P-060	**Encapsulation of Oil by Inverse Gelation**	

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INTRODUCTION

Many methods have been used to encapsulate oil, including spray drying (Tan 2005), extrusion (Yilmaz 2001), and coecervation (Bachtsi 1996). However, some of the methods may need a control of heat and pH, involves volatile materials that are hazardous to health, as well as using synthetic polymers as encapsulating matrices that are unlikeable for oral consumption. In this study, an alternative method was developed to produce oil-core capsules by dropping calcium-containing oil into alginate solution.

MATERIALS AND METHODS

Preparation of calcium chloride-in-oil emulsion
Calcium chloride solution (Panreac Quimica Sau, Spain) was prepared at concentrations of 40g/L, 80g/L and 120g/L. 80 ml of calcium chloride solution was mixed with 200 ml of sunflower oil containing non-ionic surfactant with an ultra-turrax at 13,500 rpm for 10 min to form water-in-oil emulsion.

Preparation of oil-core alginate capsules by gravitational dripping method
Sodium alginate powder, Algogel3001 (Cargill, France) was prepared at desired concentrations of 5g/L, 10g/L and 15g/L. Tween85 (1.75 ml) was added to 350 ml of alginate solution. Droplets of emulsion were extruded through multi-nozzles into alginate solution by a peristaltic pump. The alginate solution was constantly stirred at 350 rpm. The dropping height was 80 mm to ensure the formation of spherical droplets. Dropping time of droplets was 1% of total residence time in alginate solution to ensure that capsules were formed within the same curing period. Finally, capsules were filtered and washed four-fold with water to stop the gelation. Several batches of capsules were prepared by varying the calcium chloride concentration, alginate concentration and curing time while keeping all other parameters constant.

Measurement of capsule membrane thickness
Confocal laser scanning microscope (Nikon A1), was used to measure the membrane thickness of capsules. The image analysis was done using ImageJ 1.42q software. Prior to analysis, wet capsules were labelled with fluorescence Rhodamine B by incubating in 15 g/L calcium chloride solution containing 1g/L Rhodamine B. Two wet capsules that were produced under the same experimental formulation were cut half and ten membrane thickness measurements were taken from each capsule.

Measurement of capsule Young's Modulus
Wet capsules were dried overnight at room temperature until constant weight. Uni-axial compression between two parallel plates was done to measure the resulting stress when strain was imposed on dry capsules using Dynamic Mechanical Analyzer Q800 (TA Instruments, France). Three individual capsules from each batch of formulation were tested. An individual Young's Modulus analysis of each capsule was obtained by using best-fit linear regression in the linear region of the stress-strain curve limited between 5% and 15% of strain.

RESULTS AND DISCUSSIONS

Effect of curing time

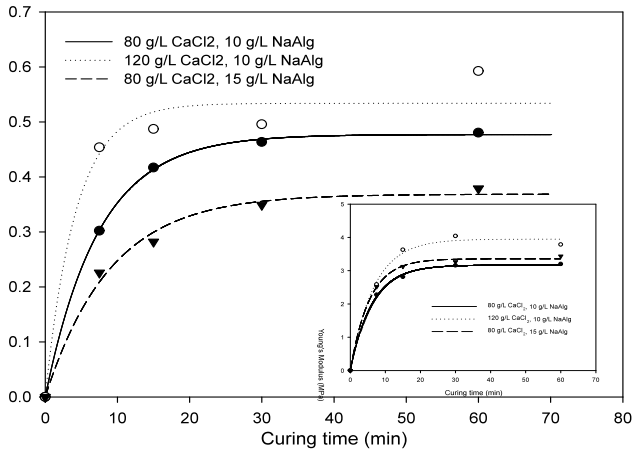

Figure 1: Effect of gelation time on mean membrane thickness and Young's Modulus of capsules

Figure 1 shows the evolution of membrane thickness of wet capsule at three experimental formulations. When the emulsion was dropped into alginate solution, thin membrane was formed around the droplets. The immediate outward diffusion of calcium ions from the surface and core of the droplets to the alginate solution has resulted in the development calcium alginate membrane. At the beginning stage, the diffusion coefficient of the calcium ions was mainly governed by the electrostatic interactions between the fixed charges on the alginate matrix and calcium ions. As the membrane thickness of the capsules increased, formation of the membrane increased slowly. This might attribute to the obstructing effect of gel matrix that hindered the diffusion of calcium ions through gel network. The calcium alginate membrane evolves continuously until

calcium ions, which diffuse from the core completely depleted. This evolution was shown in an increase in the membrane thickness of wet capsules. The effect of membrane thickness of capsules on the strength of capsules was studied using constant strain-rate compression tests on dry alginate capsules to determine their elastic (Young's) moduli. The compressive modulus of the dry calcium alginate capsules increased with curing time (Figure 1). The results may be explained by the increase in the membrane thickness of the capsules.

Influence of alginate concentration and calcium chloride concentration

The influences of varying the calcium chloride concentration in the disperse phase of emulsion and varying the sodium alginate concentration at constant gelation time of 60 min, on the wet capsules mean diameter and Young's modulus were studied individually. The results indicate that at constant alginate concentration, wet capsule mean diameter increased with an increase in calcium chloride concentration (Figure 2). As mean diameter of capsules is correlated to the membrane thickness, this means that the membrane thickness increases with an increase in the calcium chloride concentration. This may be explained in terms of calcium chloride diffusion coefficient through the gel network, where the diffusion coefficient of calcium chloride ions increases with an increase in calcium ions (Potter 1994). When the amount of calcium ions increases, more calcium ions are free to diffuse and bind with the G-block fraction in the alginate chains. This has caused an increase in binding sites and consequently an increase in the membrane thickness of the capsules. On the other hand, at constant calcium chloride concentration in disperse phase, an increase in alginate concentration decreased the wet capsule mean diameter (Figure 3). The increase in alginate concentration increased the binding sites for calcium ions and resulted in a compactly cross-linked polymer network. This closely and firmly gel structure that formed the membrane would probably cause a low membrane thickness and consequently reduced the mean diameter of capsules. Young's modulus of capsules increased with an increase in the calcium chloride concentration in the disperse phase at constant alginate concentration (Figure 2). This presumably due to that, the increase in the membrane thickness has resulted in stiff membrane layer. The influence of alginate on the strength of the capsules shows an increase with increasing alginate concentration (Figure 3). The mean diameter of capsules decreased, which also means a decrease in membrane thickness of capsules with increasing alginate concentration. This effect may logically attribute to the increase in a number of junction zones in the alginate network to cross-link with calcium ions to form stiff gel structure.

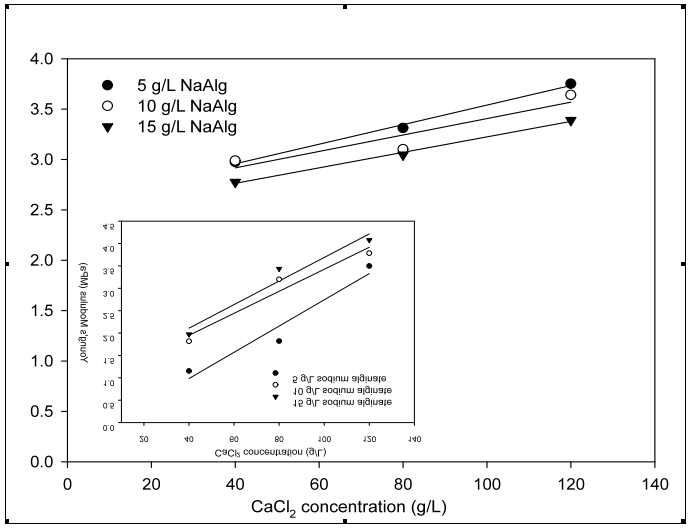

Figure 2: Effect of $CaCl_2$ concentrations on mean diameter and Young's Modulus of capsules

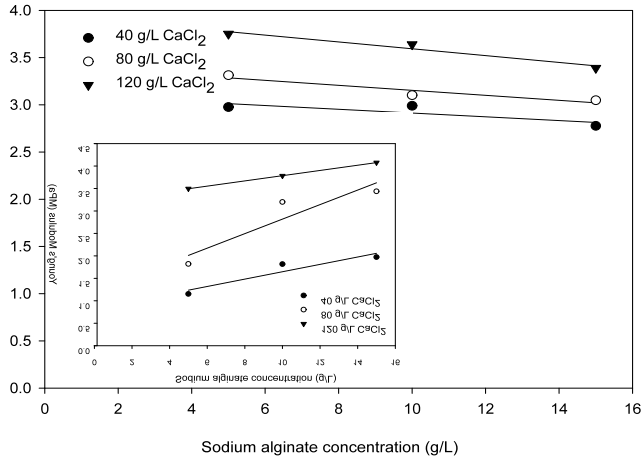

Figure 3: Effect of alginate concentrations on mean diameter and Young's Modulus of capsules

CONCLUSION

This work represents a simple, mild, and inexpensive method to encapsulate oil. Furthermore, it is also easy to customize the membrane thickness of capsules by modifying the formulation conditions; gelation time, calcium chloride and alginate concentration.

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