



A mechanistic model for swelling kinetics of waxy maize starch suspension

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ABSTRACT

The evolution of 8% suspension of waxy maize starch granule size distribution when subjected to heating to 65, 70, 75, 80, 85 and 90 °C at a heating rate of 15 °C/min were measured. Granule swelling was more pronounced at higher temperatures with an increase in the average size from 13 μm to 25–28 μm, eventually approaching equilibrium. Cryo SEM images revealed porous structure of swollen granule. The proposed model for starch swelling accounts for entropy of mixing, enthalpy of water-starch interaction and elastic restoring force. The second virial coefficient of waxy maize in aqueous medium was characterized by static light scattering. Chemical potential profile as well as the temperature profile within the granule were predicted from the solution of unsteady state diffusion and heat conduction respectively. The granule size was then inferred from the chemical potential profile. The proposed model predicts the evolution of average granule size and granule size distribution accurately.

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1. Introduction

Starches are important ingredients used in various food and non-food products. They are very useful in a number of food applications, where they may act as a source of calories, as well as thickening, stabilizing and gelling agents. Starch is the predominant food reserve carbohydrate in plants, and provides 70–80% of the calories consumed by humans worldwide. Starches are obtained from seeds, particularly corn, wheat, rice, and from tubers or roots, particularly potato, sweet potato, and cassava (Whistler and BeMiller, 1997). Starch occurs naturally as discrete particles, called granules. Starch granules range in size from sub-micron elongated granules of chloroplasts to the relatively huge oval granules of potato, which could be over 100 μm. Granule shapes include nearly perfect spheres and discs, and polyhedral or irregular granules (Chen et al., 2006). Starch granules are composed of a mixture of two polymers: a linear polysaccharide, amylose with $\alpha(1-4)$ glucosidic bonds and a highly branched polysaccharide, amylopectin with 5% of $\alpha(1-6)$

bonds which lead to a unique branched structure capable of crystallizing. Starch granules are relatively dense, are insoluble in water, and hydrate only slightly at room temperature; the dispersions formed have a relatively low viscosity. Starch granules swell when heated in an aqueous medium because of uptake of water due to a chemical potential gradient. This swelling is resisted by the elasticity of the granule network which ruptures at some point leading to leaching of starch (predominantly amylose) into the aqueous medium, thus resulting in its increased viscosity. The combined effects of increased volume fraction of granules (due to swelling) and the increased aqueous phase viscosity results in thickening of the starch dispersion, a phenomenon known as starch pasting. Starch pasting behavior greatly influences the texture of a variety of food products such as canned soups, gravies, sauces, baby foods, fruit pie fillings, puddings, batter mixes for deep fried foods etc. In non-food products, starch dispersion rheology and its pasting behavior is important in operations that range from paper coating to the fabrication of paints. Thus, it is necessary to quantify the effect of starch structure and composition on its pasting behavior in order to develop rational guidelines for modification of starch through cross linking in order to obtain desirable texture and rheological properties. This would require understanding the swelling of

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starch granules, the conditions under which they will rupture, the extent of release of its contents to the aqueous medium upon rupture and the effect of these on the rheology of suspension.

Starches isolated from different sources are known to have different molecular structures resulting in a wide range of different functionalities. Differences in functionality can be attributed to the morphology and size of the starch granules but also to the assembly and structure of the starch molecules within the starch granules (Swinkels, 1985; Singh et al., 2003). The ratio between amylose and amylopectin within the starch granules is considered important since this variable has profound effect on starch paste rheology as shown for amylose-free potato starch (Visser et al., 1997; Hoover, 2001) and high-amylose starch (Banks et al., 1974; Srichuwong et al., 2005). Other molecular properties such as starch molecular weight distribution (Blennow et al., 2001), and the degree of amylopectin branching are also known to influence the functional properties of starches (Fredriksson et al., 1998; Singh et al., 2007). The end uses of different types of starches are related to the ability of the granules to undergo swelling (Okechukwu and Rao, 1995; Vroman and Tighzert, 2009). In high amylopectin cereal starches like waxy maize starch, the granules tend to hydrate with ease, swell rapidly, and rupture to a great extent, resulting in the loss of paste viscosity, with weak bodied, stringy and cohesive pastes (Tattiyakul and Rao, 2000). Waxy maize starches, composed of nearly 100% amylopectin, have built-in viscosity stability due to the branched nature of the polymer molecule. Waxy maize is useful in a wide array of applications in the food industry because of its stability.

Two main types of crystalline starch structures have been detected by wide-angle X-ray scattering (Nara and Tsu, 1983; Galliard and Bowler, 1987; Pérez and Bertoft, 2010): the A-type structure of cereal grain starches such as maize, wheat, and rice; and the B-type structure of tuber, fruit and stem starches such as potato, sago and banana starches. An additional C-type structure composed of both A- and B-type polymorphs, has been detected in bean seed starches (Sarko and Wu, 1978; French, 1984; Jacobs et al., 1998; Lopez-Rubio et al., 2008). The type A X-ray pattern of cereal starches is indicative of parallel, double helices separated by interstitial water (Whistler and BeMiller, 1997). It has been shown (Stevenson et al., 2006; Cai et al., 2014) that amylopectin, molar mass, radius of gyration and density of pin oak acorn starch were comparable to other A-type starches. The molecular weight, radius of gyration and hydrodynamic radius of amylopectin have been investigated by laser light scattering and size exclusion chromatography (Galinsky and Burchard, 1995; Hanselmann et al., 1996; Bello-Perez et al., 1998; Millard et al., 1999; Durrani and Donald, 2000; Yoo and Jane, 2002; Fuentes-Zaragoza et al., 2010). The molecular weight of non-degraded amylopectin was reported to range from 1.70×10^8 g/mol to 5.60×10^8 g/mol, and the gyration radius of non-degraded amylopectin was reported to vary from 170 to 342 nm. The differences in the results obtained by different authors could be attributed to the different sources of samples and partly due to the sample preparation method (Cheng et al., 2006). In spite of extensive experimental investigations on starch pasting profile, quantitative prediction remains unsatisfactory. Studies have proved that granule size and its ability to incorporate water and swell can affect the function and physical properties of starch population (Crosbie, 1991; Pal, 1996). The distribution is normal for maize starch, bimodal for wheat starch, or trimodal for barley starch (Stapley and BeMiller, 2003). The initial average diameter or the length of major axis can be different among sources of starch and even within the same species or different part of the same plant. For example, the large A-type (disc-like) and small B-

type granules (spherical or polygonal) in wheat endosperm started with average diameter of 10–35 μm and 1–10 μm , respectively (Choi and Kerr, 2004). At 80 °C, the average diameter of native wheat starch granule increased from 20.42 to 44.64 μm in 1 min and further increased to 54.27 in 30 min. Rice starch granules, on the other hand, vary from 2 to 7 μm in their size (Wani et al., 2012).

Swelling of starch is characterized by an initial phase of slight swelling followed by rapid swelling and a final stage of maximum swelling (Tester and Morrison, 1990; Lagarrigue et al., 2008). Comparison of waxy and normal starches indicate that swelling power is a property of amylopectin and that amylose, lipids (Tester and Morrison, 1990) and gums (Kruger et al., 2003) inhibit swelling. Swelling power and water solubility gradually increased with increasing temperature after 60 °C for *S. epigaea* and potato starches and after 65 °C for maize starch (Cai et al., 2016). Swelling power was found to be in the order of potato starch > maize starch > barley starch > wheat starch with amylose leaching being highest for potato starch followed by wheat starch and least for maize starch (Eliasson, 1986; Tester and Morrison, 1990) and correlated well with gelatinization temperature and enthalpy of gelatinization for wheat starch (Sasaki and Matsuki, 1998). Experimental measurements of evolution of granule size distribution of corn and cowpea starch due to swelling when heated to different temperatures in the range of 67–90 °C (Okechukwu and Rao, 1995; 1996; Lagarrigue et al., 2008) indicated broadening of the distribution with an increase in mean particle size. Granule rupture was found to occur at longer times resulting in a decrease in mean diameter above 80 °C and 90 °C for cowpea and corn starch respectively.

Extensive investigations have been carried out on swelling of polymer gels (Katchalsky et al., 1951; Flory, 1953; Katchalsky et al., 1953; Katchalsky and Michaeli, 1955; Tanaka, 1978; Tanaka et al., 1980; Ricka and Tanaka, 1984; 1985; Hirotsu et al., 1987; Hooper et al., 1990; Joanny and Leibler, 1990; Achilleos et al., 2001; Kozlovskaya et al., 2006; Kopeček, 2007; Hong et al., 2008; Kabanov and Vinogradov, 2009; Gidley et al., 2010; Buenger et al., 2012). It has been demonstrated that changes in temperature (Tanaka, 1978), solvent composition (Hirotsu et al., 1987), pH (Zhang and Peppas, 2000; Kim et al., 2003) and ionic strength (Ricka and Tanaka, 1985; Hooper et al., 1990) can induce changes in the state of the swollen network. These induced changes have been exploited for drug delivery using polymer gels (Liechty et al., 2010). The validity of Flory-Rehner theory (Flory, 1953) has been demonstrated for a wide variety of systems (Prange et al., 1989; Baker et al., 1994; Urayama and Kohjiya, 1996). In some cases (Brotzman and Eichinger, 1982; Zhao and Eichinger, 1992), the additivity assumption of free energies of mixing and deformation has been shown to be unsatisfactory. Flory's theory has been extended by adding empty sites to the lattice (Lu and Hentschke, 2002). Monte Carlo simulations were presented for the equilibrium swelling of polymeric gels (Escobedo, 1996; Escobedo and Pablo, 1997).

In this manuscript, we propose a model for swelling of starch granule suspension when heated to a constant temperature. The proposed model accounts for diffusion of water into the granule as a result of chemical potential gradient. Polymer solution theory is adopted to express the chemical potential in terms of free energy of mixing as well as elastic free energy of the starch network. Experimental measurements of evolution of starch granule size distribution when heated at different temperatures are compared with model predictions. To the best of our knowledge, this is the first such model to explain swelling of starch granules in terms of their structure.

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