CHAPTER I
INTRODUCTION

Due to the environmental issues developed years by years, poly(lactic acid) (PLA) is a green polymer which becomes an alternative plastic for today with reliable industrial scale production. It can be utilized in a wide range of applications, e.g., biomedical items, agricultural field, textile and disposal packaging because of its excellent transparency, high strength, moderate barrier properties and fully biodegradability. The fact that PLA is a brittle material with slow crystallization rate, it is still limited in some demands which require the ductility and toughness. The formation of semi-crystalline structure, thus, is necessary to overcome the brittleness of PLA.

Up to the present, various nucleating agents and plasticizers additions have been applied in order to improve PLA toughness by increase of PLA crystallinity with decrease of PLA crystallization temperature. As always found, when two different phases are blended together, the phase separation appears as consequence which can be overcome by reactive compatibilizers. In addition, another practical approach being able to trigger the toughened PLA film is processing conditions control through stretching and annealing.

We believe that the development of tough PLA film with effectiveness, the key factors involved with the induced PLA crystallization and the optimized amorphous and crystalline phases, are needed to be comprehended. The highly miscible and crystalline components in the system, PLA chain mobility, high ordered microstructure as well as regular chain packing in the crystal lattice are realized in the success of the induced PLA crystallization with balancing of amorphous and crystalline phases. The studied points are (i) how the designed nucleating agent and plasticizer can contribute the good miscibility at nucleation sites and the acceleration of PLA crystallization, including PLA chain mobility, and (ii) how the stretching process can produce high ordered structure and influences on the PLA orientation. Therefore, this present work focuses on starch modified with a series of silane coupling agents (silane-starch) as nucleating agent, and the biaxial-stretching process in order to enhance PLA crystallization with systematic studies. Additionally, the

synthesized poly(L-lactic acid)-poly(ethylene glycol) triblock copolymer (PLLA-PEG-PLLA) as miscible plasticizer is also speculated for the toughened PLA film.

In the case of starch, the varied organofunctional silanes are simply coupled with starch in order to effectively form the compatible reactive PLA/silane-starch blend. For biaxial-stretching process, the different stretching rates and draw ratios are investigated which drastically change the PLA film performance. Furthermore, the PLLA-PEG-PLLA triblock copolymers with varied PLLA chains length are simply prepared by condensation reaction which is expected to improve PLA ductility effectively.

The investigations in molecular level were carefully carried out to illustrate the improved miscibility between PLA and starch or PLLA-PEG-PLLA in the entire system as well as the changes of PLA microstructure regularization by reactive blending and biaxial-stretching process, including crystallization behavior and thermal property. From an understanding in details, it enables us to establish the relationship of the PLA mechanical properties and the various microstructures and/or chain packing regularization.

In overall, this present dissertation endeavors to manifest how we can develop PLA toughness in virtues of practical approaches based on optimizing between amorphous and crystalline phases. The systematic studies were carried out to apprehend the factors related to PLA crystallization such as chains mobility, microstructural orientation, chain packing structures, etc. The information obtained from this work will be a guideline in effective and efficient development of a crystalline and tough material for other rigid polymers further.