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Fuzzy logic application to model caffeine release from hydrogel colloidosomes

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ABSTRACT

The diffusion coefficients of mass transfer in colloidosomes are sensitive to environmental changes. Also, Fick's law of diffusion cannot be solved analytically in presence more complex geometries or non-constant active agent diffusivities. Because of the approximation property and uncertainty handing of fuzzy systems, here they are employed to model diffusion coefficients in the caffeine release from hydrogel colloidosomes. The approximated diffusion coefficients are then used in the Fick's law equations, to create the fuzzy-diffusional model. The identification and validation of the fuzzy-diffusion model is obtained from experimental caffeine release curves of colloidosome samples in different shear rates, and at two time ranges. The proposed method is then compared with three other methods and depicts lower error and variance both in identification and evaluation. The diffusional-fuzzy model maintains the interpretability of the Fick's model, improves the process simulation and eliminates phenomenon and property considerations.

1. Introduction

Microencapsulation is the most frequently used technique to accomplish controlled release in the food industry. Controlled release is a method for providing additional protection to a flavoring point. The delay in release can lessen volatilisation, oxidation or chemical reaction, as a result of which the performance of the flavoring considerably improves (Pothakamury and Barbosa-Canovas, 1995; Taylor and Hort, 2007).

While many flavor encapsulants are efficient, it is essential that the flavor molecules are released when the food is consumed; otherwise the flavor, despite being in suitable condition, will not be sensed (Roberts and Taylor, 2000).

Flavor release depends on the nature of the foodstuff itself and the way in which it is consumed. The state of the foodstuff can alter substantially as it is broken down by chewing, and mixed with saliva (Voilley and Etievant, 2006).

Flavor or any other active agents are released from the controlled release delivery systems by diffusion, degradation, swelling, osmotic pressure and melting. Diffusion is controlled by solubility and permeability of compounds in food matrices., In food systems it is the dominant mechanism in controlled release from encapsulation matrices. The principal steps in the release of a flavor compound from the matrix system include: diffusion to the surface of the matrix; partition between the matrix and the surrounding food and transportation of the flavor compound away from the matrix surface (Madene et al., 2006).

Colloidosomes as an innovative class of microcapsules are formed by the self–assembly of colloidal particles to the interface of emulsion droplet. By this means, a variety of release strategies may be attainable. Colloidosomes have a wide range of applications in different areas such as pharmaceuticals, food, flavors, fragrances, and cosmetics industries (Parthibarajan et al., 2011).

The most appealing characteristic of alginate is its versatility of gel formation simply induced by various different cations. Alginate beads have been used in microencapsulation as their preparation on a laboratory scale is undemanding because of their gentle hydrogelling process, relatively economical rate and safety for use in food formulations (Madziva et al., 2005).

Fuzzy systems are intelligent paradigms that have

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approximation property, uncertainty handling, and incorporating human knowledge. Modeling and control methods based on the fuzzy systems attempt to integrate numerical and symbolic processing into a single framework. Using linguistic knowledge, fuzzy systems integrate human knowledge into the control structure. In comparison with other nonlinear approximation techniques, fuzzy systems offer a more transparent representation of the nonlinear complexes that scientists handle in biological systems such as food related problems. In this manner, processed data can be translated in a model and analyzed in an approach analogous to what the public is cognizant of (Babuška and Verbruggen, 1996).

Recently, uncertainty of bioprocesses has persuaded scientists to search for new methods of solving fundamental phenomena including mass and heat transfer.

In order to design encapsulation systems especially with controlled release possibility, it is crucial to model the release data to determine the mass transfer properties of the capsules and reaching to desire release rates (Zuidam and Nedovic, 2010). In biological systems, alterations of production conditions are inevitable. In this case, the prepared spheres of hydrogel colloidosome are not uniform and equal (Amiryousefi et al., 2016). Therefore, the release characteristics of the prepared colloidosome samples do not follow a specific pattern. In fact the diffusion coefficients of caffeine release are not constant. First, this study aims to provide a simple, rapid and non-destructive method to encapsulate caffeine as a flavor model in colloidosome. Second, to handle the uncertainties in bioprocesses, here a Diffusional-fuzzy model is introduced that approximates diffusion coefficients in the caffeine release from hydrogel colloidosomes. The approximated coefficient is then used in the Fick's law equations in order to get into more realistic solutions with fewer assumptions.

The overall structure of the paper is as follows: in Section 2 the materials and the methods are described. Results and discussions are brought in Section 3, and finally, conclusions are drawn in Section 4.

2. Materials and methods

2.1. Materials

Caffeine ($C_8H_{10}N_4O_2$) and Sodium alginate were obtained from AppliChem; calcium chloride and D-(+)-Gluconic acid δ-lactone (GDL) from Sigma; sodium poly(styrenesulfonate) (PSS, MW 70000) and Na_2CO_3 from Aldrich; and sun flower oil from Oila company.

Highly purified water was obtained by deionization and filtration with a Millipore purification apparatus. Other chemicals were all analytical reagents and used as received.

2.2. Preparation of porous $CaCO_3$ microparticles

Porous $CaCO_3$ microspheres were prepared via the process reported by Wang et al. (2006). First, 0.2 M $CaCl_2$ was rapidly poured into an equal volume of 0.2 M Na_2CO_3 solution (containing 4 g/L PSS) at room temperature (23 °C). After vigorous agitation with a magnetic stirrer, water of the precipitate was removed. Then, the precipitate was carefully washed with deionized water, and dried in an oven at 50 °C for 48 h. This simple procedure results in a highly homogeneous spherical porous $CaCO_3$ microparticles. Before loading, $CaCO_3$ microparticles were washed with acetone and dried in a vacuum oven at 50 °C.

2.3. Fabrication of colloidosome hydrogel beads

The colloidosome hydrogel beads were prepared by an emulsification technique modified from Liu et al. (2008). The parameters of Table 1 was used to optimize fabrication of the microcapsules, find the best fractions of ingredients and also model the release kinetics.

At first, porous $CaCO_3$ microparticles were dispersed in 5 mL sunflower oil through stirring for 1 h. The defined amount of aqueous solution of 1 wt% sodium alginate was added into oil, and water-in-oil emulsion was formed by stirring. Then 150 μL of freshly prepared 0.2 g/mL GDL aqueous solution was added to gelate the alginate core. The emulsion was shaken for two hours, and then left for 48 h in the experimental condition. Due to gravity, the colloidosome hydrogel beads sank into the bottom of the container. Next, the colloidosome hydrogel beads were collected by centrifugation (Hettich Zentrifugen, 12D-78532 Tuttlingen, Germany) for 5 min in 4000 revolutions per minute (rpm), and then, they were washed three times with ethanol. Finally, the obtained colloidosome hydrogel beads were redispersed in water through shaking.

2.4. Flavor loading

To evaluate flavor release from the fabricated colloidosomes, caffeine, which is a widely used flavor in processed foods and beverages, was applied as a model system. In addition to availability and usefulness of caffeine in nutrition and food industry, its form is stable in experimental conditions. Furthermore, a simple and rapid spectrometric method exists for the determination of caffeine in aqueous solutions (Purcarea et al., 2008).

The caffeine loaded colloidosome hydrogel beads were fabricated in the same way as described above except that the sodium alginate aqueous solution contained 0.3 g/L caffeine equal to 1.54 mM.

By dividing the actual caffeine content on theoretical caffeine content, the amount of caffeine loading was measured. The colloidosome hydrogel beads was treated with ultra-sonication and centrifugation to obtain the actual caffeine content. Also, the amount of caffeine was measured with spectrometery method.

2.5. Release

To evaluate the caffeine release from colloidosome, the shear rate in the range of mouth for drinks is approximately considered as 55 s^{-1} (Karaman and Kayacier, 2010). Then, the amount of caffeine release was stimated at 37 °C and neutral pH.

The most common method is to measure the release of active ingredients from encapsulates, which are dispersed in a liquid medium. To assure good mixing and the increased concentration of solution in the liquid phase, gentle agitation is applied to determine the released kinetics and diffusion coefficient (Zuidam and Nedovic, 2010).

Concentrations of released caffeine molecules were calculated by using a linear calibration curve. 4 mg of caffeine was dissolved in 10 ml of deionized water. The solution was then shaken for 2 min and diluted by half. The dilution was repeated until 8 samples were made with pure water as the lowest concentration. The absorption spectra exhibited peaks at 271 nm. Concentration was in the linear regime to minimize error (Rosenberg, 2010).

Simulated mouth condition was considered as temperature of 37 °C, neutral pH and shear rates in range of mouth shear condition for drinks. The prepared colloidosome hydrogel beads were exposed in simulated mouth condition to determine the amount of released caffeine. To assess the modeling and evaluating of caffeine release in simulated mouth condition, released amount was measured at 3 shear rates of 0, 50 and 100s^{-1} during 300 min. A magnetic stirrer with hot plate was applied to create the mentioned