CHAPTER IX
CONCLUSIONS AND RECOMMENDATIONS

Hexanoyl chitosan (H-chitosan) was synthesized by reacting chitosan with hexanoyl chloride to obtain products with various degrees of hexanoylation. The obtained H-chitosan exhibited much improvement in the solubility in organic solvents such as chloroform, dichloromethane and tetrahydrofuran. H-chitosan with degree of substitution of 3.91 was dissolved in various organic solvents in order to study the effect of the solvent on solution properties of H-chitosan. Dilute solution properties of H-chitosan in three organic solvents, i.e. chloroform, dichloromethane, and tetrahydrofuran, were studied to investigate the relationship between polymer-solvent interaction, the solution viscosity, and hydrodynamic radius. The calculated solubility parameter of H-chitosan was estimated to be 9.31 $(cal.cm^{-3})^{0.5}$ by group contribution method. This estimated value can be compared with those published values of chloroform, dichloromethane and tetrahydrofuran which were used as the solvents. The solubility parameter value of H-chitosan is close to the solubility parameter value of chloroform more than those of dichloromethane and tetrahydrofuran. The results from viscosity and dynamic light scattering technique indicated that the three H-chitosan solutions exhibited comparable polymer-solvent interactions as they are expected to be for a good solvent. A more favorable unwinding of the polymer chains was found in chloroform. This could be a result of the small discrepancy between the solubility parameters of H-chitosan and chloroform. Although chloroform, dichloromethane and tetrahydrofuran are good solvent for H-chitosan, chloroform is considered to be the best, as confirmed by the highest value of intrinsic viscosity and hydrodynamic radius.

Blends of H-chitosan and polylactide (PLA) were prepared into two forms, films and electrospun fibers for potential uses in biomedical applications. It is expected that the addition of H-chitosan helped in improvement of the physical and mechanical properties of the normally-brittle PLA. Blend films and electrospun fibers of H-chitosan and PLA were prepared by the solution-casting and electrospinning technique, respectively, from corresponding blend solutions using different organic solvents, such as chloroform, dichloromethane or tetrahydrofuran.

Effect of solvent or solution properties on the H-chitosan/PLA blend products was obviously seen in electrospinning fibers than cast films.

In the case of blend films, only the blend solutions in chloroform and dichloromethane could be cast into films at all blend compositions. TGA, DSC, SEM, and WAXD results indicated that the blend products exhibited partial miscibility between H-chitosan and PLA molecules in the amorphous phase at low H-chitosan contents (i.e. $\leq$ 40 wt.%) and became more immiscible when H-chitosan content increased. As proved by constancy of melting temperature and the observation of phase separation. In addition, the miscibility of H-chitosan/PLA blend films from solution of chloroform was better than those in dichloromethane solution. Physically, the blend films appeared to be softer and more elastic as H-chitosan content increased.

In case of electrospun fibers, the as-spun fibers from PLA solutions in all of the three solvents appeared to be round in their cross-section, with rough surface morphology, while the as-spun fibers from H-chitosan solutions in all of the three solvents were flat, with smooth surface morphology. The as-spun fibers from H-chitosan/PLA blend solutions in chloroform with the H-chitosan solution content less than or equal to 50% w/w were continuous without the presence of beads, while those from the blend solutions in dichloromethane were beaded fibers or even just beads at high H-chitosan solution content. Interestingly, the diameters of the as-spun fibers from blend solutions with H-chitosan solution content less than or equal to 50% w/w were found to decrease with increasing amount of H-chitosan portion.

The inspiration of this work was to find an economical way for improving the applicability of H-chitosan through the blending with PLA. These blend systems are expected to have considerable importance in several fields, especially in biomedical applications, because of their biocompatibility and biodegradability. They are expected to have potential uses in tissue engineering and composition-dependent biodegradability. Recommendations for future work are on the biodegradation behavior and applications in tissue engineering. Since the present studies showed that H-chitosan and PLA were only partially miscible in the amorphous phase at low H-chitosan content, the addition of a compatibilizer such as glycol might be beneficial to the final properties of the blends.