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Ultrasonic Characterization of Unyeasted Bread Dough of Different Sodium Chloride Concentrations

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ABSTRACT

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Altering the properties of dough by reducing sodium chloride (NaCl) content affects aeration processes during mixing. The effect of NaCl concentration on the bubble size distribution (BSD) in unyeasted doughs was investigated by an ultrasonic transmission technique through analysis of frequency-dependent ultrasonic phase velocity and attenuation coefficient. As NaCl concentration was decreased, the volume fraction of gas in the dough increased, resulting in a larger attenuation coefficient for the dough. From the peak in attenuation coefficient, estimates of the median radius and the width of the log-normal BSD in the dough were extracted, both of which were sensitive to the dough's NaCl concentration. As NaCl concentration was reduced, the bubble radius decreased and the width of the distribution increased, in accordance with expectations arising from changes in the dough's consistency. Over the course of 150 min, the radius increased (40–50%) and the width decreased (4–8%) for all dough formulations, consistent with changes in the BSD arising from disproportionation. These dynamic changes demonstrate that dough is an interesting soft material whose formulation can be manipulated to enable it to possess different BSDs; the diffusively driven evolution in these bubble sizes can be investigated noninvasively with ultrasound.

There is a current awareness of the nutritional advantages of low-sodium diets because high levels of sodium have been linked to high blood pressure, a major factor in cardiovascular disease (He et al 2000). Cereal products are reported to contribute about 30% of overall daily sodium intake (EFSA 2005). Reducing the sodium chloride level in baked product formulations is therefore a worthwhile objective. However, simply reducing the sodium chloride content in bread dough is not easy because salt has a number of important functions in bread baking and bread quality, such as strengthening the gluten, enhancing the handling and machinability of doughs, and improving the flavor of bread (Salovaara 1982; Linko et al 1984). Furthermore, altering the ingredient concentrations of a dough alters its mechanical properties, which affects the dough's air-entrainment capacity (Campbell et al 2001; Chin et al 2005; Bellido et al 2006; Mehta et al 2009; Koksel and Scanlon 2012).

During breadmaking, dough is subjected to a set of process operations in which the number and size of bubbles are manipulated (Campbell et al 1998). Determining the bubble size distribution (BSD) at the end of the mixing process is of crucial importance, because the aerated structure of bread depends on the BSD within the dough at this point of the process (Chin and Campbell 2005). Therefore, noninvasive monitoring of changes in the bubble population within the dough can be the basis for predicting final product quality before bread is manufactured. From a practical perspective, investigation of the BSD at the end of mixing can be most conveniently accomplished by using doughs prepared without yeast. Such unyeasted doughs are also interesting candidates for fundamental studies of time-dependent changes in bubbles, because unyeasted dough is a relatively stable, viscoelastic system that does not allow bubbles to cream out, making bubble disproportionation, an interesting physical process in itself, easier to monitor. However, it is extremely challenging experimentally to monitor bubbles in dough and to study how their size distribution and its evolution are influenced by changes in ingredient concentrations, because dough is optically opaque, bubbles are fragile, and they have rapid dynamics (Shimiya and Nakamura 1997; Bellido et al 2006; Scanlon et al 2008, 2011; Strybulevych et al 2012).

The use of low-intensity ultrasound is a promising approach for analyzing BSDs in viscoelastic media such as bread doughs because ultrasound is rapid, can be used in optically opaque systems, and is sensitive to the large density and compressibility differences between the dough matrix and the gas (Létang et al 2001; Ross et al 2004; Scanlon et al 2008). Ultrasonic pulses are launched into a material, and their propagation characteristics are used to interrogate the material's properties by measuring the phase velocity and attenuation coefficient as a function of frequency (Strybulevych et al 2007). It has been shown that these ultrasonic parameters reflect the effect of changes in matrix properties as well as changes in the BSD (Létang et al 2001; Leroy et al 2008a), and so the effects of manipulation of ingredients such as sodium chloride can be potentially evaluated.

Because the BSD in the dough, which is affected by the changes in ingredient concentrations, is central to the aerated structure of bread, the first objective of this study was to investigate the effect of reducing sodium chloride content on the BSD in dough via analysis of the frequency-dependent response of the phase velocity and attenuation coefficient of unyeasted dough by an ultrasonic transmission technique. The second objective of this study was to investigate the evolution over a long time of the BSD in doughs made with different sodium chloride concentrations to determine how disproportionation affects the BSD in unyeasted doughs.

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MATERIALS AND METHODS

Sample Preparation. Dough samples were prepared from 100 g of a strong breadmaking flour milled at the Canadian International Grains Institute pilot mill (Winnipeg, MB, Canada), 57% distilled water on a flour weight basis, and three different salt concentrations (0.8, 1.6, or 2.4% sodium chloride on a flour weight basis). A GRL-200 mixer, operating at 225 rpm, was used to blend ingredients and develop the dough. Doughs were mixed at ambient pressure for 4 min, which was shown previously to be optimal for doughs made with 2.4% salt. To reduce the number of bubbles entrained during mixing, in addition to the mixing at ambient (atmospheric) pressure, doughs with the same formulations were also mixed under reduced pressure by drawing a vacuum on the outlet of the mixing bowl (Fan et al 2013). These doughs were mixed for 2 min at ambient pressure before drawing a vacuum (pressure ≈0.04 atm), allowing the flour, water, and salt to be mixed first so that the flour particles would not be sucked out of the mixing bowl.

To prepare dough subsamples for the ultrasonic tests, a pathology blade lightly greased with mineral oil was used to excise a dough subsample disc of approximately 1–2 mm thickness from the center of the dough immediately after mixing. The remaining dough was kept in a sealed plastic container to minimize dehydration of the dough. The surfaces of the ultrasonic setup contacting the dough subsample were also lightly greased with mineral oil. Before being monitored ultrasonically for 3 h, doughs were rested for 3 h after mixing, allowing the rapid bubble dynamics that occur right after mixing to slow down during the ultrasonic tests. This long resting time was selected to enable complementary experiments, not reported here, to be performed; these experiments used a reflection ultrasonic technique (which initially required a long thermal equilibration time for reliable data acquisition) and a desktop X-ray microtomography setup (which was incapable of producing adequate quality images during the rapid bubble evolution right after mixing).

Experimental Methods. The experimental setup for testing dough subsamples was composed of an arbitrary waveform generator (AWG 33220A, Agilent Technologies, Mississauga, ON, Canada) that produced Gaussian pulses with central frequencies ranging from 1.8 to 4 MHz, a pair of Panametrics transducers with a central frequency of 2.25 MHz (Olympus NDT Canada, Alberta, Canada), an ultrasonic pulse receiver (Panametrics, Olympus NDT Canada), and a digital oscilloscope (TDS5032B digital phosphor oscilloscope, Tektronix, Beaverton, OR, U.S.A.) (Fig. 1). The ultrasonic experiments were carried out in transmission mode by placing the dough subsample in the path between the generating and detecting transducers.

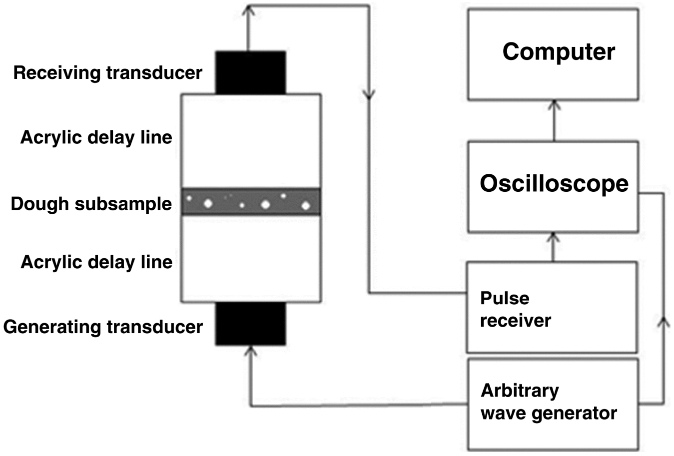

Fig. 1. Experimental setup for testing dough subsamples.

The voltage pulse emitted from the arbitrary waveform generator was sent to the generating transducer to create an ultrasonic pulse that traveled through the acrylic delay line, was partly transmitted into and through the dough subsample, and then traveled through the second acrylic delay line. The transmitted signal was detected by the second (receiving) transducer and amplified by the pulse receiver. The transmitted signal was averaged 500 times to improve the signal-to-noise ratio, and the averaged signal was sent to a computer for analysis of longitudinal phase velocity and attenuation coefficient. For measurements of the time evolution of the ultrasonic signals, data were recorded every 15 min, with the total time for each measurement (signal acquisition at five different central frequencies, plus data storage on the computer) being approximately 2 min. To acquire the reference signal, the two acrylic delay lines were placed in direct contact, and the pulse transmitted through the two delay lines was used as a reference.

The phase velocity (v) and the attenuation coefficient (α) were determined by comparing the phases and magnitudes of the Fourier transforms of the signal transmitted through the sample relative to the reference signal as follows:

$$v = \frac{\omega L}{\varphi_{\text{sample}} - \varphi_{\text{reference}}} \tag{1}$$

$$\alpha = -\frac{2\ \ln\left(A_{\text{sample}} / A_{\text{reference}}\right)}{L} \tag{2}$$

where φ, A, ω, and L are the phase and magnitude of the Fourier transform, angular frequency, and sample thickness, respectively (Strybulevych et al 2007). To determine v and α accurately, the phase and amplitude of the waves transmitted through the sample were corrected for the effects of reflections, resulting from acoustic impedance mismatch, at the interfaces between the dough sample and delay plates (Leroy et al 2011). Fluctuations in the frequency dependence of the impedance-mismatch-corrected v and α, because of signal-to-noise limitations, were smoothed out with a first-order polynomial Savitzky–Golay signal processing filter (Savitzky and Golay 1964).

For density measurements, a subsample of about 5 g was excised from the dough with a pathology blade and weighed with an analytical balance (±0.0001 g). The subsample was placed in a 25 mL specific gravity bottle (Kimble Glass, Vineland, NJ, U.S.A.) previously filled with distilled water equilibrated to room temperature. The density of the dough subsample was calculated from the weight of water displaced (Mehta et al 2009). Density measurements were performed at room temperature (23 ± 0.5°C), but with the temperature measured precisely so that dough density calculations took account of water density variation with temperature.

To determine the gas volume fraction, a two-point calibration was used, because there is a linear relationship between dough density and headspace pressure in the mixing bowl (Campbell et al 1998; Elmehdi et al 2004; Mehta et al 2009). The gas-free densities of the doughs (ρ_{gf}) were calculated from a linear extrapolation of dough density (ρ) to zero-pressure (0 atm) intercept for doughs mixed under atmospheric and reduced pressure (≈0.04 atm). This determination was performed for each dough formulation with different salt concentrations. The gas volume fraction in the dough (ϕ) was determined as follows:

$$\phi = 1 - \left(\rho / \rho_{gf}\right) \tag{3}$$

RESULTS AND DISCUSSION

Effect of Salt Concentration on Dough Density and Gas Volume Fraction in the Dough. By manipulating the salt concentration, doughs with different gas volume fractions were prepared. As salt concentration was decreased, dough density decreased and gas volume fraction in the dough increased, indicating that more air was occluded into the dough as the salt concentration was lowered (Table I). This trend can be explained by the salt's charge-shielding effect on proteins during the mixing process and the resulting improvement in protein cross-linking (Beck et al 2012). In a flour–water system, the gluten proteins have a net positive charge at normal pH (pH ≈ 6), and therefore they repulse each other, allowing faster hydration. When a small amount of salt is added to the system, it shields the charges on the protein chains, allowing the protein chains to approach each other (Miller and Hoseney 2008). This effect causes the flour to hydrate more slowly, increasing the hydration time and thus the dough development time (Hlynka 1962; Farahnaky and Hill 2007). Consequently, when doughs with different salt concentrations are mixed for a fixed period of time, the formulations with longer dough development times, that is, the formulations with higher salt concentrations, will occlude less air during that fixed mixing time (Koksel and Scanlon 2012). A decrease in dough density with a decrease in salt concentration was previously reported for doughs prepared with a fixed mixing time (Chin et al 2005).

Effect of Salt Concentration on Ultrasonic Phase Velocity and Attenuation Coefficient. The ultrasonic phase velocity (v) and attenuation coefficient (α) as a function of frequency are displayed for the different salt concentrations in Figure 2A and B, respectively. For all dough formulations tested, the frequency-dependent behavior of v and α was similar to what one would expect for a bubbly medium, regardless of salt concentration (Leroy et al 2008a, 2008b). The frequency-dependent peaks in both v and α are indicative of a low-frequency resonance arising from the bubbles entrained into the dough during mixing (Leroy et al 2008a; Scanlon et al 2008), because of the strong pulsations (periodic expansion and contraction) of the bubbles. These pulsations arise because of the large differences in density and compressibility between the gas in the bubbles and the surrounding material (dough matrix in our case) (Vagle and Farmer 1992; McClements 2009).

At frequencies well above the resonant frequencies, the phase velocities of all dough formulations approached each other and attained smaller values compared with the peak values reached near resonance. Our phase velocity values continued to decrease at the high-frequency end of our transducer bandwidth. We expect them to continue falling with a rise in frequency to attain phase velocity values of the same order of magnitude (≈1.7 km/s) to those reported for wheat flour–water doughs studied in other ultrasonic experiments (Létang et al 2001).

When the effect of salt on the ultrasonic properties was investigated, both v and α were found to be sensitive to the salt concentration. Reducing the salt concentration from 2.4 to 1.6% had a tremendous effect on both of the ultrasonic parameters investigated, although the effect of further salt reduction was not as pronounced (Fig. 2). For v, the magnitude of the peak increased and the frequency at which this peak occurred shifted toward lower values as the salt concentration was increased. The magnitude and position of the peak in α were also dependent on the salt concentration: as salt concentration was increased, both the magnitude of the peak (α_{max}) and the frequency at which this peak occurred (f_{max}) decreased. To ascertain how peaks in v and α were affected by salt concentration and thus the gas volume fraction in the doughs, α_{max} and f_{max} were plotted as a function of salt concentration (Fig. 3).

TABLE I
Effect of Sodium Chloride Concentration on Dough Density and Gas Volume Fraction in the Dough

NaCl Concentration (% fwb)[a]	Dough Density (kg/m^3)[b]	Gas-Free Dough Density (kg/m^3)	Gas Volume Fraction (%)
2.4	1,172.6 ± 4.6	1,273.8 ± 2.5	7.9 ± 0.4
1.6	1,150.9 ± 2.7	1,270.5 ± 4.6	9.4 ± 0.4
0.8	1,112.5 ± 3.0	1,268.6 ± 3.1	12.3 ± 0.4

[a] fwb = flour weight basis.
[b] Dough density values are the mean ± SD, n = 6.

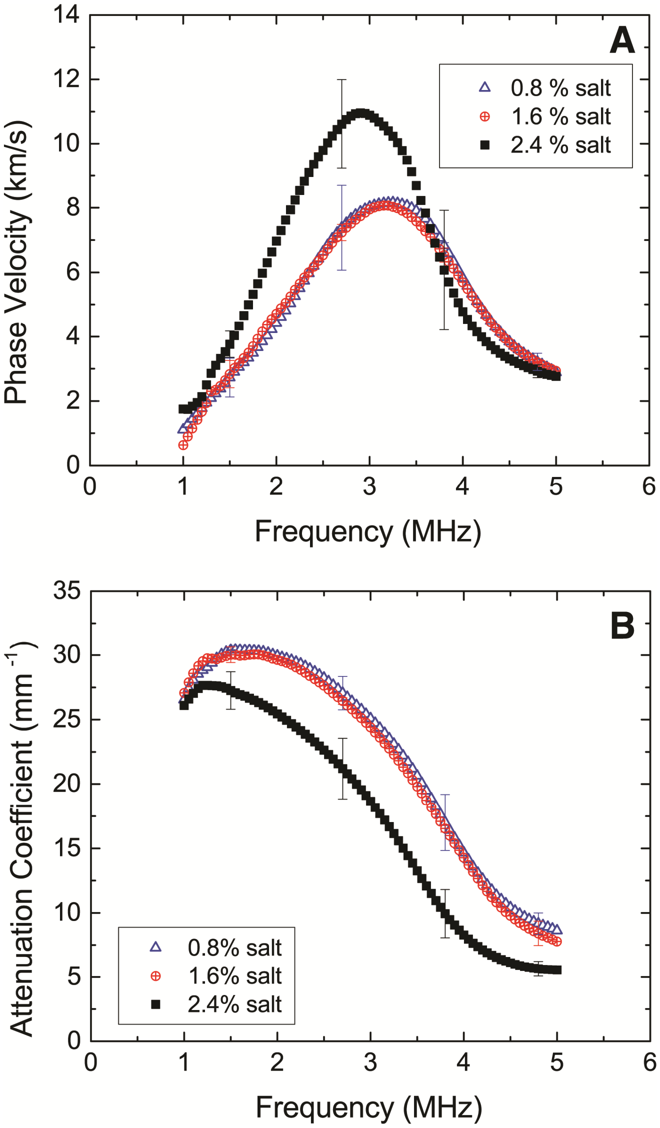

Fig. 2. Frequency dependence of phase velocity (**A**) and attenuation coefficient (**B**) as a function of salt concentration for doughs tested 5 h after mixing by the ultrasonic transmission technique. Error bars show ±1 SD.

In heterogeneous media such as dough, the total attenuation coefficient is due to the sum of scattering of the sound and dissipative processes occurring at the interface of bubbles and the dough matrix (Scanlon et al 2008). Therefore, α is expected to increase as the salt concentration decreases because of higher gas volume fraction in these doughs. It has been shown by ultrasonic transmission techniques at low frequencies (in which dough behaves as a composite of bubbles and dough matrix) that the attenuation coefficient was significantly higher for doughs with a higher concentration of bubbles (Mehta et al 2009). The attenuation coefficient in doughs is also dependent on the properties of the dough matrix. Water content and salt concentration are closely related in their effect on dough properties such as consistency, because salt addition tends to lower the flour's water absorption (Hlynka 1962). Using an ultrasonic reflection technique, Létang et al (2001) found that the attenuation coefficient decreased over the whole frequency range studied (1–11 MHz) with increasing water content. They concluded that water was affecting the ultrasonic properties of the dough matrix. It is possible that salt also affects dough matrix properties, but over this frequency range we cannot delineate a matrix effect on the attenuation coefficient from bubble effects.

Effect of Salt Concentration on Dough Bubble Sizes. Identifying the magnitude of the peak ($\alpha_{\max}$) and the frequency at which this peak occurred ($f_{\max}$) has the advantage that these parameters can be linked to the dough's BSD by the following equations (Leroy et al 2011; Strybulevych et al 2012):

$$\left(2\pi f_{\max}\right)^2 = \frac{3\kappa P_0 + 4\mu'}{\rho_M R_0^2} \qquad (4)$$

$$\alpha_{\max} = \frac{2\sqrt{3\phi}}{R_0 \exp\left(2\varepsilon^2\right)} \qquad (5)$$

where κ is the ratio of specific heat capacities of the gas in the bubble ($\kappa = 1.4$ for air), P_0 is the pressure of the gas in the bubbles, ρ_M is the density of the dough matrix, and ϕ is the volume fraction of bubbles in the dough (Leroy et al 2011; Strybulevych et al 2012). The real part of the complex shear modulus of the dough matrix (μ') was determined from the work of Leroy et al (2010), in which it was seen to follow the relation $\mu' = 10{,}900\omega^{0.234\pm0.004}$ over a wide frequency range. Because the distribution of bubbles that arises from the repetitive action of bubble subdivision is lognormal (Shimiya and Nakamura 1997; Bellido et al 2006), R_0 and ε are the median radius and the width of the lognormal BSD in the dough, respectively.

When the attenuation peak is broadened by high viscosity or a wide bubble distribution, the sharp resonant feature in the attenuation becomes washed out, and the parameter R_0 obtained from $f_{\max}$ in equation 4 does not correspond to the median radius of the bubbles but is closer to the radius of the smallest bubbles in the distribution (Leroy et al 2011). The shapes of the attenuation peaks in Figure 2B indicate that this situation applies to the dough samples investigated in this study. Thus, the values of R_0 extracted in our calculations for all salt concentrations by using equation 4, with the appropriate ρ_M (Table I), are in fact underestimates of the median radius. Furthermore, although the use of equation 5 to characterize the peak attenuation is a good approximation in this regime for which the peak is broad, the next step in our analysis—inserting ϕ (Table I) and these underestimated values of R_0

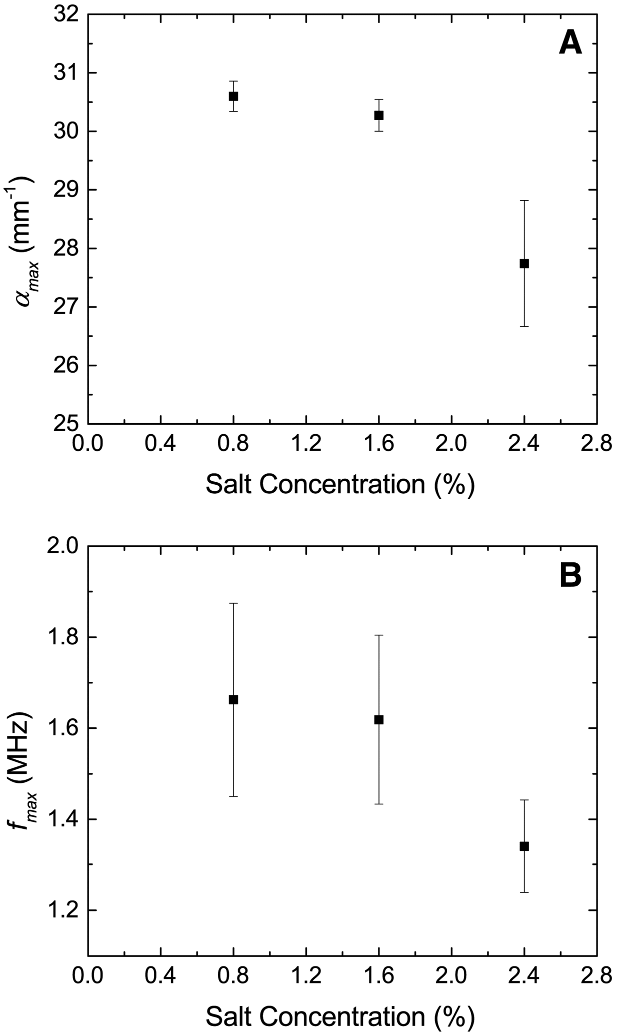

Fig. 3. The effect of salt concentration on the magnitude of the peak in attenuation coefficient (**A**) and the frequency where the peak in attenuation coefficient occurs (**B**) for doughs tested 5 h after mixing by the ultrasonic transmission technique. Error bars show ±1 SD.

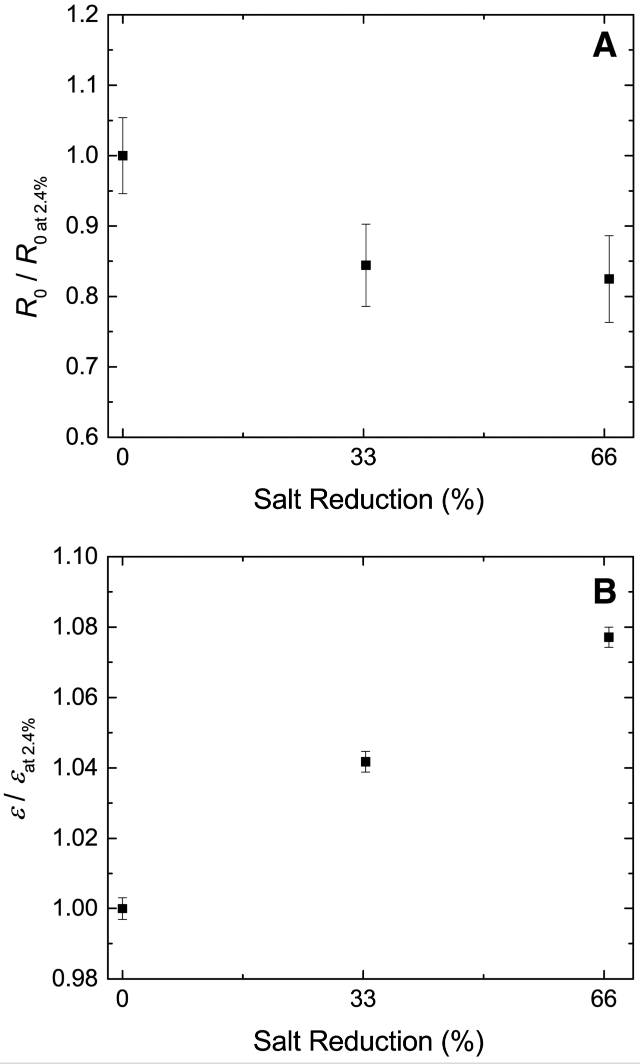

Fig. 4. The effect of salt concentration on the changes in the normalized bubble radius (**A**) and the normalized distribution width (**B**) for doughs tested 5 h after mixing. Error bars show ±1 SD.

into equation 5 to obtain ε—results in overestimates of the width of the BSD. This limitation, however, does not preclude achieving our main goal of using this method to track the relative changes in bubble radius and distribution width with salt concentration and evolution time. Therefore, to display these changes, our calculated values of R_0 and ε for each salt concentration were normalized with respect to the R_0 and ε of the dough with 2.4% salt concentration, enabling the BSD comparison between doughs with different salt concentrations. The effects of salt reduction on normalized radius ($R_0/R_{0\,\mathrm{at}\,2.4\%}$) and normalized distribution width ($\varepsilon/\varepsilon_{\mathrm{at}\,2.4\%}$) are presented in Figures 4A and B, respectively. It was found that $R_0/R_{0\,\mathrm{at}\,2.4\%}$ decreased but $\varepsilon/\varepsilon_{\mathrm{at}\,2.4\%}$ increased as salt concentration was reduced. Similar to our results, X-ray microtomography experiments showed that a higher median bubble size and a lower standard deviation were seen in a slack dough formulation compared with its stiff counterpart (Bellido et al 2006). Nevertheless, no direct comparison can be made considering that Bellido et al (2006) manipulated both the water and salt concentration, whereas the water level was constant for our experiments. In a study in which confocal laser scanning microscopy was used to investigate the microstructure of dough by measuring two-dimensional hole sizes on thin slices of dough, an increase in the bubble cross-section size was reported with increasing water content (Upadhyay et al 2012). Because our doughs with low salt concentration will be insufficiently hydrated (Farahnaky and Hill 2007), the bubble size results of Upadhyay et al (2012) were consistent with these findings that low-salt doughs yield smaller bubble sizes. In another study in which dough microstructure after 105 min of leavening was investigated by X-ray microtomography, the highest bubble surface-to-volume ratio was obtained for the lowest water content (Mastromatteo et al 2013). Although it is difficult to make a direct comparison, both results were consistent in the sense that the average bubble size decreased (i.e., surface area per unit volume increased) as the salt concentration was reduced or water content increased.

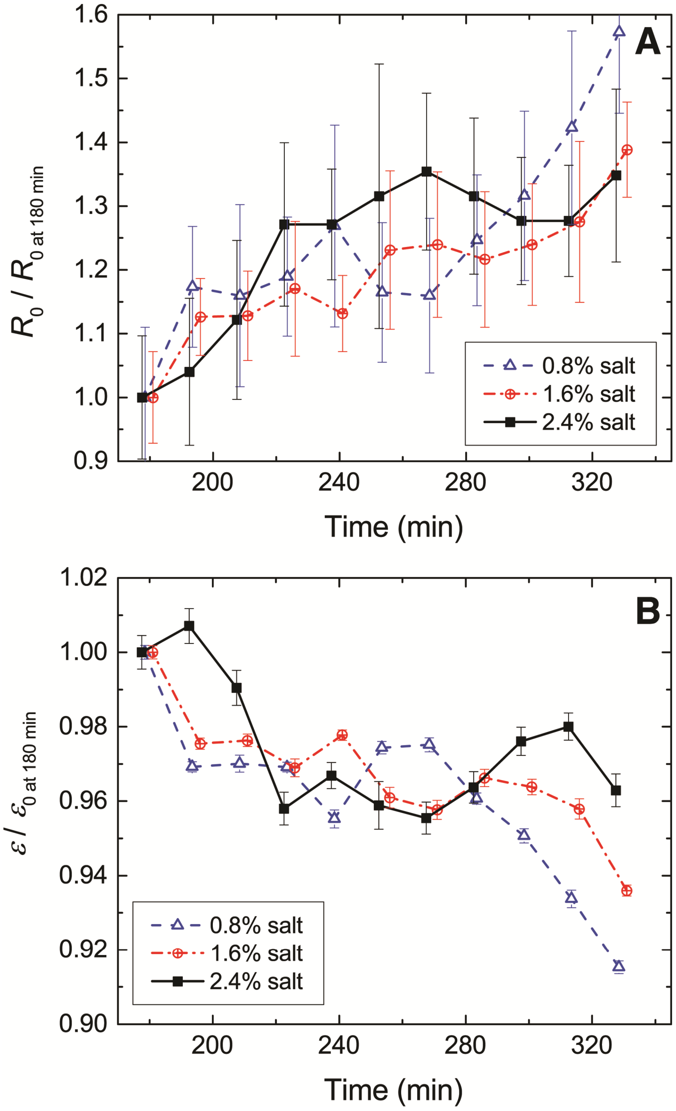

Fig. 5. The effect of salt concentration on the time-dependent changes of the normalized bubble radius (**A**) and the normalized distribution width (**B**) in the dough. Error bars show ±1 SD.

Effect of Salt Concentration on Time Evolution of Dough Bubble Sizes. In Figure 5A, the time evolution of bubble sizes, as determined by the ultrasonic transmission technique, is presented for the different salt concentrations. For each concentration, the values of R_0 determined from equation 4 were normalized with respect to the radius at 180 min, which was the start of the ultrasound experiments. This normalization allowed a meaningful comparison of the time evolution of bubble sizes in doughs with different salt concentrations. The corresponding time-dependent changes in the widths of the BSD are shown for each salt concentration and time in Figure 5B. As with Figure 4B, ε was determined by inserting the values of R_0 from equation 4 and ϕ from Table I into equation 5. Figure 5B shows the relative changes in ε as a function of time for each salt concentration by plotting the values of ε for each concentration normalized by ε at the same salt concentration 180 min after mixing.

From the start of data acquisition, there were substantial changes in the BSD of the doughs over the course of 150 min, with the radii showing increases that were typically in the range of 40–50% and with the widths decreasing by as much as 8%. We did not find any convincing evidence that salt reduction had a significant influence on the rate of change of bubble size and polydispersity with time. The large error bars in R_0, which resulted from the difficulty in pinpointing $f_{\max}$ when attenuation peaks were broad and were modulated by ripples resulting from noise, were one of the confounding factors making it difficult to delineate a possible dependence of the rates of change on salt concentration. It may still be worth noting that at the latest measurement time of 330 min the reduced-salt formulations appeared to have the most pronounced changes (the radius R_0 increased by 57 ± 13, 39 ± 7, and 35 ± 14% for the dough formulations containing 0.8, 1.6, and 2.4% sodium chloride, respectively, and ε decreased by 8.5, 6.4, and 3.7% for the least to most salt, respectively). These results for all salt concentrations were consistent with changes in the BSD arising from disproportionation (Shimiya and Yano 1988; Kokelaar et al 1996; Shimiya and Nakamura 1997; Murray and Ettelaie 2004). An increase in the relative median bubble size and a decrease in the width as time progressed in unyeasted doughs have been previously reported (Shimiya and Nakamura 1997; Leroy et al 2008b; Strybulevych et al 2012). Using light microscopy, Shimiya and Nakamura (1997) reported a 153% increase in median bubble radius and an 11.1% decrease in width over the course of 157 min in tests that started soon (3 min) after mixing. Their data showed an increase in bubble size that was 3–4 times larger than our observations over a later time interval of similar duration. Using an ultrasonic transmission technique, Leroy et al (2008b) reported a 29% increase in median bubble radius and a 4.6% decrease in width for doughs tested 53 min after mixing, over the course of 45 min. The results of Leroy et al (2008b) translate into approximately half the change in the median bubble size in one-third of the time when compared with our results. Although one should be careful in the comparison of results for different doughs prepared under different conditions, the results from these studies clearly support the idea that the rate of change in the BSD in dough slows down as the dough is aged.

CONCLUSIONS

An ultrasonic transmission technique has been described for measurement of bubble sizes and their time evolution to evaluate the effect of salt concentration on dough aeration. Frequency-dependent ultrasonic phase velocity and attenuation coefficient were seen to be sensitive probes of changes in the air volume fraction in dough that were brought about by a decrease in the salt concentration from 2.4 to 1.6%. Further reduction in salt concentration did not significantly influence the ultrasonic phase velocity or the attenuation coefficient. Analysis of the frequency dependence of these ultrasonic parameters showed that the typical sizes of the smallest bubbles in the BSD became smaller with salt reduction and that the width of the BSD increased as the salt concentration was reduced. The BSD exhibited a substantial time dependence for all salt formulations, even for relatively long times after mixing (3–5 h), with the changes in bubble sizes and distribution widths showing the classic signatures of disproportionation. Accordingly, ultrasonic transmission is a powerful tool in the breadmaking process for examining diffusively driven time-dependent changes in bubble concentration and their size distribution.

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