtain the best final products for particular use of biodegradable polymers, many factors related to the structure, properties and performance should be considered. In this study we try to modify the properties of PBS through the solid-phase deformation methods which can improve the mechanical properties by means of the molecular orientation and the development of fine-structures. On the other hand, the coloring of biodegradable polymers is open to utilization in various ways. The PBS was colored by synthetic food color additives. The mixing of PBS and food color additives was successfully performed in the melt state of PBS by use of a twin screw extruder. The colored PBS sheets can be stretched to draw ratio as high as 4 times by means of rolling methods. The changes of appearance, transparency, and mechanical properties were studied. The mechanical properties were directly related to draw ratio (rolling ratio), and the tensile strengths of the drawn sheets were also improved by the molecular orientation of PBS. This process work is important means to develop the non-toxic and 'green' plastics which have many potential applications.

### EXERIMENTAL

*Materials and sheets preparation* Poly (butylene succinate), PBS, studied in this work is a commercial BIONOLLE #1010 obtained from Showa Highpolymer Co. Ltd. Natural color, Curcumin (CI 75300) and four kinds of commercially available synthetic food colors, Phloxine B (CI45410), Sunset Yellow FCF (CI15985), Fast Green FCF (CI42053), Brilliant Blue FCF (CI 42090), were used as the color additives. Mixing of PBS and color additives was conducted by use of a twin screw extruder. Extrudates through T-die were quenched by a pair of chill rolls at 40 °C. Colored PBS sheets of 0.8 mm thickness and 50 mm width were obtained.

*Rolling* The solid-phase deformation of PBS sheets was conducted by using a laboratory size rolling device (Figure 1). Rollers with a diameter of 120 mm and a length of 150 mm were heated with internal cartridge heaters. The rolling mill was equipped with an apparatus for applying tension forces to the sheet during rolling. Rolling was conducted at 60 °C.

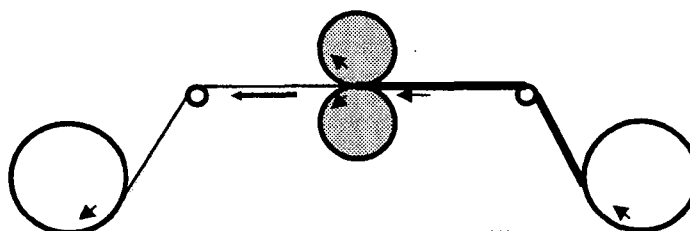


Fig.1 Schematic drawing of rolling mill

## RESULTS AND DISCUSSION

Extruded PBS sheets were semi-crystalline and the appearances were white and opaque. Coloring of PBS sheets by natural color, Curcumin, and synthetic food color additives was successfully conducted. The color additives used in this study showed very good dispersion in PBS sheets by melt-mixing. The colored sheets show the good appearances and clear colors. As-extruded PBS sheets were highly crystallized and deformed with necking propagation after yielding.

Both visual estimation and haze measurement cause remarkable changes of optical clarity of colored PBS sheets with rolling. There is a sharp drop in the haze measured for sheets with rolling ratio between 1.5 and 2.5, corresponding to a change from opaque sheets to clear sheets (figure 2).

The mechanical properties of PBS were improved by the rolling. Tensile modulus and breaking stress of PBS sheets increased with the increase of rolling ratio. To know the thermo-mechanical properties, measurements of temperature dependence of dynamic mechanical properties were conducted. DMA curve of extruded sheet shows the  $\alpha$  dispersion temperature at about  $-26\text{ }^{\circ}\text{C}$  corresponding to the glass transition temperature,  $T_g$ , of PBS. The change of  $\alpha$  relaxation temperature with rolling is of interesting. The peak temperature of the loss modulus  $E''$  increased with the increase of rolling ratio. Also, the dynamic storage modulus increased with increasing rolling ratio. These phenomena give information for crystallization and the molecular orientation in the PBS sheets.

### Conclusion

The deformation behavior of colored biodegradable aliphatic polyester, PBS, was investigated to know the effect of molecular orientation on the properties of PBS sheets. The results can be summarized as follows;

1. Coloring of PBS sheets by natural and synthetic food colors was successfully conducted.
2. The enhancements of mechanical and thermo-mechanical properties of PBS sheets can be performed by plastic deformation.
3. The molecular orientation of PBS films increased by the rolling process, which accompanied by the increase of glass transition temperature and the transparency.

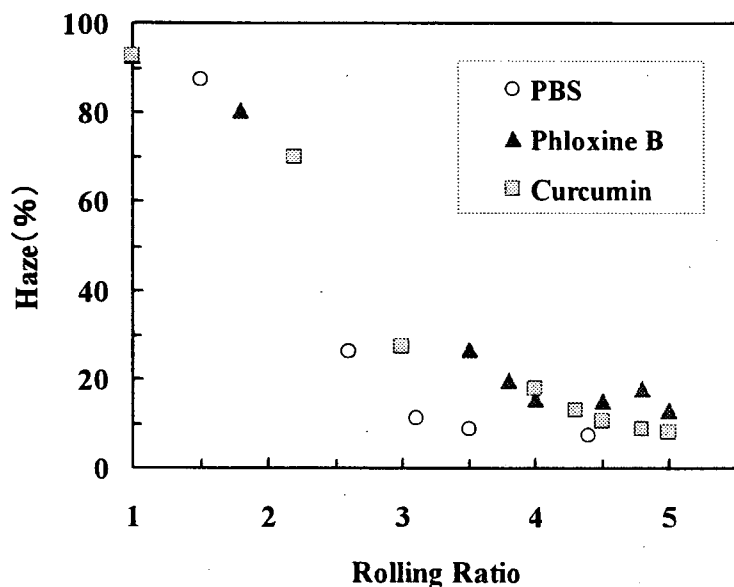


Figure 2 Increase of transparency with rolling of PBS and colored PBS sheets.