INFLUENCE OF POLYMER–SURFACTANT INTERACTION ON THE MICROCAPSULE FORMATION

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Water soluble nonionic derivatives cellulose like hydoxypropylmethyl cellulose (HPMC) is physiologically harmless and is preferred for use in pharmaceutical formulations and the foodstuffs In commercial products, besides polymers, some other components like low–molar–mass surfactant are often present. Interaction between polymer and surfactant could change the adsorption layer around the oil droplets effecting emulsion stability and hence possibility for film formation, Sovilj V. et al. (1993), Kelley D. et al. (2003). The properties and structure of surfactant–polymer complexes depend on the molecular characteristics of polymer and surfactant, Goddard E. D. (2002,1993).

In this paper, the influence of surfactant–polymer interaction on the emulsion properties and possibility for microcapsule formation has been investigated. Emulsion was prepared dispersing sunflower oil in the mixture of nonionic polymer, HPMC, and ionic surfactant, sodium dodecylsulfate (SDS). The emulsions viscosity, stability, particle size and particle size distribution, depending on various mechanisms of HPMC–SDS interaction, were measured. After preparation, the emulsions were spray dried to obtain the microcapsules.

MATERIALS AND METHODS

Hydroxypropylmethyl cellulose (HPMC), pharmaceutical grade, obtained from Colorcon Ltd., England, was used without further purification. Sodium dodecylsulfate (SDS), purity > 99%, was obtained from Merck, Germany, with critical micelle concentration of 0.244%, determined by conductometric titration at 20°C, Sovilj V. (1998). As dispersed phase sunflower oil ("Sunce" – Sombor) was used.

The emulsions were prepared by dispersing 20% (w/w) of sunflower oil in the 1% (w/w) HPMC solution containing various SDS concentrations (0, 0.15, 0.25, 0.35, 0.45, 0.55, 0.75, 1.0, 1.5 and 2.0%) at 20°C, by means of homogenizer Ultraturrax T–25 (Janke&Kunkel, Germany). The emulsions were left for 24 h to reach equilibrium.

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Rheological measurements were performed using rotational viscometer RV20 ("Haake" – Germany) at 20°C. Continuous loop method was applied, Djaković Lj. et al. (1990). Particle diameters of emulsions were measured from microphotography using Qwin software and expressed as volume–surface mean value, Kelley D. et al. (2003). For stability tests, the emulsions were transferred into the graduated cylinder and stored for two months. The extent of creaming was characterized by a creaming index H (%). The higher the creaming index, the worse is emulsion stability.

Microencapsulation was performed by spray drying of the emulsion in a Mini Spray Dryer (Büchi, Switzerland). The microcapsules were obtained in a powder form.

RESULTS AND DISCUSSION

The concentrations of SDS corresponding to certain mechanisms of HPMC–SDS interaction were previously determined by rheological measurements of 1%HPMC solution in presence of various SDS concentrations, Sovilj V. et al. (2003). It was determined that HPMC–SDS interaction starts at 0.15% SDS concentration and finishes at 1.0%. At the SDS concentration of 0.15%, SDS molecules start to bind on the HPMC and cause the increase in viscosity to the maximum one and after that decreases until reach constant value at the end of interaction, Goddard E. D. (2002), Hormnirun P. (2000). To investigate the influence of HPMC–SDS interaction on the emulsion properties, the SDS concentrations are chosen to cover the region before, during and after interaction. The changes of apparent viscosity ha of emulsions at characteristic SDS concentrations are presented on Fig. 1. During the interacting region the viscosity changes are pronounced. Creaming index H, which is measure of emulsion stability, is calculated after 7 days of storage and are presented also on Fig. 1. It is evident that stability of emulsions depends on the HPMC–SDS interaction and follows viscosity changes of emulsion Kelley D. et al.(2003). More stable emulsions are formed at SDS concentrations during the interacting region.

The mean diameters as well as polidispersity of emulsions decreases with increasing SDS concentration (from d_{vs} = 11.3 μm to 2.73 mm, at 0.35% SDS). After that the values (d_{vs} and polydispersity) are almost

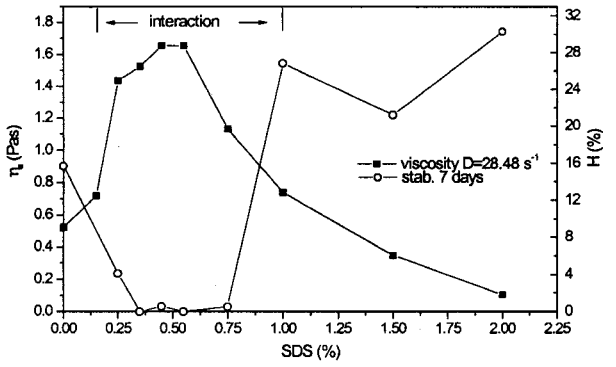
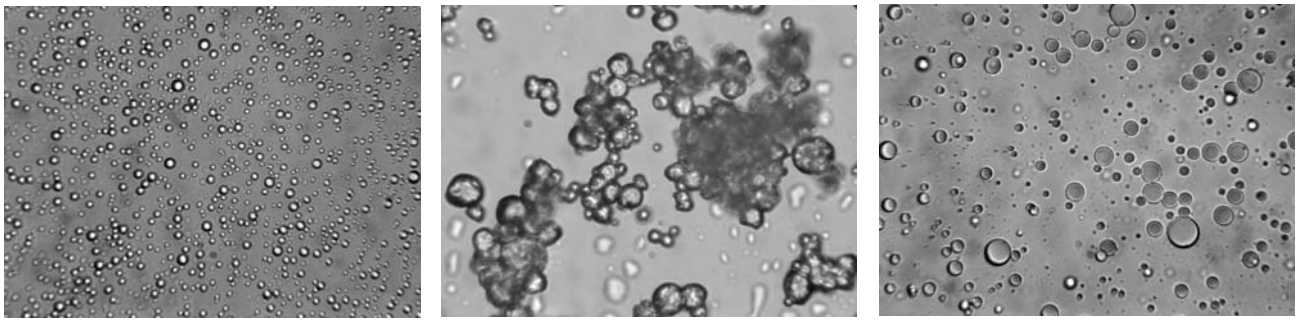


Figure 1. Changes of apparent viscosity η_a of emulsions and creaming index, H, with SDS concentrations.

CONCLUSIONS

At the SDS concentrations at which HPMC–SDS interaction in continuous phase takes place, viscosity of emulsions rises with SDS concentration, reaches maximum, and then decreases. The stability of emulsions after 7 days is increased during the interacting region due to increasing viscosity of continuous phase and stable film forming around the oil droplets. Particle mean diameter of emulsion decreases with increasing SDS concentration and is not influenced by HPMC–SDS interaction. The possibility for microcapsules formation and their stability is pronounced in the interacting region.



a) b) c)

Figure 2. Microphotography of 20% emulsions of sunflower oil in 1% HPMC and 0.75% SDS (a) obtained microcapsules (b) dispersion of microcapsules in water (c).

constant, independent on SDS concentration and it could be expected that emulsion are more stable. That is not true because of decreasing viscosity of continuous phase and changes of adsorption layer structure due to HPMC–SDS interaction. The structure of adsorption layer around the oil droplets is important for microcapsules formation, Sovilj V. (1993, 1996).

Spray drying of the emulsions gave white powder of microcapsules in the form of aggregates at all SDS concentrations. Oil leaching immediately after preparation is determined at microcapsules obtained from emulsion at SDS concentrations before and after interaction. It could be expected because before interaction only HPMC molecules are adsorbed on the oil droplet. On the other hand, after interaction on the oil surface only SDS molecules are adsorbed and HPMC is solubilized with SDS molecules and desorbed from the oil surface. In that case, possibility for stable microcapsule formation is failed. At the interacting region formed HPMC–SDS complexes are surface active and intermolecular connecting with SDS molecules forming stable film around the oil droplets. The formed microcapsules are stable and have good redispersibility in water. Redispersibility ability increases with increasing SDS concentrations, because of electrostatic repulsion between charged groups of SDS molecules. Photomicrograph of emulsion, stable microcapsules and dispersion of microcapsules in water are presented on Figure 2.

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