Microencapsulation of cinnamon oil for antimicrobial coating application

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INTRODUCTION

Nowadays, it gets more and more important that packaging can play a big role in extending of food shelf life, for example by incorporation of active substances into the structure of packaging. However, some active compounds (e.g. antimicrobials) are sensitive to external conditions such as high temperature and shearing during the processing (Coma et al., 2008). To protect the quality of food, direct coatings on food or on packaging material are applied, that avoids active substances to be exposure to high temperature and shear force (Coma, 2008). The antimicrobial function can be achieved using bioactive substances such as essential oils, which contain volatile antimicrobial compounds. In this research work, various active formulations, in form of stable emulsion based on whey protein (WP), gum arabic (GA) and cinnamon essential oil that can be applied as coating on cellulosic food packaging material have been evaluated.

The antimicrobial activity of essential oil and extracts has been recognized for many years with wide spectrum of application in food preservation, pharmaceutical and natural therapies since 19th century (Lopes et. al, 2011; Kapoor, 2008; Cooke, 2000). The quality and quantity of Essential oils (Eos) varies depending on the harvesting seasons, plant part (eg. leaves, seeds, roots), biotope, and also methods of obtaining of essential oils because of different composition (Papachristos et al. 2004; Faleiro et al, 2002; Marino et al, 1999; Juliano et al, 2000). Cinnamon is a commonly used spice in food industry and there are its various species around the world. Some previous studies proved that Chinese cinnamon (*Cinnamomum cassia*) essential oil with key component cinnamaldehyde show higher antimicrobial activity than other kinds of cinnamon species (Pritam, 2013). The encapsulation of drugs, flavors, fats and others by complex coacervation with whey protein (WP) and gum arabic (GA) was reported previously. However, different core ingredients may have different influence on capsule formation (Weinbreck et al., 2004). The WP and GA form complexes by electrostatic interaction, which is mainly affected by ionization degree of WP.

MATERIALS AND METHODS

Cinnamon oil extracted by steam distillation is purchased from Jiangxi global natural spice co. Itd, China. It has density 1.05g/ml, and contains more than 80% cinnamaldehyde (yellow color). It is soluble in ethanol and acetone. Whey protein 80% (Davisco) has been obtained from sweet dairy whey. The product is a homogeneous, free flowing, semi-hygroscopic powder with a bland flavor. Gum Arabic E-414 (JAR, Poland), a mixture of polysaccharides obtained from Acacia Senegal tree. It occurs as a white powder with pale yellow or amber lumps. It is highly soluble in water and various aqueous solutions even up to 500 g/ l.

Whey protein (Davisco) dispersion was prepared in demineralised water with final concentration 4% w/w, using magnetic stirrer, at room temperature. 4% gum arabic (JAR, Poland) dispersion was also prepared with the same procedure. The pH of both solutions was adjusted to 4.0 or 3.5 using aqueous HCl solution. The ultrasonic processor UP400S (400 W, 24kHz) (Hielscher, Germany) was used to obtain emulsion system with oil-in-water microcapsules. Firstly, cinnamon oil was slowly added into whey protein dispersion (drop by drop) and ultrasonicated for 3min. Secondly, arabic gum dispersion was added slowly into WP/Oil mixture and ultrasonicated for 3min. Different ratios of WP/GA/EO materials (Tab. 1), and different pH were prepared in order to find the optimum conditions of emulsion formation with the highest stability.

Table 1. Different ratio of WP/GA/EOs with different pH

P. No.	WP	GA	EO	pH	Notes*
I	40g	20g	10g	4.0	WP/GA=2:1 (Weinbreck, 2004)
II	40g	20g	6g	4.0	WP/GA=2:1 (Weinbreck, 2004)
III	40g	20g	4g	4.0	WP/GA=2:1 (Weinbreck, 2004)
IV	40g	20g	6g	3.5	
V	30g	20g	6g	3.5	
VI	60g	20g	6g	3.5	WP/GA=3:1 (Miri, 2010)

The stability of formed émulsions was evaluated using fast and simple method applied in paper coating industry, where a drop of emulsion with different concentration was placed on a piece of paper and the liquid spreading including formation of transprent oil spots was checked.

RESULTS AND DISCUSSION

Different ratio of WP/GA/EO was tested as well as different pH (3.5 and 4.0), as shown in the Table 1.

The ratio WP/GA=2:1 at pH 4.0 was reported in the literature (Weinbreck 2004). The concentration of essential oil was tested with ratio shell/core: 3:5, 1:1, and 3:2 to find a proper ratio to ensure the highest stability of emulsion. The capsule formation process and their final properties can be affected by pH and temperature (Bartkowiak, 2001).

Therefore, different ratios of all three components were evaluated using fast and simple "drop coating" method.. For coating dispersions with pH 4.0, the fast spreading of water was observed with small amount of released oil around the drop; that means that the oil was not encapsulated and the capsules were not stable. The most stable emulsion was observed in the case of samples (II) of shell/core ratio = 1:1, and 6g of encapsulated oil (10%).

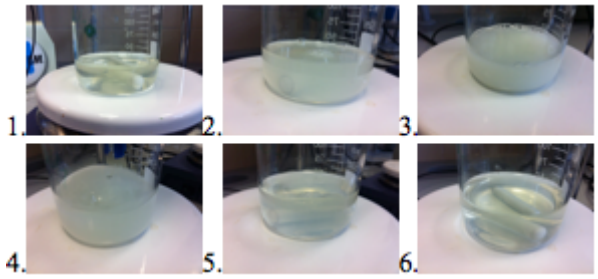

Fig 1. pH adjustment of whey protein 4% wt. solution (picture 1-6 corresonds to pH 7.0, 5.6, 5.0, 4.0 3.8, 3.5).

Protein solubility and denaturization can be affected by pH, heat and protein concentration (Dissanayake, 2013). The isoelectric point (pI) of whey protein is around 4.5. During the pH adjustment process using HCl aq. solution, whey protein start to form white precipitates below pH 5.6, and solution turn again to be clear below pH 3.8, which can be seen in the picture (Fig. 1). Therefore finally pH 3.5 was selected, which is below pH 3.8, to create microcapsule emulsion.

According to the drops on paper test results, for those six samples the best sample is VI. After drop of emulsion was placed on paper, one could observe only small amount of water around precipitated capsules without any signs of released oil. After 24 hours, the emulsion is stable and the their colour is white without any change with time. For other samples, the colour of the emulsion is yellow, or it turns to yellow after 24 hours, which is the typical colour of cinnamon oil and indicates that such systems are not stable.

Therefore, only the best suspension (VI) of microcapsules was observed using optical microscope N-800M (fig. 2). The average size of stable coacervate capsules is around 1-2μm.

Due to characteristic of oils, the encapsulation methods for specific oils can be different. Some tests with other systems such as ethylcellulose coacervation using ethanol/water (Bartkowiak et al) were performed. However in that case the emulsion containing cinnamon oil wasn't stable. The reason probably is the solubility of cinnamon oil in organic solvents such as ethanol and acetone that make impossible to form stable oil-in-ethanol emulsion as the first stage of EC coacervation process.

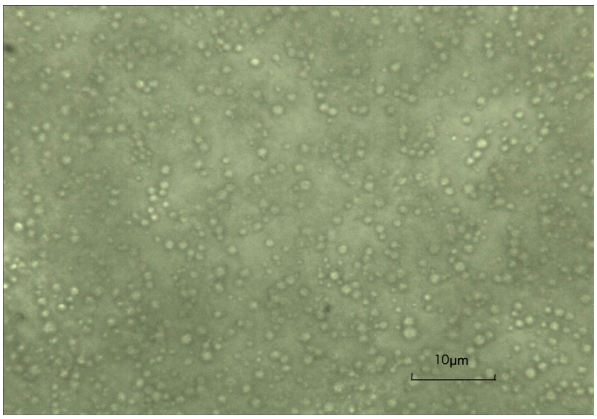

Fig 2. Microcapsules containing cinnamon oil (optical microscope, N-800M)

For further work, both emulsion rheology and antimicrobial activity of formed coating also will be studied. The activity of natural antimicrobial substances can be affected by different conditions, such as pH, temperature and film forming polymer content etc. The behavior of essential oils can differ, depending on the composition and processing methods. Burt (2004) suggests, that in food application, the effect of EOs directly present in food and in other ways such as incorporated into packaging can be different. So, further experiments will be performed to confirm the antimicrobial activity of cinnamon oil incorporated in coatings in contact with different kinds of food.

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ACKNOWLEDGMENTS

The research leading to these results has received funding from the European Union Seventh Framework Programme [FP7/2007-2013] under grant agreement n° 290098 (project NewGenPack).