

CHAPTER VII

CONCLUSIONS

Three practical approaches based on balancing of crystalline and amorphous phases, were applied to overcome PLA brittleness which are (i) compatible nucleation, (ii) plasticizer addition, and (iii) processing conditions control. In the case of the compatible nucleation, starch was simply modified with different organofunctional silane coupling agents. From the detailed structural analysis by ^1H - ^1H TOCSY NMR spectrum, CPMS successfully formed covalent bonds with starch. The CP-starch obtained was consequently coupled with PLA for the reactive PLA/CP-starch blending. As compared to PLA, the reactive PLA/CP-starch blend brought not only the effective compatibility improvement but also the 10-fold increased X_c with decreased T_g for 10 °C. Furthermore, CP-starch was developed to be thermoplastic starch (mTPS) which was subsequently blended with PLA. The physical properties of PLA/mTPS blend performed the potential in use as mulch film such as fully biodegradability, relatively low oxygen permeability, and accelerated PLA degradation, etc. For processing conditions control, the BO process was applied and the PLA microstructure regularization change was followed by X-ray techniques. By systematically varying stretching rates and draw ratios, the relationship between PLA mechanical properties and microstructure regularization was established. At low stretching rate of 3 mm/s, the mesophase and large PLA crystals with several tens of nanometers were found, leading to significant increase of tensile strength but slightly improved elongation at break. When the stretching rate and the draw ratio were increased to a certain high level (75 mm/s stretching rate combined with 5×5 draw ratio), many isotropically small PLA crystals in δ -form were produced. This resulted in the drastically improved toughness for 4 times, as compared to conventional PLA blown film. In the approach of plasticizer addition, PLLA-PEG-PLLA triblock copolymer with varied PLLA chains length was prepared by simple condensation of L-lactic acid and diol-PEG. From the replacing of PEG by PLLA-PEG-PLLA, the phase separation due to self-recrystallization of PEG was clearly obstructed although the PLLA chain length was extended. The crystallization of PLA

in the system was significantly accelerated by PLLA-PEG-PLLA without recrystallized PEG phase in PLA matrix. Moreover, the flexibility of PLA/PLLA-PEG-PLLA films was drastically enhanced, similar to that of PLA/PEG homopolymers film.