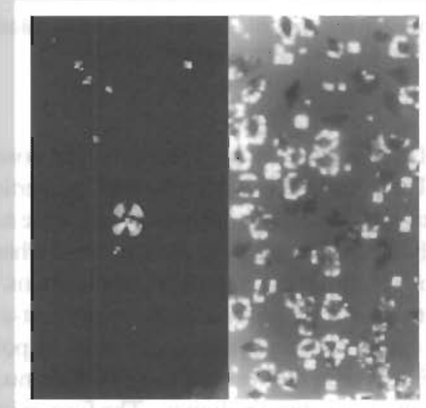
Summary: The effects of various additives: poly(D-lactic acid) (PDLA), talc, fullerene C_{60}, montmorillonite, and various polysaccharides, on the non-isothermal crystallization behavior of poly(L-lactic acid) (PLLA), during both the heating of melt-quenched films from room temperature, and the cooling of as-cast films from the melt, was investigated. When the melt-quenched PLLA films were heated from room temperature, the overall PLLA crystallization was accelerated upon addition of PDLA or the stereocomplex crystallites formed between PDLA and PLLA, the mixtures containing PDLA, and the mixture of talc and montmorillonite. No significant effects on the overall PLLA crystallization were observed for talc, C_{60}, montmorillonite, and the mixtures containing C_{60}. Such rapid completion of the overall PLLA crystallization upon addition of the aforementioned additives can be ascribed to the increased density (number per unit volume or area) of PLLA spherulites. When the as-cast PLLA films were cooled from the melt, the overall PLLA crystallization completed rapidly, upon addition of PDLA, talc, C_{60}, montmorillonite, and their mixtures. Such rapid overall PLLA crystallization is attributable to the increased density of the PLLA spherulites and the higher nucleation temperature for PLLA crystallization. In contrast, the addition of various polysaccharides has no significant effect, or only a very small effect, on the overall PLLA crystallization during heating from room temperature or during cooling from the melt. This finding means that the polysaccharides can be utilized as low-cost fillers for PLLA-based materials, without disturbing the crystallization of the PLLA. The effect of additives in accelerating the overall PLLA crystallization during cooling from the melt, decreased in the following order: PDLA > talc > C_{60} > montmorillonite > polysaccharides.



Polarization optical photomicrographs of *pure* PLLA, and the PLLA-F film, with the fullerene additive, during cooling from the melt (Process IIB). Both of the photomicrographs were taken at 120 °C.

Non-Isothermal Crystallization Behavior of Poly(L-lactic acid) in the Presence of Various Additives

Hideto Tsuji,[*1] *Hiroki Takai,*[1] *Norio Fukuda,*[2] *Hirofumi Takikawa*[3]

[1] Department of Ecological Engineering, Faculty of Engineering, Toyohashi University of Technology, Tempaku-cho, Toyohashi, Aichi 441-8580, Japan
E-mail: tsuji@eco.tut.ac.jp
[2] Aichi Industrial Technology Institute, Nishishinwari Hitotsugi-cho, Kariya, Aichi 448-0003, Japan
[3] Department of Electrical and Electronic Engineering, Faculty of Engineering, Toyohashi University of Technology, Tempaku-cho, Toyohashi, Aichi 441-8580, Japan

Received: November 4, 2005; Revised: December 20, 2005; Accepted: December 21, 2005; DOI: 10.1002/mame.200500371

Keywords: additives; biodegradable polyesters; crystallization; fillers; fullerenes; nucleation

Introduction

Poly(L-lactic acid) (PLLA) has been intensively investigated because it is biodegradable, producible from renewable resources, nontoxic to the human body and the environment, and can be composted.[1–6] Moreover, PLLA has a high mechanical performance, comparable to that of commercial polymers such as polystyrene and poly(ethylene terephthalate), and is utilized as a biomedical material in tissue regeneration and as a matrix in drug delivery