ULTRASONIC ENCAPSULATION OF CINNAMON FLAVOR TO IMPART STABILITY FOR BAKING APPLICATIONS

A Project Paper

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ABSTRACT

Cinnamon flavor is fairly unstable when subjected to cooking methods, specifically baking. Cinnamaldehyde, the main constituent of cinnamon that imparts flavor, is particularly volatile when subjected to high baking temperatures and is susceptible to degradation. Cinnamon is also known to have antifungal properties, making its interactions with yeast undesirable in foods such as bread. Therefore, a method is desired to protect cinnamon from both volatility and unwanted reactions. The objective of this study was to encapsulate cinnamon flavor and investigate the protective effects of the constructed capsule on heat stability and yeast interactions. Capsules were formed by ultrasonication induced cross-linking of polysaccharides chitosan and pectin. Cinnamaldehyde, the principle component of cinnamon essential oil, was incorporated with carrier oil into the polysaccharide capsules at an oil:polysaccharide ratio of 20:1 (w/w). Capsule formation was verified using Fourier-transform infrared spectroscopy and scanning electron microscopy. The formulation was spray-dried and heat studies were conducted to determine the stability of the encapsulated flavor at baking temperatures (150-250°C). Gas chromatography with flame ionization detector was utilized to quantify the amount of cinnamaldehyde remaining in heated samples. Compared to unencapsulated cinnamaldehyde and the non-sonicated formulation, the ultrasonicated capsules exhibited significantly higher cinnamaldehyde retention at high temperatures (>200°C). Thermal gravimetric analysis corroborated increased heat stability of the capsules. Yeast growth studies were also performed and yeast (*Saccharomyces cerevisiae*) displayed more growth

when subjected to encapsulated cinnamaldehyde versus pure cinnamaldehyde of the same concentration.

BIOGRAPHICAL SKETCH

Curtis Gong is a Master of Professional Studies student in Dr. Alireza Abbaspourrad's lab group. Before starting at Cornell, Curtis graduated from Northeastern University in 2017 with a Bachelor of Science Degree in Chemistry and a minor in Food Systems, Health, Equity, and Sustainability. Curtis hails from Detroit, Michigan.

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I would like to thank Dr. Alireza Abbaspourrad for giving me the opportunity to conduct research within his lab group. His motivation and expertise have been significant components of my research. I would like to thank my graduate student mentor Michelle Lee for guiding me throughout the duration of my time at Cornell. The extra effort and patience she has exercised to ensure my success is greatly appreciated. Additionally, I would like to acknowledge all the members of the Abbaspourrad lab group for offering advice, and Peter Lawrence and Mary Godec specifically for their direct assistance with my experiments. Lastly, I would like to thank Cornell University and the Food Science Department for providing me a welcoming and resourceful environment to conduct my research. My time at Cornell has been short, but I have learned and accomplished so much.

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CHAPTER 1

1. INTRODUCTION

Cinnamon is a spice derived from the bark of *Cinnamomum* trees. It is commonly utilized in cooking, particularly baking, and is commercially available in powder, stick and oil forms. Cinnamon flavor stems from its essential oil, which imparts a characteristic aroma and taste. Essential oils are extracted from source plants and can be used in various applications including food and cosmetics. The principal component of cinnamon essential oil is cinnamaldehyde, which plays a large role in the flavor definition (Tian et al., 2015). Cinnamaldehyde is volatile at high temperatures, which can present problems when cinnamon is used in applications such as baking. The elevated temperatures can cause unwanted flavor degradation and evaporation from foods (Hermanto et al, 2016). Baked goods can consequently develop unwanted off-flavors or become less flavorful and therefore less desirable. On a larger scale, companies producing food products with cinnamon at high processing temperatures may be required to increase the amount of the ingredient in the formulation in order to produce the desired flavor. Natural flavors in general can be very expensive, therefore a solution is desired to decrease the ingredient quantity, especially at industrial levels, while still retaining full flavors.

Cinnamon has been known to exhibit antibacterial and antifungal properties. This has been exploited in applications such as pharmaceuticals and food packaging (Ali et al., 2005; Sanla-Ead et al., 2012). While beneficial in medicinal some food settings, these properties can be inhibitory during baking. Yeast is used as a leavening agent in various baked products for its ability to produce carbon dioxide which assists in rising and texture development. In baked

goods such as cinnamon rolls and breads, the antifungal properties of the cinnamon may inhibit the yeast's ability to produce gas, leading to unrisen, denser breads (Pattison & von Holy, 2001).

In a baking or cooking setting, the functionality and efficiency of cinnamon can be increased by protection of the flavor itself. Encapsulation is a method of flavor protection that can impart stability to cinnamon at high baking temperatures (Cevallos, Buera, & Elizalde, 2010; Hermanto et al., 2016; Petrovic, Stojanovic, & Radulovic, 2010). Capsule wall materials provide a barrier that heat must penetrate before reaching the encapsulated flavor. In this study, encapsulation also provides a physical barrier for cinnamon flavor that can weaken the antagonistic reactions that cinnamon has with yeast. By encapsulating cinnamon flavor, baked goods can subsequently retain that flavor longer and produce goods of desired structure and texture.

There are several approaches to encapsulation. The formation of emulsions is common when formulations contain immiscible layers. Emulsions impart stability to the formulation in addition to simultaneously affording capsule formation (Borodina et al., 2014; Hermanto et al., 2016). They require energy to form, be it through chemical or physical means. Application of mechanical energy is often used in unstable systems to form emulsions. Examples of mechanical methods include homogenization and ultrasonication. Ultrasonication is particularly useful in supplying energy to form emulsions and capsules. Specifically it is the application of sonic waves of frequencies greater than 20 kHz in order to produce agitation. This agitation produces cavities or pockets of gas or liquid within the system that are capable of implosion. The release of energy generates free radicals that assist in the bonding of capsule materials and the overall capsule formation (Borodona et al., 2014; Hashtjin & Abbasi, 2015). In a colloidal system, hydrophobic ingredients, such as cinnamon flavor, can concurrently be incorporated into the

newly formed capsules. Both the emulsion and construction of capsules lend stability to the encapsulated ingredient.

This study presents a formulation to encapsulate cinnamon flavor via ultrasonication. The emulsified microcapsules are constructed from polymeric crosslinking of two polysaccharides: chitosan and pectin. These food grade materials are commonly used ingredients in various food applications. In the presence of cavitation and free radicals, chitosan and pectin in aqueous solution are well-suited to form linkages and encapsulate the hydrophobic cinnamaldehyde. The accessibility of amine groups in chitosan and carboxyl groups in pectin can provide the basis for amide bond formation. The strength of these linkages supplies stability to the polysaccharide complex and to the capsule as a whole. This formulation was investigated for its ability to impart heat stability to cinnamon flavor in addition to restricting the flavor's undesirable interactions with yeast for baking applications.

CHAPTER 2

2. MATERIALS AND METHODS

2.1 Materials

Materials and reagents were commercially available and purchased: Trans-Cinnamaldehyde (99%) (Acros Organics), chitosan (medium molecular weight) (Sigma Aldrich), Pectin (from citrus peel) (Sigma Aldrich), vegetable oil (Wegmans Food Market, Ithaca, New York), maltodextrin (bulksupplements.com), hydrochloric acid (VWR) and ethyl acetate (EMD).

2.2 Emulsion and Capsule Preparation

Aqueous solutions of chitosan (0.5% wt.) and pectin (0.5% wt.) were prepared and adjusted to pH 2 with hydrochloric acid. Cinnamaldehyde and vegetable oil were mixed at a 7:3 (w/w) ratio to comprise the total oil phase. The aqueous solutions (equal volumes of chitosan and pectin solution) were layered onto the oil mixture at a 1:20 dry ratio (w/w) respectively. The mixture was covered and placed in an ice bath to be ultrasonicated via ultrasonic probe (750 W, 20 kHz, 15 minutes) (VibraCell 750 Ultrasonic Processor, Sonics and Materials Inc.)

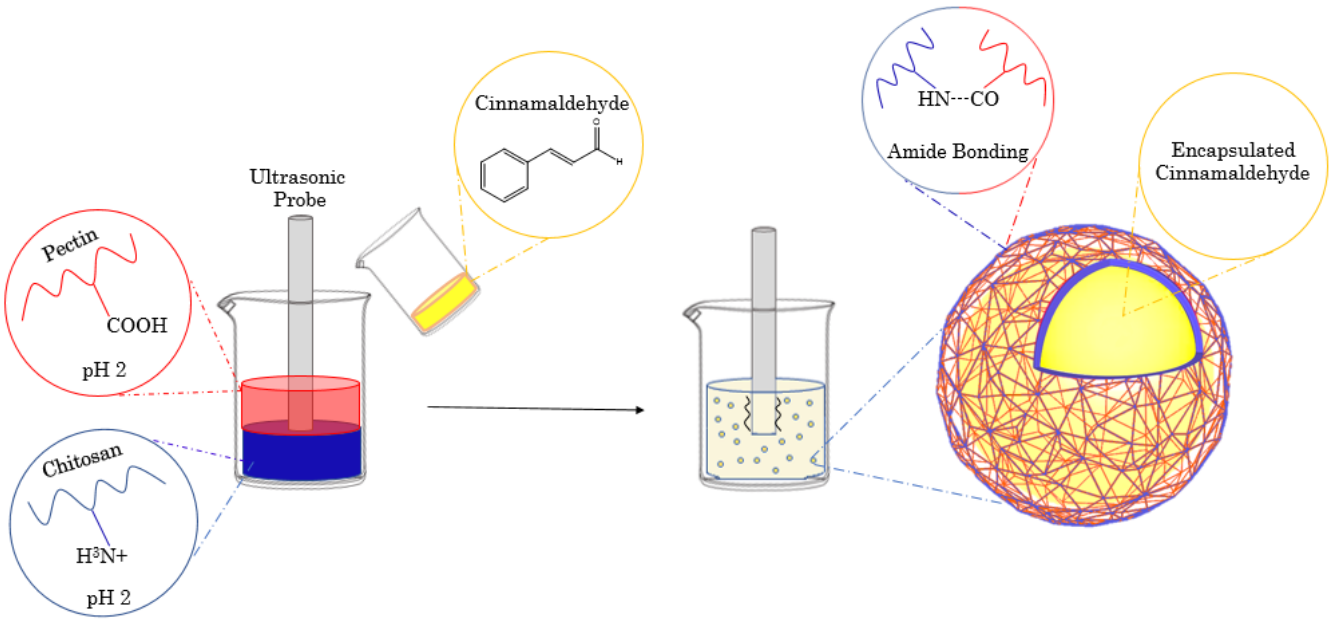

Figure 1. Cinnamaldehyde encapsulation mechanism schematic detailing capsule formation via ultrasonication induced polymeric cross-linking

2.3 Capsule Characterization

Emulsion particle sizes and zeta potentials were measured (Zetasizer Nano-ZS, Malvern Panalytical). Emulsion samples were frozen (-20°C) and freeze-dried (FreeZone Freeze Dryer, Labconco). Freeze-dried samples were utilized for Fourier-Transform Infrared (FTIR) measurements (IRAffinity-1s Fourier Transform Infrared Spectrophotometer, Shimadzu). Measurements were conducted in a range of 400-4000 cm^{-1} with a resolution of 4 in %Transmittance mode and Happ-Genzel apodization function. Emulsion samples and individual capsule components were air-dried to be examined by scanning electron microscopy (SEM).

2.4 Formulation Spray-Drying

Maltodextrin (27.5 dry wt. %) was mixed into the ultrasonicated formulations to assist in spray-drying powder formation. The emulsions were subjected to spray-drying (150°C inlet temperature, 55°C exhaust temperature) and powder was collected.

2.5 Gas-Chromatography (GC) Analysis

2.5.1 Heat Treatment

Spray-dried samples were heated in a furnace (Thermolyne Benchtop Furnace, Thermo Scientific) at 50, 100, 150, 200, and 250°C for 10 minutes each. The samples were left uncovered in order for the cinnamaldehyde to evaporate. Pure cinnamaldehyde and non-sonicated, physical mixtures of the formulation, of equal cinnamaldehyde concentration to the spray dried samples, were subjected to the same heat treatment. Heating was conducted in triplicate.

Heated samples were cooled to room temperature. Ethyl acetate (1 mL) and water (1 mL) were added to all samples for extraction. Samples were vortexed for 1 minute, sonicated for 10 minutes, and vortexed for an additional 1 minute. 100 μL of the organic layer of each sample were added to vials for GC-analysis.

2.5.2 Gas Chromatography-Analysis

Gas chromatography with flame ionization detection (Hewlett Packard 5890 Series II Gas Chromatograph with Flame Ionization Detector, GenTech Scientific) was utilized to analyze the samples. 0.8 µL of each sample was injected with an initial column temperature of 60°C ramped to 250°C at a rate of 10°C/min. The injector temperature was 250°C, with an inlet pressure of 17 kPa. Hydrogen was utilized as the carrier gas. Two injections were performed for each sample. Data was collected and analyzed via PeakSimple Chromatography Data Systems (SRI Instruments).

2.6 Thermogravimetric Analysis

Thermogravimetric analysis of pure cinnamaldehyde, spray-dried, encapsulated cinnamaldehyde, and capsule components, were conducted. A temperature range of 25-600°C was observed.

2.7 Yeast Growth Studies

Saccharomyces cerevisiae was cultured for yeast growth experiments. Stock solutions of encapsulated and unencapsulated cinnamaldehyde (6.3 mg/mL) were produced. To 50 µL of yeast, 100x (990 µL yeast peptone dextrose (YPD), 10 µL cinnamaldehyde stock) and 200x (950 µL YPD, 5 µL cinnamaldehyde stock) dilutions of both stocks were added to separate wells of a 24-well plate. Untreated yeast was plated as a control. Three replicates were conducted for each treatment. Negative controls of each isolated stock and YPD were plated. The plate was analyzed by plate reader (SpectraMax iD3 Microplate Reader, Molecular Devices) for 15 hours and absorbance was measured.

CHAPTER 3

3. RESULTS AND DISCUSSION

3.1 Emulsion and Capsule Formation

Chitosan and pectin were chosen as polymer materials for the fabrication of emulsion capsules because of their availability of functional groups, namely amine and carboxyl groups respectively, which participate in ultrasonication-induced polymer cross-linking. This is evident with the two polymers' ability to form polyelectrolyte complexes at neutral pH ranges. To prevent this from occurring before ultrasonication and the inclusion of the oil phase, the pHs of the aqueous chitosan and pectin solutions were adjusted to a pH value of 2 (Bordina et al., 2014). Upon ultrasonication of the aqueous and oil layers, high energy initiates the cross-linking reactions between chitosan and pectin. Amide bond and hydrogen bond formation construct a stable capsule structure while simultaneously incorporating the hydrophobic oil phase inside (**Figure 1**). In addition, cavitation produced from the high frequency sonic waves generates free radicals that promote these chemical reactions and facilitate the encapsulation process. This structure imparts a great degree of stability to the susceptible cinnamaldehyde. Ultrasonication generates concentrated amounts of heat, therefore emulsions were sonicated while submerged in ice and covered to reduce any evaporation of the flavor compound.

3.2 Capsule Characterization

Table 1. Particle Size and Zeta Potential Measurements of chitosan polymer capsule and encapsulated cinnamaldehyde emulsion

Sample	Mean Particle Size (μm)	Zeta Potential (mV)
Chitosan-Pectin Capsule	1.595	35.23
Chitosan-Pectin-Oil-Emulsion	12.326	44.3

Particle size and zeta potential measurements of the polymer capsules and emulsion were made (**Table 1**). As expected, the inclusion of oil in the complex increased the particle size significantly. The amount of oil included will affect the size and can also impact emulsion stability. The experimental oil amount was determined by screening various polysaccharide to oil ratios and observing which amount yielded the best cinnamaldehyde encapsulation efficiency. A ratio of 1 part dry polymer to 20 parts oil produced the best efficiency and was therefore utilized. This amount of oil is large and will produce a large particle size, however as this formulation is designed for baking applications, it is envisioned that large particles would not be an issue if the emulsion (or spray-dried powder) was dispersed in dough.

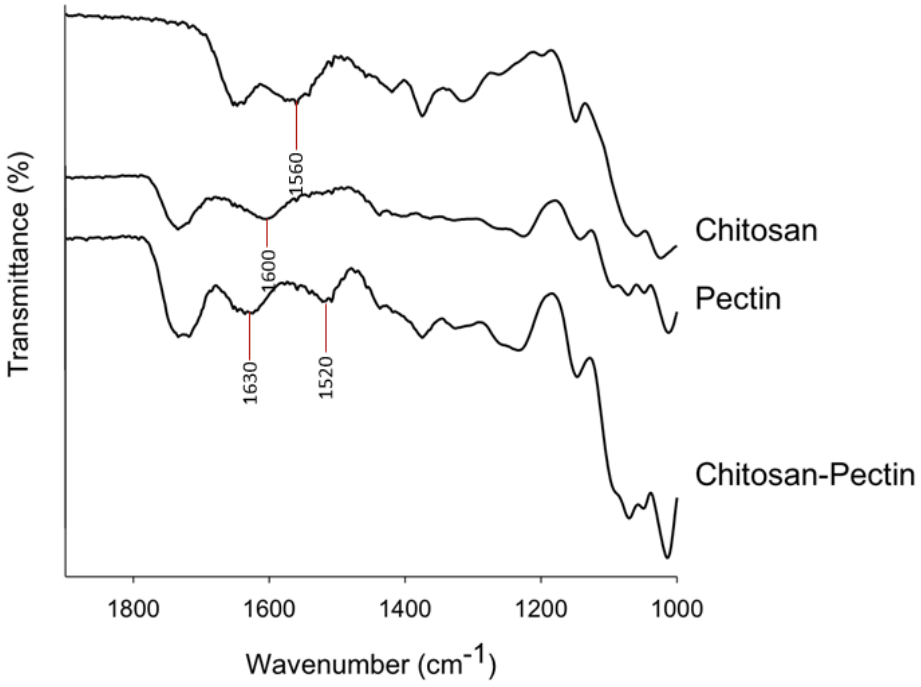

Figure 2. FTIR measurements of polymer capsules and chitosan and pectin components detailing spectral changes corroborating formation of amide bonds.

Fourier transform infrared measurements were taken of the polymer capsule and of its polysaccharide constituents in order to confirm the formation of capsules (**Figure 2**). The presence of amide bond formation is evidenced by several peaks. A new peak at 1630 cm^{-1} of the chitosan-pectin spectrum appears and is attributed to the carbonyl stretch (Amide I) of the newly formed amide bonds (Coimbra et al., 2011; Maciel, Yoshida, & Franco, 2015). The peak at 1600 cm^{-1} of the pectin spectrum correlates to carboxyl functional groups. This peak is diminished in the chitosan-pectin spectrum, suggesting the participation and subsequent depletion of the carboxyl groups in amide binding. Additionally, a shift is seen in the wavenumber of the amine bend (N-H) from the chitosan spectrum to the chitosan-pectin capsule spectrum. This decrease from 1560 cm^{-1} to 1520 cm^{-1} signifies the conversion of a primary amine, in this case the amine functional groups of chitosan, to a secondary amine, characteristic of amide bonds (Coates 2006).

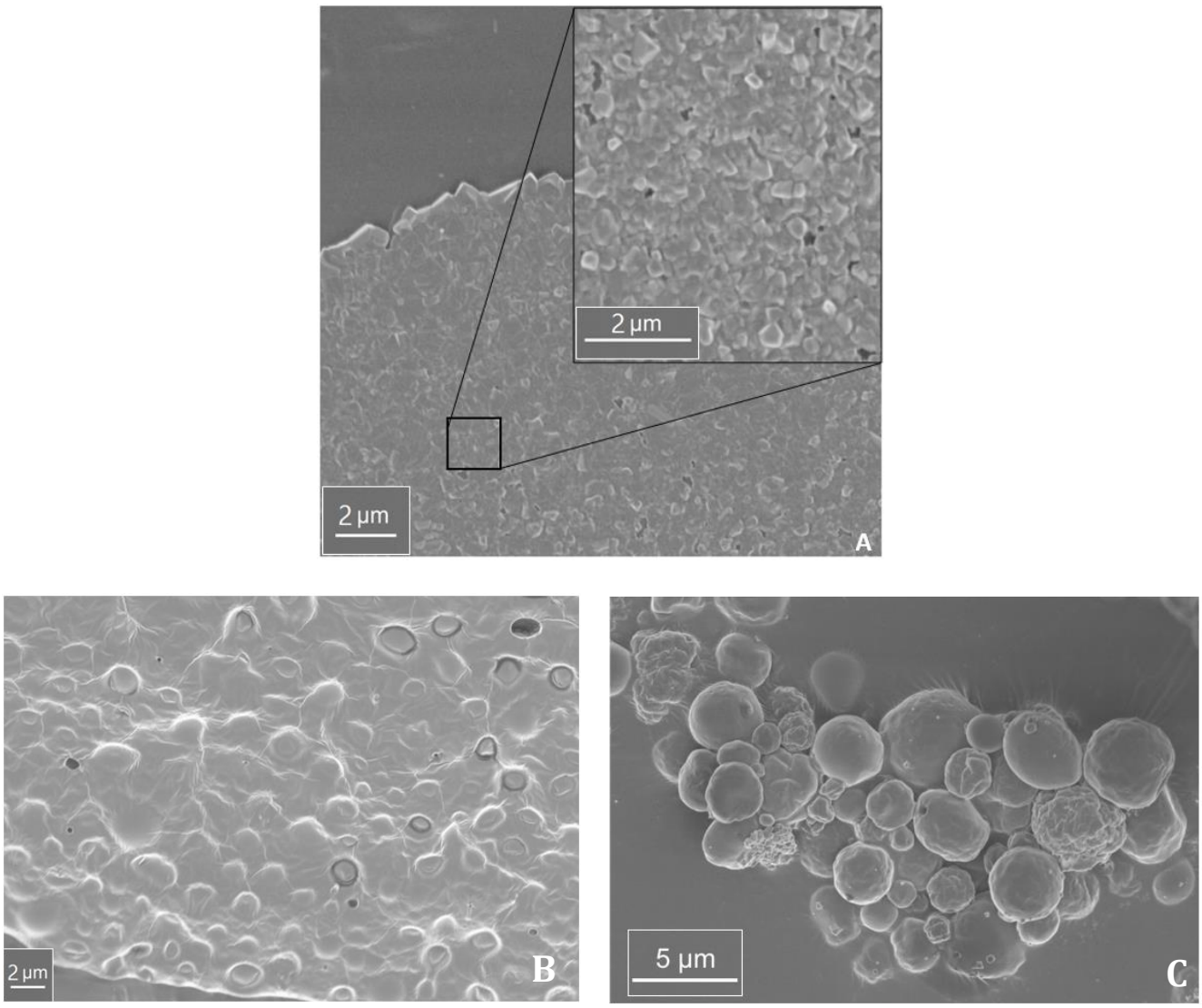

Figure 3. SEM images of chitosan-pectin capsules corroborating capsule formation and morphology: (A) chitosan-pectin capsules, (B) Encapsulated cinnamaldehyde emulsion, (C) Spray-dried encapsulated cinnamaldehyde

With the confirmation of the formation of amide bonds via FTIR, scanning electron microscope images were taken of the capsules to further corroborate the construction of spherical capsules (**Figure 3**). The samples in images (A) and (B) of **Figure 3** were air-dried, which can explain the observation of a film. However, spherical shapes seem to be imbedded into these films, suggesting capsule formation. Image (C) has clearly defined spherically capsules, again confirming the encapsulation process.

3.2 Gas Chromatography Analysis

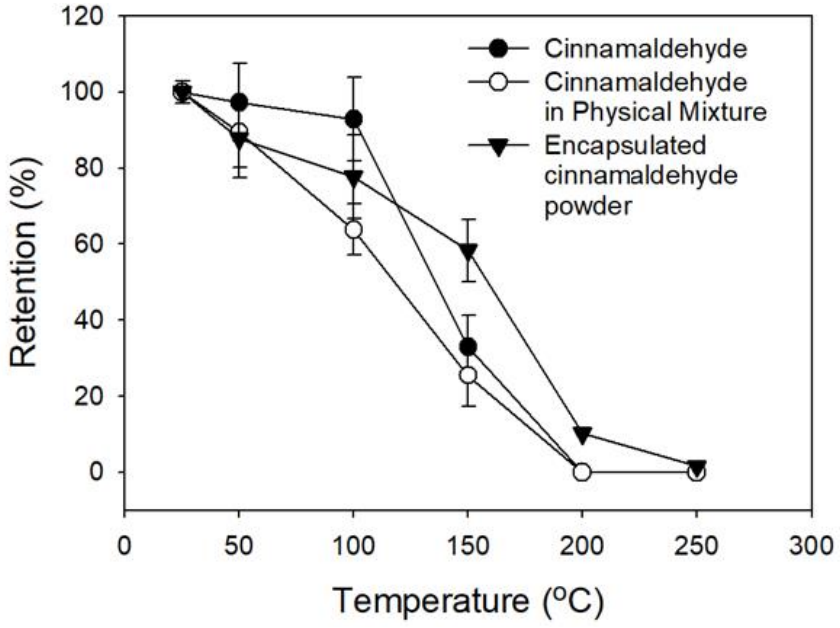

Figure 4. Gas chromatography derived cinnamaldehyde retention of encapsulated cinnamaldehyde versus unencapsulated cinnamaldehyde and physical mixture

After confirmation of capsule formation, gas chromatography was utilized to determine cinnamaldehyde retention of the spray-dried encapsulated cinnamaldehyde before and after heating. Spray-drying was conducted in order to produce a dry powder suitable for baking applications. For comparison, pure cinnamaldehyde and a non-sonicated physical mixture containing all the same components as the encapsulated cinnamaldehyde, were analyzed.

Samples (spray-dried, pure cinnamaldehyde, physical mixture) were left at room temperature or heated at either 50, 100, 150, 200, or 250°C. Extraction was performed in ethyl acetate as cinnamaldehyde exhibited good solubility within the solvent. Vortexing and low-intensity sonication were necessary to rupture the capsules and release the flavor into the organic layer. This organic phase was injected into the GC and cinnamaldehyde retention was calculated based on peak areas collected in PeakSimple, by comparing to standards of cinnamaldehyde (**Figure**

4). The process of spray-drying induced unavoidable heating of the encapsulated cinnamaldehyde. Initial gas chromatography analysis yielded a retention of 48% of the original cinnamaldehyde, post-processing. This information was factored into the final analysis. Cinnamaldehyde retentions for the furnace heating process were accordingly adjusted to represent a 100% retention of the starting cinnamaldehyde amount after spray-drying. This allowed for a direct comparison to be made between all samples.

Cinnamaldehyde has a boiling point of close to 250°C, however as observed by GC analysis and TGA data, evaporation of the compound seems to occur between 100-150°C. Therefore, the retention differences between samples aren't particularly of any concern at the lower heated temperatures. In addition, for baking applications, the range of 150-250°C is of more interest. At 150°C and 200°C, the encapsulated cinnamaldehyde has significantly higher flavor retention than both the unencapsulated cinnamaldehyde and physical mixture. 200°C represents a typical baking temperature and the encapsulated formulation shows an improved 15% cinnamaldehyde retention versus the pure cinnamaldehyde and physical mixture controls, where all the flavor was lost (0% retention). Even at 250°C, the encapsulated formulation retained a small proportion of the cinnamaldehyde (1.5%) whereas the other samples did not. Application wise, one would be able to retain more flavor with the encapsulated cinnamon in baked rolls or bread. Less amounts of the flavor could even be utilized, which at an industrial level could save a lot of money.

3.3 Thermogravimetric Analysis

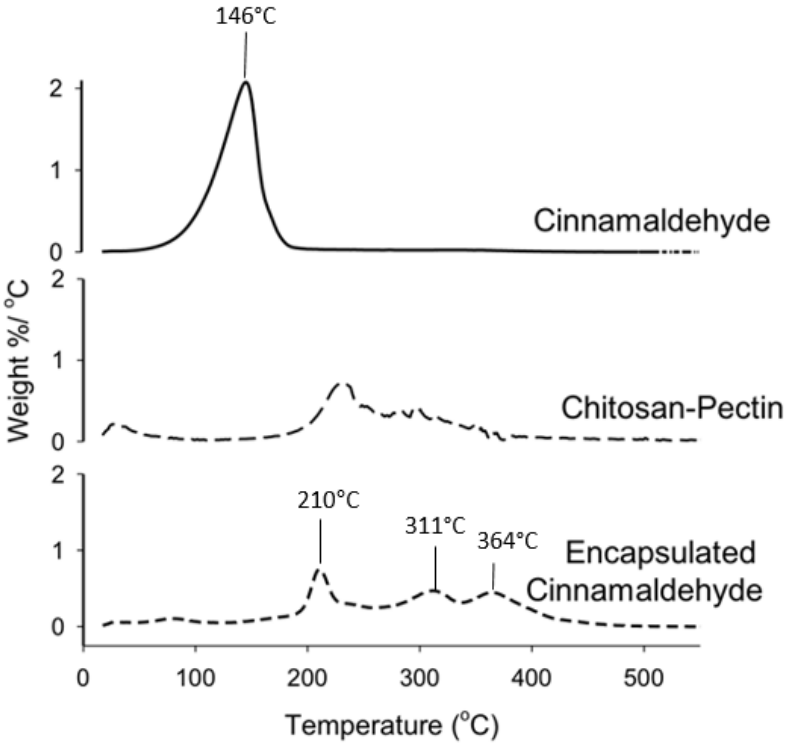

Figure 5. Thermogravimetric analysis of encapsulated cinnamaldehyde and capsule components versus pure unencapsulated cinnamaldehyde

Thermogravimetric analysis was conducted on the spray-dried powder and its constituents to further characterize the stability of the encapsulated cinnamaldehyde (**Figure 5**). The curve observed when heating pure cinnamaldehyde shows bulk degradation around 146°C. In comparing the encapsulated cinnamaldehyde curve, there is no such peak close to this temperature. As cinnamaldehyde comprises a large proportion of the spray-dried formulation (~48%), one would expect to see a large peak at that general temperature point if no heat stability difference existed between it and pure cinnamaldehyde. This is not the case, and a large peak corresponding to sample weight loss of the encapsulated cinnamaldehyde is not detected until 210°C. Additional peaks are seen in this spectrum at 311°C and 364°C. The cinnamaldehyde may be degrading at 210°C or even in conjunction with other capsule components at higher

temperatures. Regardless, this demonstrates that the encapsulation of cinnamaldehyde does indeed impart heat stability to the flavor. Heat must first penetrate or degrade the capsule before reaching the inside contents. This obviously requires higher temperatures or longer times to evaporate the cinnamaldehyde versus free, susceptible cinnamaldehyde.

3.4 Yeast Growth Studies

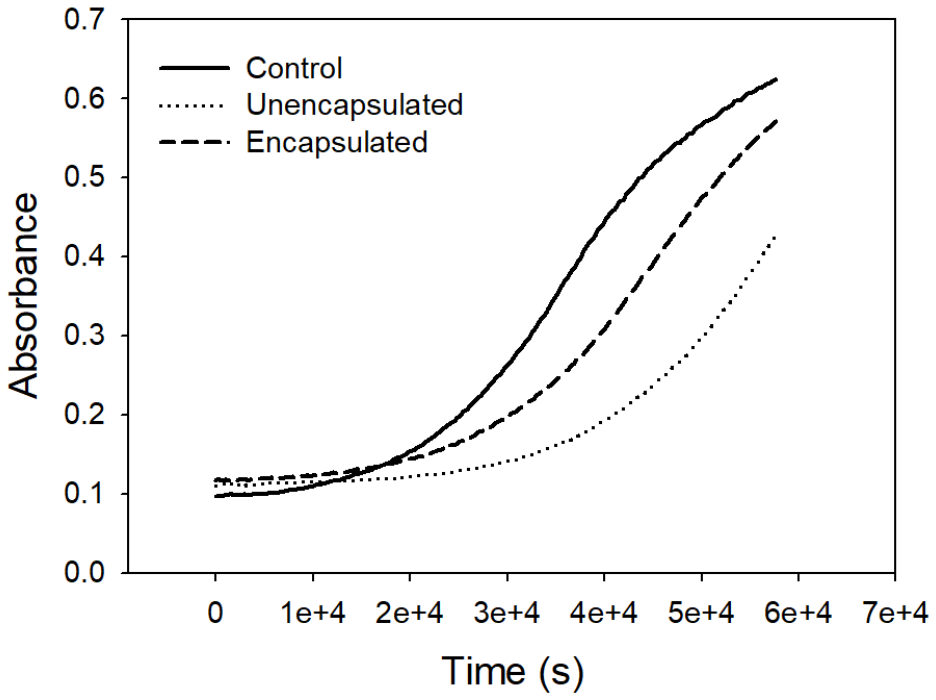

Figure 6. Yeast growth curves of untreated, encapsulated cinnamaldehyde treated, and unencapsulated cinnamaldehyde treated yeast

In addition to demonstrating increased cinnamaldehyde heat stability from encapsulation, yeast growth studies were conducted to examine the effect encapsulation had on cinnamaldehyde's antifungal properties. *Saccharomyces cerevisiae* was allowed to grow in wells after treatment with either pure cinnamaldehyde or encapsulated cinnamaldehyde (**Figure 6**). A control group with no treatment exhibited the greatest amount of growth. Between the

cinnamaldehyde treatments, the yeast growth was greater when subjected to encapsulated cinnamaldehyde than pure cinnamaldehyde. This indicates that encapsulation does in fact inhibit the antifungal properties of cinnamaldehyde. The polymer capsule provides a barrier that limits interactions between cinnamaldehyde and yeast and the yeast is allowed to grow properly. This has implications for baking as proper growth of yeast in dough allows for increased rates of fermentation and subsequent gas production, yielding better volume and texture.

CHAPTER 4

4. CONCLUSIONS

This study proposes a formulation for encapsulating cinnamaldehyde by utilizing ultrasonication. Chitosan and pectin are polymers that were found to be suitable for forming a capsule capable of incorporating the hydrophobic cinnamon flavor in an aqueous environment. This microcapsule formation was verified via FTIR analysis which indicated the construction of amide bonding. Capsule morphology was also observed under SEM, where spherical particles were indeed discerned. Gas chromatography was utilized to determine cinnamaldehyde retention in samples heated at baking temperatures. Compared to pure cinnamaldehyde and the physical mixture of the formulation, the encapsulated cinnamaldehyde displayed significantly higher flavor retention at the high temperatures. This indicated that the encapsulation imparted improved heat stability to the encapsulated flavor. Thermogravimetric analysis also corroborated this fact. When comparing the heating curves of pure and encapsulated cinnamaldehyde, the encapsulated formulation depicted cinnamaldehyde loss at a higher temperature than that of pure cinnamaldehyde, representative of improved heat stability. Additionally, yeast growth tests were conducted and *Saccharomyces cerevisiae* showed superior growth when subjected to encapsulated cinnamaldehyde versus subjection to pure cinnamaldehyde of the same concentration. This signifies the reduction of the antifungal properties of cinnamaldehyde due to encapsulation. In conjunction, the improved heat stability and reduced yeast interaction serve to demonstrate that this particular formulation and method for encapsulating cinnamon flavor can yield baked products of higher flavor and texture quality than unencapsulated cinnamon flavor. In addition, less amounts of encapsulated flavor would be required to achieve the same organoleptic qualities of normal cinnamon, potentially reducing costs for food manufacturers.

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