CHAPTER I

INTRODUCTION

Polylactide or polylactic acid (PLA) is well-known as an environmental-friendly material, due to its biodegradability, biocompatibility, and renewability. It has attracted increasing interest in various markets, such as packaging, textile, automotive, and biomedical devices. However, PLA is too brittle at room temperature, because of its high glass transition temperature (T_g) which is in the range of 55 to 65 °C. Its low toughness is also a major drawback for expanding its applications as commodity plastic materials. In accordance with other brittle plastics, PLA can be toughened via plasticization, copolymerization, and blending with varieties of flexible polymers, such as natural and synthetic rubbers [1-4].

Among various types of synthetic rubbers, isoprene rubber (IR) and acrylonitrile-butadiene rubber (NBR) are the promising toughening agents for PLA [4]. The former exhibits high ability to improve elongation at break, whereas the latter improves Izod impact strength of PLA. However, tensile strength and modulus of PLA decreased with the incorporation of all type of rubbers. Natural rubber (NR), a major natural products widely produced in Thailand, has also been employed as a toughening agent for PLA. NR composes of *cis* 1,4- polyisoprene which produces excellent physical properties, i.e., high strength, high flexibility, high impact strength, low heat buildup, and very good tear resistance [5]. Although NR has been introduced into many thermoplastics as a toughening agent, the application in PLA is limited because of a dissimilarity in their solubility parameters. Increasing of NR's polarity could be achieved by converting its double bonds to epoxides, generating epoxidized natural rubber (ENR) [6, 7].

ENR has been introduced as a toughening agent for various polar polymers. The introduction of NR and ENR leaded to a decrease of tensile strength, but a slight increase in elongation at break and impact strength of Polyamide (PA6).

Improvement of impact strength of the blends could be explained in terms of the blend morphology. Large-sized particles of NR in polymer matrix were the cause of its premature failure. Because of the significant improvement in toughness of polymers is compromised with a great loss in strength and stiffness, crosslinked ENR has been employed. Typically, incorporation of crosslinked ENR increased tensile properties, but decreased impact strength of the blends. This was probably due to the fully-crosslinked rubber fraction changed the morphology from small particles to large patches after crosslinking, leading to poor dispersion of rubber domains. [5]

In this research group, Nguyen et. al [8, 9] had studied crosslinking efficiency of ENR by OH-capped polylactic acid derivatives. It was found that not only crosslinked structure, grafted and simple blend structures were produced during the crosslinking process. A variation in contents of different chemical structures leaded to an improvement of tensile strength and elongation at break, compared to those of uncured ENR. These cured materials with tunable properties showed high potential for use as toughening agents for brittle PLA, as the materials contain flexible rubber domains and PLA blocks that can enhance compatibility with PLA. In addition, it has been proven in ref. 10-13 that hydroxyl-terminated lactic acid prepolymers are major products from the glycolysis of PLA resins by ethylene glycol. These prepolymers are promising starting materials for further developing of degradable thermoplastics [10-12] and thermoset (co)polyesters [13]. Therefore, potency of PLA glycolyzed products as polymeric crosslinkers for ENR is of interest of this dissertation. Effects of content of PLA-glycolysates (GPLA) and their chain lengths on chemical structures and properties of the GPLA-cured ENR products are examined. Efficiency of the curing process and its products crosslink density are measured. Tensile and thermal properties of the cured products are investigated. Feasibility of using the GPLA-cured ENR materials as toughening agents for commercial PLA resin is examined in the latter part of this work. Effects of structures of GPLA-cured ENR products and their blend compositions on impact and tensile properties, and morphology of the toughened PLA are studied.