Microencapsulation with gum arabic and spruce galactoglucomannan

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Introduction

Gum arabic (AG), the dried exudate from Acacia trees, is probably the most well-known and widely used capsule material in food field due to its unarguable suitability for microencapsulation purposes. AG is a highly branched and complex heteropolysaccharide (arabinogalactan) containing a small amount (~1%) of covalently bound protein (Dickinson 2003). Unlike many other polysaccharides, AG possesses excellent emulsifying properties and low solution viscosity despite its high molecular weight (Buffo et al. 2001).

During thermomechanical pulping of spruce wood, paper industry produces process water containing high amount of *O*-acetyl-galactoglucomannan (GGM) which is a predominant hemicellulose in spruce wood (Willför et al. 2008). GGM can be effectively extracted from the process water and futher spray-dried to powder using the method developed by Finnish research group (Willför et al. 2003; Xu et al. 2007). The molecular structure of GGM consists of mannose and glucose unit backbone with branches of galactose units (Xu et al. 2007). The hydroxyl groups in the mannose units are partly substituted by acetyl groups (degree of acetylation 16%). Also small amount of protein (~1%) as an impurity exists in GGM. Thus far, no commercial usage of GGM in food field exists and therefore, there is a great interest to find possible food applications for GGM. From the microencapsulation perspective, GGM could be a potential capsule material since it has many properties that are needed for encapsulation purposes such as water-solubility, emulsifying capacity and low solution viscosity (Mikkonen et al. 2007; Willför et al. 2008). Moreover, the costs to produce GGM were estimated to be reasonable (Persson et al. 2007) which makes GGM even more interesting alternative for microencapsulation.

The aims of the study were 1) to compare the capability of GGM, AG and mixture of GGM&AG (50:50) to encapsulate α-tocopherol (hydrophobic model substance) and 2) to characterize water sorption properties of GGM, AG and GGM&AG microcapsules.

Material and methods

The spray-dried GGM (39 kDa) was obtained from an industrial-scale isolation trial from process water of a Finnish pulp mill (Willför et al. 2003; Xu et al. 2007). Gum arabic (*Acacia Seyal*) used was purified and spray-dried powder and it was a commercial product (Valspray F 25500) of Valmar, France. α-tocopherol was from Merck (Darmstadt, Germany).

Capsule materials (10% w/w) were dissolved in distilled water at 60 °C. The solutions were cooled and stored in a refrigerator overnight. α-tocopherol (2% w/w) was added into the solutions and the solutions were first homogenized with Heidolph homogenizer (Diax900, Heidolph, Germany) for 2 x 2 min at 26000 rpm and then with a Microfluidizer (Microfluidics model 110y, Newton, MA) for 10 min at pressure of 500 bar. During preparation, temperature of emulsions was kept below 10 °C with a cooling ice bath. Mean droplet size of emulsion was determined using a laser diffraction particle size analyzer (LS230, Coulter, Fullerton, CA). To obtain microcapsules, emulsions were

frozen at −20 and −80 °C and then freeze-dried (72 h, pressure <0.01 mbar). Powders were stored in the dark in vacuum desiccators over P_2O_5 prior to usage.

Water sorption properties of microcapsules (~100 mg) were determined by equilibrating triplicate samples at 25 °C for 8 days in vacuum desiccators over saturated salt solutions. The desired relative humidity (RH) conditions were obtained with the following salts: LiCl (11.4%), CH_3COOK (23.7%), $MgCl_2$ (32.9%), K_2CO_3 (44.3%), $Mg(NO_3)_2$ (53.6%), $NaNO_2$ (65.9%), NaCl (76.2%) and KCl (85.5%). The water content of samples was obtained gravimetrically by daily weighing. The Guggenheim-Anderson-DeBoer (GAB) and the Brunauer-Emmett-Teller (BET) models were fitted to the experimental data as described by Roos (1993).

To analyse α-tocopherol content, microcapsules (50 mg) were broken down to release α-tocopherol from the matrices by dissolving them in distilled water containing ascorbic acid and ethanol. After adding saturated KOH, the mixture was stirred vigorously and saponified in a boiling water bath (85 °C) for 15 min. α-tocopherol was partitioned into heptane, and the content of α-tocopherol was analyzed by normal phase HPLC with an Inertsil silica column (5-μm particle size, 4.6 mm x 250 mm; Varian Chromapack, Middelburg, The Netherlands). The mobile phase (flow rate 2 mL/min) contained 3% 1,4-dioxane in heptane, and α-tocopherol was detected with fluorescence detection (FLD) using an excitation wavelength of 292 nm and emission wavelength 325 nm and quantified with an external standard method. At least duplicate analyses of each sample were performed. Encapsulation yield was calculated as: microencapsulated α-tocopherol (g/100 g solids) / calculated value of α-tocopherol in emulsion (g/100 g solids) x 100.

Results and discussion

Encapsulation yield (EY) illustrates the percentage of α-tocopherol that was encapsulated into capsule matrices. During an effective microencapsulation process, none or only minor losses of encapsulated component may exist. In our study, EY values for AG, GGM and GGM&AG were at moderate level: 59, 45 and 40%, respectively. These values revealed that encapsulation process caused losses of α-tocopherol with every capsule material, which may be related to the insufficient stability of emulsions during emulsion preparation, freezing and freeze-drying, and as a result some α-tocopherol might remain unprotected to the surface of capsule matrices.

Higher EY values were obtained with AG than GGM suggesting that AG was better emulsifier/stabilizer for α-tocopherol than GGM. This suggestion was also supported by the analysis of emulsion droplet sizes: AG emulsions had smaller droplet size (mean 2.9 μm; median 2.8 μm) than GGM&AG (mean 3.6 μm; median 3.2 μm) and GGM emulsions (mean 4.3 μm; median 3.8 μm). Generally, minimum droplet size has been related to maximum emulsion stability and encapsulation yield (Soottitantawat et al. 2003). Probably smaller droplet size and further better EY values could be obtained by using higher homogenization pressure and/or longer homogenization time while preparing emulsions (Chen and Wagner 2004). In addition, the optimal process parameters for AG and GGM emulsions are not necessarily similar. Previously, it has been shown that with the same emulsion process parameters different hydrocolloids (e.g., gum arabic, guar gum, locust bean gum) resulted in notably different emulsion particle sizes and emulsion stability behaviour (Huang et al. 2001).

Water sorption properties of food materials depend on many factors, e.g., chemical composition, physicochemical state (amorphous/crystalline) and physical structure, and are important to know for predicting potential changes in food storage stability (Labuza and Altunakar 2007). In our study, water sorption of microcapsules differed from each other (**Figure 1**). AG microcapsules adsorbed most water and GGM microcapsules adsorbed least water at the same RH which suggests more

hydrophobic nature of GGM than AG. This phenomenon may be related to the differences in molecular weights and proportion of branched structures.

Water sorption isotherms represent the steady-state amount of water held by the food solids as a function of storage RH (=water activity x 100) at constant temperature (Labuza and Altunakar 2007). All water sorption isotherms had sigmoid shapes as shown in **Figures 2–4**. The GAB isotherms were applicable for all microcapsules over the whole water activity range tested whereas the BET isotherms showed good agreement with the experimental data only up to water activity 0.44 which water activity range was used in modeling.

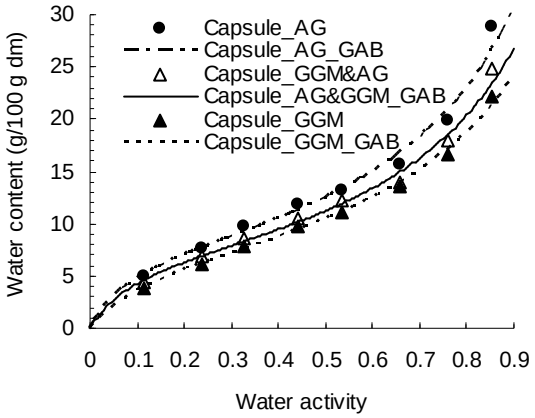

Figure 1: Comparison of water sorption of different microcapsules at 25 °

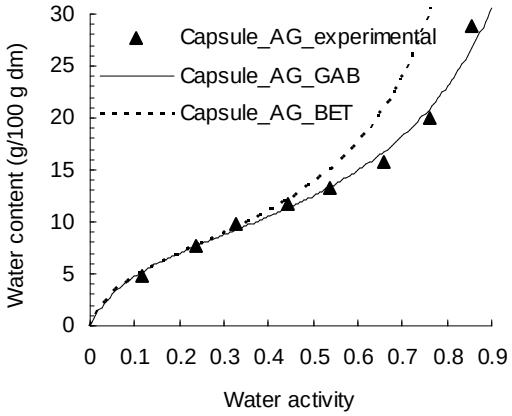

Figure 2: Water sorption isotherm of AG at 25 °C

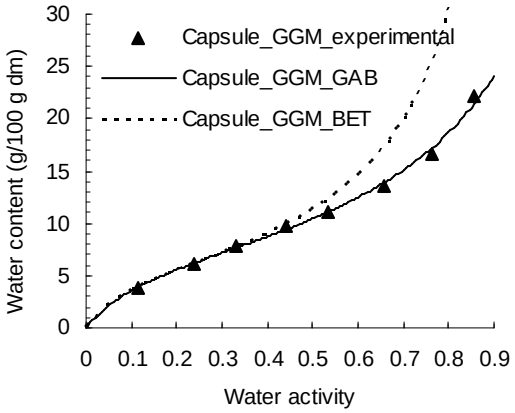

Figure 3: Water sorption isotherm of GGM at 25 °C

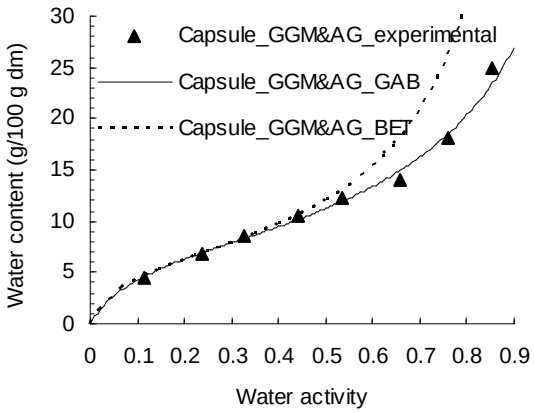

Figure 4: Water sorption isotherm of GGM&AG at 25 °C

Conclusions

The present study suggested that

1) Hydrophobic model substance α-tocopherol could be encapsulated to AG, GGM and GGM&AG matrices. However, further research is needed to improve encapsulation yield. Higher encapsulation yield may be obtained by optimizing the parameters in emulsion preparation process (e.g., the homogenization time and pressure and a ratio of capsule material to encapsulated component) for each capsule material.

2) Water sorption of microcapsules differed from each other -AG microcapsules adsorbed slightly more water compared to GGM microcapsules. The GAB water sorption isotherm was in agreement with experimental data for all microcapsules over the whole water activity range tested.

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