CHAPTER I

INTRODUCTION

1.1 Motivation

Emulsifier is an essential substance for preparing emulsion. It is added into the mixture of oil and water in order to reduce an interfacial tension. Emulsifiers can be classified into 2 main categories, small molecular surfactants and polymeric surfactants. Small molecular surfactants, such as sorbitan monosterate, glyceryl sterate, and stearyl alcohol, are normally composed in detergents, lotions and creams. Whilst, polymeric surfactants, such as protein and polysaccharide, are widely used in food grade emulsions. Each type of emulsifier has its particular advantages. Normally, small molecular surfactants have high efficiency on generating small emulsion droplets during emulsification. Polymeric surfactants have high efficiency on providing long term stability. However, there are some disadvantages on using those surfactants. Some small molecular surfactants do not provide strong enough droplet-droplet repulsion. On the other hand, polymeric surfactants exhibit low efficiency on generating small droplets during emulsification.

Protein and polysaccharide show many emulsifying properties. Most protein is highly effective emulsifier due to amphiphilic property, or in other words, they contain both hydrophilic and hydrophobic moiety in the structure. Moreover, protein shows amphoteric property, or containing both positive and negative charges in the structure to promote emulsion stability through electrostatic repulsive force. However, the electrostatic repulsive force decreased around isoelectric point leading to droplet flocculation.

Viscous polysaccharides used to stabilize emulsion by thickening the aqueous phase. They are insensitive to change properties in pH and salt. However, they are less effective to use as emulsifiers because they mostly have hydrophilic regions, but they have least hydrophobic regions.

Nowadays, many researchers made an effort to improve these properties via (i) the covalent linking of proteins to polysaccharide, such as ovabumin-dextran conjugates [1], whey protein isolate-maltodextrin conjugates [2], caseinate-dextrans conjugate [3] (ii) using a naturally occurring protein-polysaccharide complex, such as gum Arabic [4]. Nevertheless, the conjugation of protein to polysaccharide was mostly prepared by dry-heat treatment which is complicated process and time consuming. Furthermore, gum arabic is necessary to used in high concentration to emulsify O/W emulsion [5].

From literature review, we found that polysaccharide conjugate protein showed good emulsifying properties with good stability. Therefore, it is our interest to synthesize sodium phosphorylated chitosan (PCTS), which is phosphate derivative of chitosan.

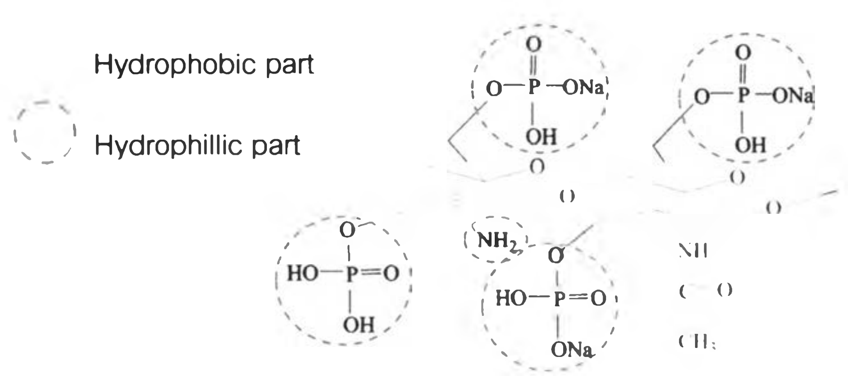

Figure 1.1 Structure of sodium phosphorylated chitosan indicating the hydrophobic and hydrophilic part

From the structure of PCTS (Figure 1.1), PCTS is an amphiphilic polymer. It contains hydrophobic at axial plane of main chain and hydrophilic side groups. The hydrophobic and hydrophilic balance can be adjusted simply by varying degree of deacetylation of chitosan. Molecules with high DD promoted the formation of O/W emulsion, whereas those of lower DD promoted the formation of W/O emulsion [6,7]. Moreover, PCTS is amphoteric polymer as well as protein due to possess both positive amino groups and negative phosphate groups in order to stabilize individual emulsion droplets. PCTS can be dissolved in water because the presence of the phosphate groups breaks the inter- and intra-molecular hydrogen bonding of chitosan [8]. Furthermore, many researches were reported about polymers modified with phosphoric groups that phosphoric polymers exhibit cytocompatibility and bioabsorbability [9-11].

Therefore, we hypothesize that PCTS should be a new attractive polymer for using as emulsifier, i.e. reactive agents encapsulators, especially for pharmaceutical and drug delivery system, including in personal care products.

1.2 Objectives

In order to reach our hypothesis, the objectives of this work were divided into two sections. First, the emulsifying properties of PCTS, and physical mechanism for unstable emulsion using PCTS as emulsifier were investigated. Second, effect of systematic conditions, viz. pH, ionic strength, temperature, to disturb the stabilization of emulsion prepared by using PCTS as emulsifier was studied.

1.3 Scope and thesis organization

Owing to its properties as described above, PCTS was chosen in this study. The scope of this works can be divided into 2 parts. Emulsifying capability of PCTS, emulsifying stability and physical mechanism of unstable emulsion were described in Part 1. Emulsifying capability was determined in terms of Hydrophilic-Lipophilic Balance (HLB), Critical Micelle Concentration (CMC), emulsion type evaluation, and emulsifying efficiency. HLB and CMC are important factors for emulsifier because HLB indicates the balance of strength of hydrophilic and lipophilic moiety in emulsifier molecules, while CMC indicates the lowest concentration of PCTS to form stable micelle. Then, emulsifying stability was studied in terms of storage appearance alteration and droplet size alteration. In order to explain the mechanism of unstable emulsion in detail, physical mechanism of unstable emulsion was investigated by observing the unstable emulsion droplets though confocal laser scanning microscope, and by evaluating the reversibility of stable-unstable emulsion under simple pH variation. The reversibility of stable-unstable emulsion was considered in terms of zeta potential, droplet size, and droplet size distribution under pH variation.

Systematic conditions (pH, ionic strength and temperature) influenced to destabilization of the emulsion were investigated in Part 2. Emulsion was prepared using 1%w/v PCTS as emulsifier and diluted in various environmental pH, ionic strength, and temperature. The physical mechanism was studied to explain the destabilized phenomena for each systematic condition.

Chapter 2 provided the background information related to the scope of this thesis. This chapter presented a brief introduction of emulsifier, nano-emulsion, mechanism to stabilize emulsion, mechanism to destabilize emulsion and literature review for this thesis.

Chapter 3 provided experimental methods in detail which were divided into 2 parts. The emulsifying capability, emulsifying stability, and physical mechanism of unstable emulsion were described in Part 1. While systematic conditions and methods viz. pH, ionic strength, and temperature providing to investigate the physical mechanism of destabilized emulsion were described in Part 2. Characterization including HLB, CMC, emulsion index, macroscopic observation, zeta potential, droplet size, and droplet size distribution, were also provided in this Chapter.

Chapter 4 described the results and discussion. Emulsifying capability, emulsifying stability, and physical mechanism of unstable emulsion were discussed in Part 1. While systematic conditions such as pH, ionic strength and temperature, were set to investigate the effect of destabilization of emulsion. Physical mechanism using to explain destabilized phenomena was discussed in detail in Part 2.

Finally, Chapter 5 provided conclusions of this work.