# Formation of wormlike micelle structure in phosphatidylcholine aqueous mixture

博士学位論文内容要旨

# Abstract

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In past decade, wormlike micelles have drawn intention in basic research and application due to their distinctive characteristics. Wormlike micelles are defined as flexible, long cylindrical, and polymer-like structure showing viscoelastic properties. These wormlike micelles are formed by the self-assembly of the surfactant molecules in both aqueous media; called normal wormlike micelle, and non-polar organic solvents; called reverse wormlike micelle. Due to the formation of wormlike micelles in either aqueous or non-polar media, they can be applied in various field including oil field applications, personal care products, biosensors, tissue engineering and drug delivery systems.

Normal wormlike micelle can be formed by several types of surfactants including nonionic, cationic, zwitterionic surfactants. On the other hands, reverse wormlike micelle has been reported in systems containing lecithin and organic solvent or oil with small amount of water.

Lecithin or phosphatidylcholine (PC) is one of phospholipids containing choline as a head hydrophilic group. Phosphatidylcholine composes of two fatty acids in molecule; while, lysophosphatidylcholine (LPC) has one fatty acid at sn-1 position of glycerol backbone giving LPC has more hydrophilicity than PC

We observed the gelation of mixed phosphatidylcholine in aqueous solution was found at high temperature. We assumed that gelation caused by wormlike micelle structures of two different self-assembly structures of phosphatidylcholine; therefore, the objective of this study is to elucidate how wormlike micelles of phosphatidycholine aqueous mixtures are formed. In this study,

pulsed-field-gradient nuclear magnetic resonance (PFG-NMR), differential scanning calorimetry (DSC) and rheological experiment were perform to evaluate the effect of temperature and total concentration of phosphatidylcholine as well as the ratio between phosphatidylcholine and lysophosphatidylcholine on the formation of wormlike micelles.

In first experiment, the viscosity of individual phospholipids; LPC or PC, were lower than 2 mPa·s which close to that of water. The mixtures of two phospholipids at total concentrations ≥56 mM showed a marked maximum in viscosity around LPC molar fraction 0.5 – 0.7, which was ascribed to wormlike micelle formation. In addition, the results indicated that a minimum concentration of both LPC and PC were prerequisite for formation of phosphatidylcholine wormlike micelles in aqueous solutions. The diffusion coefficient of the phosphatidylcholine, as measured using pulsed-field-gradient stimulated spin echo NMR, suggested that LPC and PC form spherical micelles and vesicle structures, respectively. Individual spherical micelles of LPC and a vesicle structure of PC were found at low temperature in the mixtures of LPC and PC. The formation of wormlike micelle is reflected in a steep decrease of the diffusion coefficient at temperatures above 55°C.

In second experiment, more details on thermally induced phosphatidylcholine wormlike micelle were given. Wormlike micelle can be formed after the melting of crystalline like structure in both LPC and PC at higher 40°C detected by DSC. Line width at half maximum in NMR showed the steep increased indicated the restricted in alkyl chain after wormlike micelle formation. The diffusion coefficient of mixtures, as measured by PFG-NMR, also showed the decrease after wormlike micelle structure was formed. For rheological response, the formation of wormlike micelle causes the mixtures switch from low viscosity, Newtonian fluid to viscoelastic, shear-thinning fluid.

Overall, this finding proposed the method in creating wormlike micelle structure by mixture of two surfactants, more hydrophilicity with *CPP* less than 1/2 and the other with *CPP* higher than 1/2. The better understanding in self-assembly on the formation of phosphatidylcholine wormlike micelles could help fabricating the structure for food matrix, delivery systems or biomimetic materials.