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A mathematical model of the effect of pH and food matrix composition on fluid transport into foods: An application in gastric digestion and cheese brining

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The question of bioaccessibility of nutrients within a food matrix has become of increasing interest in the fields of nutrition and food science as bioaccessibility is the precursor to bioavailability. By analyzing the propagation of the wetting front of acidic water in raw carrot core and Edam cheese as model systems, we show that the diffusion of the acidic water is dependent on the pH of the gastric fluid and the food matrix. In addition, we demonstrate that the diffusion of NaCl during cheese brining is also dependent upon the concentration of the NaCl. This demonstrates that Fickian diffusion, along with a concentration dependent diffusion coefficient, is a valid model for describing concentration profiles in multiple food systems.

Utilizing the diffusion rates found at various pH levels (1.50, 2.00, 3.50, 4.30, 5.25 and 7.00), we developed a model to describe the measured non-linear rate of soluble solid loss during digestion at various constant pH levels. Additionally, we have developed a model to predict the likely rate of soluble solid loss during digestion in the stomach where pH decreases with time. This model can be used to help understand and optimize the relationship between food structure/composition and food degradation in the human stomach, which may help in the development of novel foods with desired functionality.

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1. Introduction

1.1. Cheese brining

The brining process is critical in cheese making. The salt content affects the moisture content, texture, and flavor of the cheese. Absorption of salt facilitates the removal of whey and reduces the moisture content of the cheese, which improves the shelf life of the cheese. Salt also regulates the growth of bacteria within the cheese curd which affects the flavor and acidity of the cheese [\(Floury et al., 2009;](#page--1-0) [Wendorff, 2010](#page--1-0)). The acidity of the cheese has a marked effect on the texture of the cheese. For instance, a low curd pH (4.7) can cause soft cheeses to become gritty whereas cheese curd at an optimal pH (5.2) remains creamy [\(Hynes, Delacroix-Buchet, Meinardi, & Zalazar,](#page--1-0) [1999\)](#page--1-0).

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There have been a variety of models used to depict either the diffusion of solutes into foods or the leaching of solutes into a surrounding liquid ([Diaz, Wolf, Kostaropoulos, & Spiess, 1993; Schwartzberg &](#page--1-0) [Chao, 1982; Turhan & Kaletunç, 1992](#page--1-0)). Typically, these processes are described using Fickian diffusion; however in the case of multicomponent systems, such as moisture reduction and salt uptake in cheese during the brining process, there have been multiple approaches. The concentration of salt in cheese during the brining process has been studied extensively and has been modeled primarily by using either Fickian diffusion [\(Bona, Silva, Borsato, Silva, & Fidelis, 2010; Naesens,](#page--1-0) [Bresseleers, & Tobback, 1981; Pajonk, Saurel, & Andrieu, 2003; Turhan](#page--1-0) [& Kaletunç, 1992](#page--1-0)) or non-Fickian diffusion, in particular the Maxwell– Stefan approach ([Payne & Morison, 1999; Verschueren, Engles,](#page--1-0) [Straatsman, van den Berg, & de Jong, 2007\)](#page--1-0), with the assumption that the diffusion coefficient is constant, except in the study of [Pajonk et al.](#page--1-0) [\(2003\)](#page--1-0). [See [Floury, Jeanson, Aly, and Lortal \(2010\)](#page--1-0) for a comprehensive review on diffusion of solutes in cheese. For a detailed description of the Maxwell–Stefan approach, see [Krishna and Wesselingh \(1997\).](#page--1-0)] However, the unreliability of the Fickian diffusion model to describe the moisture content of cheese may be a result of the fact that moisture transport in porous media is both heterogeneous and dependent upon the moisture content, like that seen with NaCl. Thus, the effective

Abbreviations: AW, acidic water; EDC, effective diffusion coefficient; MCMC, Markov chain Monte Carlo; SGF, simulated gastric fluid.

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diffusion coefficient (EDC) is not constant as assumed in the aforementioned works.

In the instance of sorption or desorption, the dependence of the EDC on moisture content is well established [\(Crank, 1975\)](#page--1-0) and has been shown to be relevant to cheese in the modeling work of [Payne and](#page--1-0) [Morison \(1999\)](#page--1-0) and the experimental work of [Guinee and Fox \(1983\).](#page--1-0) In each instance, the EDC increased concomitantly with an increase in moisture content. This activity is not only restricted to cheese, but has also been demonstrated in the absorption of water in peas [\(Waggoner](#page--1-0) [& Parlange, 1976](#page--1-0)), soybean, corn, cotton seeds [\(Phillips, 1968](#page--1-0)), and rice [\(Gomi, Fukuoka, Takeuchi, Mihori, & Watanabe, 1996](#page--1-0)), in the dehydration of carbohydrates ([Aldous, Franks, & Greer, 1997\)](#page--1-0), and in the drying of gelatin and sugar solutions [\(Yamamoto, 1999\)](#page--1-0).

1.2. Bioaccessibility of nutrients as a result of gastric digestion

In food science and nutrition the question of the bioaccessibility of a food (the fraction of a nutrient released from a food matrix) is of increasing interest. In order for a nutrient to become bioavailable (absorbed by and used by the host), it must first become bioaccessible through the processes of mastication, stomach digestion, and enzymatic reactions within the small bowel. Both the degree of solid loss (or the extent of nutrient loss from the food matrix/particle) and the amount of remaining material are critical to human health; in that the leached soluble fraction is readily available for absorption in the small bowel and the remaining insoluble fraction may act as a substrate for bacterial growth within the small and large bowel.

In order to develop novel foods which either increase the rate of bioaccessibility (in the case of foods targeted for consumption by athletes, infants, and the elderly) or decrease the bioaccessibility (in the case of food matrices which encapsulate probiotics and thus may need to remain intact throughout digestion), a deeper understanding of human digestion is required as well as the ability to predict the behavior of food matrices within the human gastrointestinal tract.

Digestion begins with the mastication of food particles. While mastication is not essential to the chemical breakdown of food particles in the stomach ([Davenport, 1982](#page--1-0)), it is beneficial in that mastication mixes the food with the saliva which lubricates and softens the foods. Additionally, a key enzyme in saliva, α -amylase, aids in the digestion of starch during gastric digestion when the pH of the gastric contents is greater than three ([Bornhorst & Singh, 2012](#page--1-0)). Most importantly, the process of mastication reduces the particle size which increases the surface area of the particles allowing for more contact with and increased absorbance of gastric fluid upon entering the stomach.

The degradation of food particles within the stomach is complex and results from both mechanical forces and chemical reactions. Peristaltic movements within the stomach compress the food bolus, which can result in fracturing of food particulates, and may facilitate erosion or shearing due to the mixing of the stomach contents. Tenderization of food particles results from the absorption of gastric fluid; both acidic hydrolysis and enzymatic reactions facilitate the leaching of nutrients from the food particle making the nutrients bioaccessible. The extent of soluble solid loss, and hence nutrient bioaccessibility, as well as the erosion of the particle are also dependent upon the food structure, preparation, particle size, meal volume and composition, viscosity, pH, and temperature.

There have been few models proposed in the literature that describe food digestibility. Current mathematical models which describe food digestibility have primarily focused on the wet mass retention ratio of the studied food (wet sample weight after digestion time divided by the initial wet sample weight) [\(Kong & Singh, 2009b\)](#page--1-0); however, the wet mass retention ratio may be more suitable in describing the satiety properties of a food matrix rather than the bioaccessibility of nutrients within a food matrix. As observed in studies with raw and roasted almonds, the wet mass retention ratio increased with digestion time

as the food matrix absorbed gastric fluid, whereas the dry solid mass decreased over the digestion time [\(Kong & Singh, 2009a\)](#page--1-0). The absorption of gastric fluid in almonds also resulted in volume swelling. In raw almonds there was an increase in the initial volume by 10 to 30%, depending upon the initial particle size [\(Kong & Singh, 2009a](#page--1-0)). Thus the wet retention ratio does not necessarily provide information on soluble solid loss (or bioaccessibility), rather it may provide information on the change in volume of the food particle as a result of swelling from the absorption of gastric fluid. Increased particle size reduces the rate at which the food particle empties from the stomach and increases the volume of the bolus, both of which are directly related to the feeling of fullness or satiety [\(Marciani et al., 2001](#page--1-0)).

Recently, the dry solid loss of carrot was described using the Weibull function [\(Kong & Singh, 2011\)](#page--1-0); however, this description of the dry solid loss does not adequately describe the solid loss after an extended time (e.g. 36 hours). Such empirical models, although useful for inference purposes, do not incorporate any of the mechanisms that influence digestibility. For instance, the Weibull function does not describe the process of diffusion of the gastric fluid into the food particle, which may be pH dependent. Models of the mechanisms underlying the degradation of food particles will yield a better understanding of the interaction and role of different processes in the breakdown of food particles.

It is well known that the pH of the gastric contents after a meal is not static in time, rather the pH of the gastric contents can reach approximately 5 within minutes after the ingestion of a meal, and may take up to 2–3 h before the gastric contents reaches a pH below two [\(Barlow, Hinder, DeMeester, & Fuchs, 1994; Malagelada, Longstreth,](#page--1-0) [Summerskill, & Go, 1976; Mattioli et al., 1992](#page--1-0)). In previous in vitro studies of carrot digestion, the pH of the simulated gastric fluid (SGF) had a marked effect on the amount of soluble loss from the food matrix [\(Kong & Singh, 2011](#page--1-0)). The rate of soluble loss from a food matrix in the stomach is therefore not static in time and cannot be predicted based on a static pH level.

To understand the complexity of gastric digestion and to build models which accurately describe the underlying digestive mechanisms, it is essential to determine the effects of different factors (food structure, preparation, particle size, meal volume and composition, viscosity, pH, and temperature) on gastric digestion prior to investigating the interactions between those factors. Here, we consider only the effect of pH on digestion as the pH directly affects enzymatic reactions, varies over the digestive time, and has been shown to have a significant effect on the amount of soluble loss from the food matrix.

Given these results, it is our hypothesis that the rate of diffusion of fluid/solute into a food particle is not only dependent upon the concentration of the fluid/solute diffusing into the food particle, but is also dependent on the pH of the fluid and the structure of the food matrix. Furthermore, these dependencies contribute to the increased rate of soluble loss and protein degradation in particles submerged in gastric fluid with low pH. Note that our hypotheses are independent of pepsin-mediated pH and food effects on the rate of diffusion of the fluid into the food particle during gastric digestion (i.e., pepsin was not included in the SGF to ensure that any difference observed in the propagation of the acidic fluid between food matrices was a result of the food structure). Even though pepsin effects are excluded from this study, this does not preclude the important role of pepsin-dependent effects on the rate of diffusion of the gastric fluid into a food particle. Understanding the pepsin-independent effects is a prerequisite to understanding the pepsin-dependent effects. We aim to demonstrate this by developing a dynamic, mechanistic mathematical model that describes the soluble loss of a food particle due to degradation by acidic hydrolysis in the human stomach using raw carrot and Edam cheese as model food systems. As mentioned earlier, there has yet to be a mechanistic model proposed in the current literature. Specifically, we will describe the propagation of the wetting front into the food particle, which has a major influence on the rate of nutrient release during

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