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学 位 論 文 要 旨
Dissertation Abstract

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学位論文題目：
Title of Dissertation

Study of O/W Nano-Emulsion Formation Using an Isothermal Low-Energy Emulsification Method in Food System
（食品系における一定温度下での低エネルギーな乳化方法を用いたO/Wナノエマルションの調製に関する研究）

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Nano-emulsions can be fabricated using a number of different approaches, which are usually categorized as either high-energy or low-energy methods. However, the preparation of nano-emulsions from food ingredients such as vegetable oil using isothermal low-energy emulsification methods has rarely been reported. On the other hand, it is reported that a mixture of polyglycerol polyricinoleate (PGPR) and polyglycerol laurate has a great emulsifying capacity, and consequently, fine oil-in-water (o/w) emulsions can be formed, without mechanical energy, such as homogenizer. However, there have been few reports of PGPR being used to prepare o/w emulsions without the patent. The role of PGPR in making a fine emulsion for a mixture system with polyglycerol fatty acid ester (PGFA) is also not clear.

The objective of this research was to clarify the emulsification mechanism of PGPR in the mixture system, and furthermore, to investigate the applications of o/w nano-emulsion formation as an isothermal low-energy emulsification method in food system.

Phase diagrams were constructed to elucidate the optimal process for preparing fine emulsions. In all the systems examined in this study, the phases, including the liquid crystal phase (Lc) and sponge phase (L_3), spread widely in the phase diagrams. We examined droplet size of the emulsions prepared from each phase and found that o/w nano-emulsions with droplet sizes as small as 50 nm were formed by emulsifying either from a single L_3 phase or a two-phase region, Lc + L_3. These results indicate that L_3 or Lc or both is necessary to form an o/w nano-emulsion whose average droplet diameter is less than 50 nm for PGPR and PGFA mixtures used as surfactant.

In chapter 2, we investigated how phase behavior changes by replacing water with glycerol in water/mixture of PGPR and hexaglycerol monolaurate (HGML) /vegetable oil system, and studied the effect of glycerol on o/w nano-emulsion formation using an

isothermal low-energy method. In the phase behavior study, Lc + L_3 expanded toward lower surfactant concentration when water was replaced with glycerol in a system containing surfactant HLP (a mixture of PGPR and HGML). O/W nano-emulsions were formed by emulsification of samples in a region of Lc + L_3. In the glycerol/surfactant HLP/vegetable oil system, replacing water with glycerol was responsible for the expansion of a region containing Lc + L_3 toward lower surfactant concentration, and as a result, in the glycerol/surfactant HLP/vegetable oil system, the region where o/w nano-emulsions or o/w emulsions could be prepared using an isothermal low-energy emulsification method was wide, and the droplet diameter of the prepared o/w emulsions was also smaller than that in the water/surfactant HLP/vegetable oil system. Therefore, glycerol was confirmed to facilitate the preparation of nano-emulsions from a system of surfactant HLP.

We attempted to prepare β-carotene o/w emulsions using isothermal low-energy emulsification method, with surfactant HLP as an emulsifier. β-Carotene emulsions prepared in this study had droplet sizes ranging from 40 to 100 nm. It was therefore confirmed that nano-emulsions containing β-carotene can be prepared by using this emulsification method without high-pressure homogenization. When weight ratio of surfactant to oil was higher, prepared β-carotene nano-emulsions had smaller droplet sizes and more transparent appearance. It should be noted that even nano-emulsions prepared using lower surfactant concentrations had relatively small average droplet diameters (ca. 100 nm), narrow distributions (PDI < 0.1), and monomodal particle size distributions. Moreover, we examined the influence of storage time and temperature on stability of β-carotene nano-emulsions, and it was confirmed that β-carotene nano-emulsions prepared using this emulsification method had relatively high storage stability.

These results suggest that isothermal low-energy emulsification method, with surfactant HLP as an emulsifier is suitable for preparation of β-carotene nano-emulsions, which may be important for some commercial applications.