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Review

# Ultrasound as a tool to study bubbles in dough and dough mechanical properties: A review

Filiz Koksel [a,*], Martin G. Scanlon [a], John H. Page [b]

[a] Department of Food Science, University of Manitoba Winnipeg, MB, Canada, R3T 2N2
[b] Department of Physics and Astronomy, University of Manitoba Winnipeg, MB, Canada, R3T 2N2

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ABSTRACT

Investigation of dough mechanical properties using low-intensity ultrasound is now reasonably well established. In this review, an introduction to the fundamentals of ultrasound propagation in non-scattering and in scattering media is followed by several examples of how low-intensity ultrasound is used as a research tool for exploring the bubble size distribution in breadmaking dough and evaluating dough's mechanical properties. Utilization of ultrasonic techniques for quantitative assessments of bubbly dough structure and characterization of dough mechanical properties as affected by dough formulation are pointed out.



**Contents**



## 1. Introduction

Like many foods, bread owes its appeal largely due to the quality of its aerated structure (Campbell & Martin, 2012). Accordingly, understanding aeration processes during breadmaking has been the focus of several studies, beginning with the pioneering work of Baker and Mize (1941) published >70 years ago. At several steps of breadmaking, the dough matrix and the gas phase of the bread dough interact, and together they influence various "aeration" phenomena (*i.e.*, bubble entrainment, disentrainment, break-up, growth, disproportionation, coalescence), all of which affect the quality of the final product. Therefore, investigations of the mechanisms by which the gas phase is modified are a focal point for breadmaking unit process operations such as mixing (Baker & Mize, 1937, 1946; Campbell, Rielly, Fryer, & Sadd, 1998; Chin, Martin, & Campbell, 2005; Martin, Chin, & Campbell, 2004; Trinh, Lowe, Campbell, Withers, & Martin, 2013), sheeting (Leong & Campbell, 2008), fermentation (Babin et al., 2006; Bonny et al., 2004; Chiotellis & Campbell, 2003; Turbin-Orger et al., 2012) and baking (Babin et al., 2008; Dobraszczyk, 2004; Wagner, Quellec, Trystram, &

* Corresponding author.
*E-mail address:* Havva.KokselUstundag@umanitoba.ca (F. Koksel).

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Lucas, 2008; Whitworth, 2008). Given the importance of the gas phase to understanding how bread quality and dough process operations are controlled, a comprehensive grasp of bubbles and their influence on dough properties is desirable.

In a typical breadmaking process bubbles are introduced into the dough during mixing. Entrainment of gas bubbles during mixing is absolutely essential for the production of a high quality bread loaf; yeast produces carbon dioxide to grow the bubbles during fermentation, but yeast cannot create new gas bubbles, so that the bubbles incorporated during mixing are the sole nucleation sites for pooling of carbon dioxide as breadmaking proceeds (Baker & Mize, 1941).

The influence of dough aeration during mixing on dough's rheological properties has been intensively studied (Chin & Campbell, 2005a, 2005b; Chin et al., 2005). In their studies, Campbell and colleagues showed that not only does the concentration of bubbles in the dough at the end of mixing affect dough's mechanical properties, but the energy input necessary to optimally develop doughs is a function of dough's gas volume fraction (Chin et al., 2005). Even though there is no one optimal bubble size or concentration to bake the 'perfect' loaf of bread, it has been shown that strong flours produce doughs with a lower gas volume fraction at the end of mixing compared to weak flours, and doughs from strong flours produce loaves with finer cellular structures and higher volumes (Baker & Mize, 1946; Campbell, Rielly, Fryer, & Sadd, 1993; Hayman, Hoseney, & Faubion, 1998). The dough that emerges from the mixer is therefore a composite material (Scanlon & Page, 2015) comprised of highly compressible components – its air bubbles, and an essentially incompressible component – its dough matrix (Wang, Dai, & Tanner, 2006). As a result, the concentration of bubbles (Bloksma, 1981; Chin & Campbell, 2005a, 2005b; Chin et al., 2005), and perhaps their sizes (van Vliet, 1999), affect the dough's rheological properties. Consequently, the success of all subsequent processing operations performed on dough depends on the nature of the bubble size distribution in the dough.

Despite the need for a deeper understanding of how dough mechanical properties, aeration and breadmaking performance interact, the literature is not replete with techniques that are capable of bringing dough microstructure and rheology together. Several methods focus on investigation of dough's microstructure. These methods include light microscopy (Carlson & Bohlin, 1978), conventional bench-top X-ray microtomography (Bellido, Scanlon, Page, & Hallgrimsson, 2006), magnetic resonance imaging (De Guio, Musse, Benoit-Cattin, Lucas, & Davenel, 2009), and confocal laser scanning microscopy (Upadhyay, Ghosal, & Mehra, 2012). When the emphasis is on dough mechanical properties, descriptive (Farahnaky & Hill, 2007; García-Alvárez, Salazar, & Rosell, 2011; Kokelaar, van Vliet, & Prins, 1996; Linko, Härkönen, & Linko, 1984; Lynch, Dal Bello, Sheehan, Cashman, & Arendt, 2009; Van Steertegem, Pareyt, Brijs, & Delcour, 2013) and fundamental (Bloksma, 1973; Campos, Steffe, & Ng, 1997; Dobraszczyk, 2004; Hibberd & Wallace, 1966; Mastromatteo et al., 2013; Shewry, Popineau, Lafiandra, Belton, & Lellis, 2001; Upadhyay et al., 2012) rheological techniques have been extensively employed. A versatile technique, which has the potential of reconciling information on the microstructure of complex media and their rheology, and hence characterizing dough aeration as well as dough's mechanical properties, is low-intensity ultrasound (Elmehdi, Page, & Scanlon, 2003, 2005; Koksel, Strybulevych, Page, & Scanlon, 2014; Leroy, Fan, Strybulevych, Bellido, Page and Scanlon, 2008; Leroy, Pitura, Scanlon, & Page, 2010; Létang, Piau, Verdier, & Lefebvre, 2001; Scanlon, Page, Leroy, Elmehdi, Fan and Mehta, 2011; Scanlon, Elmehdi, Leroy, & Page, 2008).

An ultrasound wave is a mechanical wave which propagates as small deformations in the dough. By measuring the nature of ultrasonic propagation in a material, information about material properties can be obtained as a function of frequency (Coupland, 2004; McClements & Gunasekaran, 1997; Povey, 1997). Because the ultrasonic parameters can be measured over a range of frequencies, and since the waves of different frequencies are associated with different wavelengths, the interaction of ultrasound at specific frequencies with microstructural components of a given size permits structural investigations at several length scales. Furthermore, ultrasonic techniques are rapid and non-destructive and have proven to be well suited for studying optically opaque systems such as dough (Létang et al., 2001; Ross, Pyrak-Nolte, & Campanella, 2004; Scanlon et al., 2008; Strybulevych et al., 2012).

In this review, the fundamentals of ultrasonic propagation in wheat flour dough are reported. Dough is a complex material that is the base material for many food products. Understanding its mechanical behavior is therefore important for controlling the process operations conducted on it and for predicting the quality of end-products derived from it, the most important of which is bread. Because gas bubbles are an ingredient in all doughs, there is an emphasis on ultrasonic investigation of bubble structural information, and how bubbles, dough ingredients and dough processing conditions affect the mechanical properties of dough. The insights gained from ultrasonic investigations on dough have implications for processing of other aerated food products.

## 2. Principles of ultrasound propagation in dough

Longitudinal waves are the most common types of ultrasonic waves that are used for testing foods. There are other possible modes of ultrasonic propagation, such as shear waves or surface waves (McClements & Gunasekaran, 1997; Povey, 1997). However, these other types of waves are not so useful for the main focus of this review, which is to explore the use of ultrasound to study the bubbles distributed in the dough matrix. To pursue this goal, longitudinal waves are the most suitable since they are uniquely sensitive to bubbles and can propagate useful depths in fluids, weak gels and foods (Coupland, 2004; Kudryashov, Hunt, Arikainen, & Buckin, 2001; Saggin & Coupland, 2004). Throughout this review, unless otherwise stated, the ultrasonic techniques use longitudinal waves to probe the properties of dough and/or the bubbles in dough.

One of the simplest ways of representing acoustic waves is by using sinusoidal waves, as shown in Fig. 1a, which shows the oscillating displacements associated with a sinusoidal wave at a fixed position as a function of time. By using a sinusoidal wave, the three fundamental characteristics of acoustic waves – frequency ($f$), amplitude ($A$) and phase ($\varphi$) – can be demonstrated. Frequency is the number of times that a wave cycles per unit time and thus the temporal variation of a wave is determined by its frequency (Povey, 1997). Amplitude is the measure of the magnitude of the change in the positions of the molecules of a medium from their equilibrium positions as a result of sound propagation through the medium. Phase is a measure of the shift in the waveform with time and position relative to a reference point and is measured in radians or degrees. While Fig. 1a shows only the temporal variation of a sinusoidal wave at a fixed position, it is important not to forget that a traveling wave depends on both time and position. For a sinusoidal wave propagating in the positive $x$ direction, this spatio-temporal variation can be written:

$$\xi(x,t) = A\sin(\omega t - kx + \varphi)$$

where $\xi(x,t)$ is the displacement at position $x$ at time $t$, $\omega = 2\pi f$ is the angular frequency, $k = 2\pi/\lambda$ is the wavenumber, and $\lambda$ is the wavelength (the distance over which a snapshot of the waveform repeats). Here the phase constant $\varphi$ accounts for the possibility that the displacement may not be zero when both $x$ and $t$ are zero. As time advances, the position of any point on the wave (such as the crest of any one of the oscillations) also increases so that the quantity $\omega t - kx$ is constant, implying that the velocity of the phase oscillations in the wave, or the phase velocity, is given by $v = x/t = k/\omega$. The simplest type of wave is a plane wave, for which the displacement is the same in any given plane perpendicular to the direction of propagation.

A complex waveform, *i.e.*, a pulse, may depart drastically from a simple sinusoidal wave shape; however, no matter how complex a

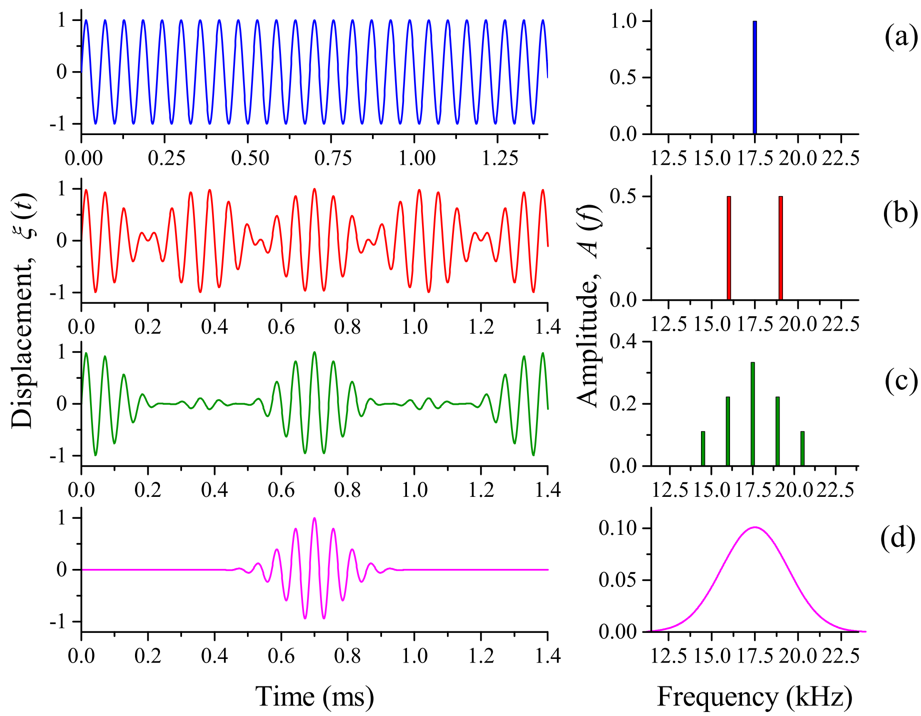


**Fig. 1.** (a) A continuous wave composed of a single sinusoidal component, (b) periodic pulses generated by the superposition of two sinusoidal waves with different frequencies, (c) periodic pulses generated by the addition of five frequencies; notice the increasing separation between pulses (b to c), (d) generation of an isolated pulse by the summation of a continuous distribution of frequencies.

waveform is, it can be broken down into combinations of sinusoidal waves of different frequencies, different amplitudes and different phases (Everest, 1989). In Fig. 1b and c, generation of periodic pulses by superposing sinusoidal waves of two or more frequencies is presented, while in Fig. 1d, generation of an isolated pulse by superposition of sinusoidal waves with a continuous distribution of frequencies is shown.

Information about a material's properties is contained in its ultrasonic velocity and attenuation coefficient (Coupland, 2004; McClements & Gunasekaran, 1997). Ultrasonic signal velocity is determined by time taken for the start of an ultrasonic pulse (*i.e.*, its initial onset) to travel a certain distance though a material, and is given by the ratio of this distance and time (Cobus, Ross, Scanlon, & Page, 2007). When the medium through which an ultrasonic pulse is traveling is dispersive, velocity is frequency dependent, so that using signal velocity to characterize a material's dynamic mechanical properties may introduce inaccuracies (Cobus et al., 2007). In such media, two additional distinct velocities - group velocity and phase velocity - are needed to describe ultrasound propagation (Page et al., 1996). Group velocity is the pulse velocity, which can be determined accurately, so long as the pulse is not distorted, by the arrival time of the peak of the pulse, whereas the phase velocity is the velocity of each of the frequency components that constitute the pulse. Phase velocity at a specific frequency can be determined using Fourier analysis, from the difference in the phase of the Fourier transform of the signal that has propagated through the sample relative to the input signal (Cobus et al., 2007). Throughout this review, unless otherwise stated, the experimental ultrasonic techniques used on doughs report the phase velocity ($v$). During ultrasonic wave propagation, the amplitude of the waves decay with distance travelled, *i.e.*, they are attenuated (Cobus et al., 2007). This decay is measured with the attenuation coefficient, $\alpha$ (Coupland, 2004; Povey, 1997). Attenuation is mainly caused by absorption and scattering. Absorption is observed in all media (both homogeneous and heterogeneous) due to heat conduction (thermal) losses, viscous losses, and molecular relaxation processes (McClements, 1991; McClements & Gunasekaran, 1997). Scattering is negligible in homogeneous media (McClements & Gunasekaran, 1997) and occurs when an ultrasonic wave comes across a discontinuity in velocity and density (*i.e.*, an inhomogeneity) and changes its direction(s) of propagation (McClements, 1991). Absorption mechanisms lead ultimately to the conversion of ultrasonic energy into heat while scattering alters the trajectory of the incident ultrasonic wave (McClements & Gunasekaran, 1997). The magnitude of different attenuation mechanisms is influenced by the thermal and physical properties of the constituent phases, the size and distribution of the inhomogeneities, and the frequency used (McClements, 1996).

The wavenumber ($k$) is a convenient way of expressing both the ability of the material under investigation to transport the strain induced by an external excitation (as a traveling ultrasonic wave) and how much the material dissipates the ultrasonic energy as the wave propagates through (Cobus et al., 2007), so that the wavenumber is complex, $k = k' + ik''$ (Leighton, 1997). The real part of $k$ is equal to the angular frequency, $\omega$ ($= 2\pi f$), divided by $v$, while the imaginary part of $k$ can be written in terms of $\alpha$ (Cobus et al., 2007).

$$k = \omega/v - i(\alpha/2) \quad (1)$$

In this review, we have chosen to adopt the convention for $\alpha$ in which it is defined as the intensity attenuation coefficient, since much of the focus is on scattering, whereby the intensity attenuation coefficient is the reciprocal of the scattering mean free path. With this convention, $k'' = -\alpha/2$. At an interface between two media, part of the ultrasonic energy is reflected whereas part is transmitted if the acoustic properties of two media are different (McClements & Gunasekaran, 1997). The relevant difference between the acoustic properties of two media is the specific acoustic impedance, $Z$, which is complex (Leighton, 1997). This characteristic impedance $Z$ for ultrasonic waves is analogous to the electromagnetic impedance for light waves (related to the refractive index) and is both a property of the material and of the type of wave traveling through it (Kleppe, 1989). For a plane wave, $Z$ is related to the wavenumber, $k$, as:

$$Z = \rho\omega/k \quad (2)$$

where $\rho$ is the density of the material through which the ultrasound is propagating. Combining Eqs. 1 and 2 gives the relationship between $Z$, $v$ and $\alpha$ as:

$$Z = \rho v / \left(1 + i\frac{v\alpha}{2\omega}\right) \tag{3}$$

For some foods, the imaginary part of $Z$ is very small compared to the real part ($\alpha << \omega/v$) so that, as a first approximation $Z = \rho v$ (McClements, 1991). Bread dough strongly attenuates ultrasound, especially around the resonance frequencies of bubbles that it contains (see Section 2.2.2.1, Scattering from bubbles and the resonance phenomenon), and thus this approximation cannot be made for bread dough in the ultrasonic frequency range.

Depending on the material under investigation, a particular ultrasonic technique such as transmission or reflection may be of interest (McClements, 1991). In highly attenuating media, such as bread dough, ultrasonic wave propagation over a wide range of frequencies may be challenging in thick samples (Fan, Scanlon, & Page, 2013; Leroy, Fan et al., 2008; Létang et al., 2001; Scanlon et al., 2008). When this is the case, reflection techniques may be preferred over transmission techniques for studying the acoustic properties of highly attenuating media (Povey, 1997). However, reflection techniques have the disadvantage that only surface properties can be interrogated (Coupland, 2004), with the ultrasonic properties being extracted from the part of the pulse that is reflected from the surface of the sample being tested. Accordingly, even with limitations of signal loss, transmission techniques have the advantage that the properties of a sample as a whole can be interrogated since the ultrasonic parameters are extracted from the part of the pulse that is transmitted into and through the sample.

A typical experimental set-up for ultrasonic transmission tests (within the MHz frequency range) for bread doughs is presented in Fig. 2. Acquisition of the signal that propagates through the dough sample and acquisition of the reference signal are carried out as shown in the right and left sides of Fig. 2, respectively. In both cases, the first (generating) transducer converts a voltage pulse that is sent from the pulse generator to an ultrasonic pulse (Awad, Moharram, Shaltout, Asker, & Youssef, 2012; Khairi, Ibrahim, Yunus, & Faramarzi, 2015). The ultrasonic pulse first travels through an acrylic delay line that is thick enough to separate the initial noise created when the ultrasound signal is generated. For the reference signal, two delay lines are in contact so that the ultrasonic pulse propagates through the first and the second delay line, and then reaches the second (receiving) transducer. For the signal through the dough, the dough (<1 mm thickness) is sandwiched between the two delay lines so that the ultrasonic pulse propagates through the first delay line, then the dough sample, and then the second delay line. The signal that has propagated through the sample and the two delay lines reaches the receiving transducer where it is converted to a voltage pulse. The received signals are amplified by the pulse receiver and then stored for analyses (Koksel et al., 2014). As indicated above, the preferred method of analysis for acquisitions in the MHz frequency range involves taking the Fast Fourier Transforms (FFT) of the pulses, since then, over the bandwidth of the transducers, the phase velocity can be determined from the phase difference between sample and reference signals, and the attenuation from the FFT amplitude ratio once corrections for reflections at the input surfaces are applied.

Finally, it is worth mentioning that because transducers have a finite diameter, the ultrasonic signals are not simple plane waves, but spread out with distance from the source due to diffraction effects. For accurate measurements, it is often necessary to make corrections for the resulting diffraction losses, especially at low frequencies where transducers may only be a few wavelengths in diameter (Breazeale, Cantrell, & Heyman, 1981; Papadakis, 1975). These corrections depend on the ratio $x\lambda/a^2$, where $x$ is the propagation distance, $\lambda$ is the wavelength, and $a$ is the radius of the transducer. However, the very high attenuation in materials such as bread dough means that samples must be very thin in order for transmitted signals to be sufficiently large to be detected, and as a result, diffraction effects can generally be neglected. They will therefore not be discussed further here.

### 2.1. Ultrasound propagation in non-scattering systems

Wave propagation through non-scattering systems is relatively simple compared to that in scattering systems (Javanaud, 1988). Single-phase or homogeneous materials, *e.g.*, fluid foods such as wine and honey, do not scatter ultrasound, whereas multi-phase or heterogeneous foods such as bread dough, ice-cream and tofu do.

The attenuation in non-scattering materials is due to absorption since these materials do not scatter ultrasound because they do not contain any discontinuities (McClements, 1991). For a longitudinal ultrasonic wave traveling in a non-scattering material, the amplitude (proportional to the square root of intensity) of an ultrasonic wave, $A$, decreases exponentially according to the equation:

$$A = A_0 e^{-\alpha x/2} \tag{4}$$

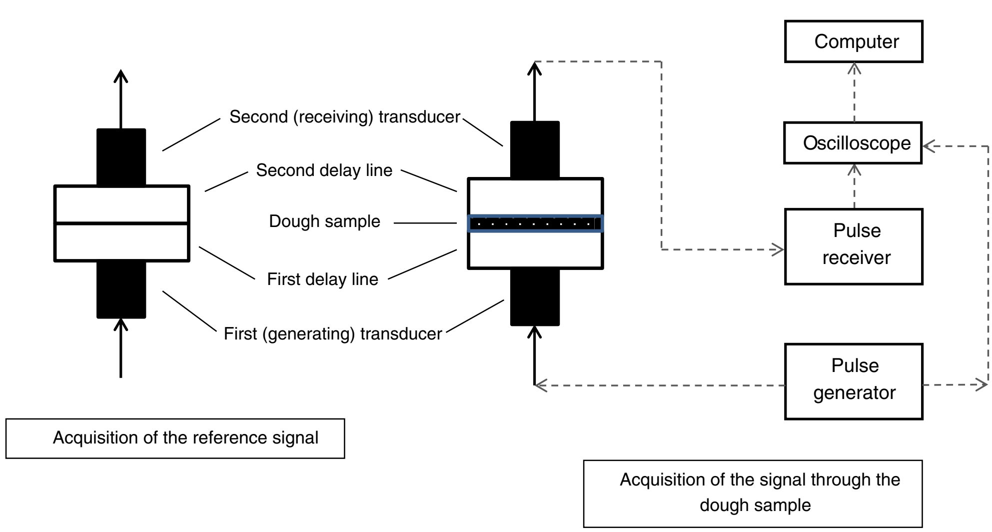


**Fig. 2.** Experimental set-up for testing dough samples in transmission mode (Reproduced, by permission of AACC International, from Koksel, F., Strybulevych, A., Page, J. H., and Scanlon, M. G. 2014. Ultrasonic characterization of unyeasted bread dough of different sodium chloride concentrations. Cereal Chem. 91:327-332).

where $A_0$ is the ultrasonic wave's amplitude at position $x = 0$ and $x$ is the distance from the surface of the material.[1]

For non-scattering materials in which attenuation is negligible (*i.e.*, $\alpha \ll \omega/v$), the velocity of sound is independent of frequency and is solely determined by the elasticity and density of the material through which it is propagating (Povey, 1997). At a specific frequency, the wavenumber, $k$, is related to the longitudinal modulus, $M$, of the material under investigation through the relation:

$$v = \omega/k = \sqrt{M/\rho}, \tag{5}$$

which has been referred to as Wood's equation (Povey, 1997). For homogeneous liquids and gases, $M$ is equal to the bulk modulus of elasticity, $B$, which is the inverse of the adiabatic compressibility, $\kappa_S$. In solids, the propagation of ultrasound is more complex compared to fluids, since solids can support shear waves in addition to longitudinal waves (Kudryashov et al., 2001; Saggin & Coupland, 2004). For solids, $M$ is a combination of $B$ and the shear modulus ($G$):

$$v = \sqrt{(B + 4G/3)/\rho} \tag{6}$$

Consequently, provided that the density is measured independently, by measuring the ultrasonic velocity, the longitudinal modulus of a material, which is equal to the bulk modulus for liquids and a combination of the bulk and the shear moduli for solids, can be obtained (Cobus et al., 2007). With a similar approach, the longitudinal viscosity ($\eta_L$), a measure of dissipation of the acoustic energy associated with longitudinal wave propagation, can be obtained (Dukhin & Goetz, 2009). This approach has found some application in investigations of fruit juice properties (Laux, Gibert, Ferrandis, Valente, & Prades, 2014; Laux et al., 2013). When density is measured independently, longitudinal viscosity can be obtained by measuring the ultrasonic velocity and attenuation coefficient ($\eta_L = M''/\omega \approx 2\alpha\rho v^3/\omega^2$ where $M''$ is the imaginary part of the complex longitudinal modulus and $M'' = B'' + 4G''/3$), for low attenuating materials (Dukhin & Goetz, 2009; Laux et al., 2013).

### 2.2. Ultrasound propagation in scattering systems

Ultrasonic propagation in scattering (heterogeneous) media is more complex compared to that in non-scattering media. Bread dough is a great example of a scattering system, since it is composed of gas bubbles entrained into the dough during mixing, within a matrix of hydrated wheat flour (mostly wheat starch and gluten proteins), yeast, dissolved salt and some other ingredients (depending on the type of bread). Gas bubbles are especially important in ultrasonic investigations of bread dough due to their effect on ultrasound propagation, and for this reason, the focus of this review is on gas bubbles as the scatterers of ultrasound in dough.

In practice, ultrasonic propagation in a medium with scatterers can conveniently be described by dividing the ultrasonic spectrum into categories pertaining to scatterer size (McClements, 1996): (*i*) low frequency regime (wavelength, λ, is very much larger than scatterer size), (*ii*) intermediate frequency regime (strong, resonant scattering region), and (*iii*) high frequency regime (wavelength is much smaller than scatterer size) (Fig. 3). In this review, the focus is on ultrasonic investigations of bread dough and bubbles in bread dough so that we discuss the effective medium properties of the dough matrix and bubbles in the low frequency regime, the bubble size distribution in dough in the intermediate frequency regime and the properties of the dough matrix in the high frequency regime. For more information on diffusive wave transport in the intermediate frequency regime where rigid particles are of interest, readers are referred to Page, Schriemer, Bailey, and Weitz (1995).

#### 2.2.1. Low frequency regime

In the low frequency regime (λ>>scatterer size), attenuation is not only due to intrinsic absorption but also due to absorption caused by the viscous and thermal damping mechanisms associated with scattering (Cents, Brilman, Versteeg, Wijnstra, & Regtien, 2004; McClements, 1991). In this regime, total intrinsic absorption is obtained by adding the absorption that occurs in each phase of the multi-phase material, and it depends on the volume fraction of these phases and not on the size of the scatterers (Cents et al., 2004). Viscous and thermal scattering occurs at the boundary between a scatterer and the medium surrounding it, *e.g.*, at the boundary between a gas bubble and its surrounding bread dough matrix. Both viscous and thermal scattering can substantially influence the velocity and attenuation of ultrasound because both the amplitude and the phase of the incident wave change as a result of scattering (McClements, 1991). Viscous scattering is due to the density difference between the scatterer and the medium surrounding it, *e.g.*, in the case of air bubbles in bread dough, the density of gas-free bread dough is approximately 1000 times greater than the density of air. This density difference causes the scatterer, *i.e.*, the bubble, to oscillate because the inertia of the scatterer is not the same as that of the medium surrounding it. As a result, a fraction of the incident wave is scattered, and, due to the damping of the movement of the scatterer by the viscosity of the medium, a fraction of the ultrasonic energy is converted into heat (Cents et al., 2004; McClements, 1996). Thermal scattering occurs because of thermal fluctuations as an ultrasonic wave propagates through a medium. These thermal fluctuations occur when the thermal properties of scatterers and the medium surrounding them are different so that as sound is traveling through, the change in temperature per unit volume within the scatterer and within the medium surrounding it are different. This temperature differential acts as a driving force for heat transfer through the interface between the scatterer and the medium surrounding it and leads to loss of some of the ultrasonic energy as heat (Cents et al., 2004; McClements, 1996).

In the low frequency regime, an effective medium approach has been shown to describe wave propagation reasonably well in bubbly liquids (Wilson, Roy, & Carey, 2005) and viscoelastic media such as bubbly gels (Leroy, Fan et al., 2008; Leroy, Strybulevych, Page & Scanlon, 2008; Strybulevych, Leroy, Scanlon, & Page, 2007). An effective medium approach has also been shown to be suitable for describing wave propagation in bread dough (Mehta, Scanlon, Sapirstein, & Page, 2009). According to effective medium theory (EMT), in the low frequency regime, inhomogeneous media can be considered to a good approximation as equivalent homogeneous media with modified properties. This approximation holds provided that the effects of scatterers can be accounted for by the use of modified 'effective' properties such as the 'effective compressibility' and 'effective density' of the medium (Gaunaurd & Uberall, 1981). In such cases, for example for bread dough, although the ultrasonic velocity strongly depends on the volume fraction of the gas bubbles in the dough, their size distribution is not a factor, and Eq. (5) is still valid (Wilson & Roy, 2008). At such long wavelengths, the velocity is determined by the effective compressibility and density of the medium, which can be estimated by their volume-fraction-weighted averages. This simple effective medium approximation accounts for the fact that the effective compressibility of a bubbly mixture is dominated by the highly compressible bubbles, while the effective density is dominated by matrix, leading to a remarkably slow sound velocity that can be less than the velocity of either component on its own. This model was first proposed by Wood (1941) for fluid mixtures such as bubbles in a liquid, and later generalized to also include dispersions of solid scatterers by Urick (1947). Hence, these effective

[1] Note that the attenuation coefficient defined by Eq. (4) is the attenuation coefficient that characterizes the exponential decay of the intensity ($I = I_0 e^{-\alpha x}$), which is proportional to the square of the amplitude.

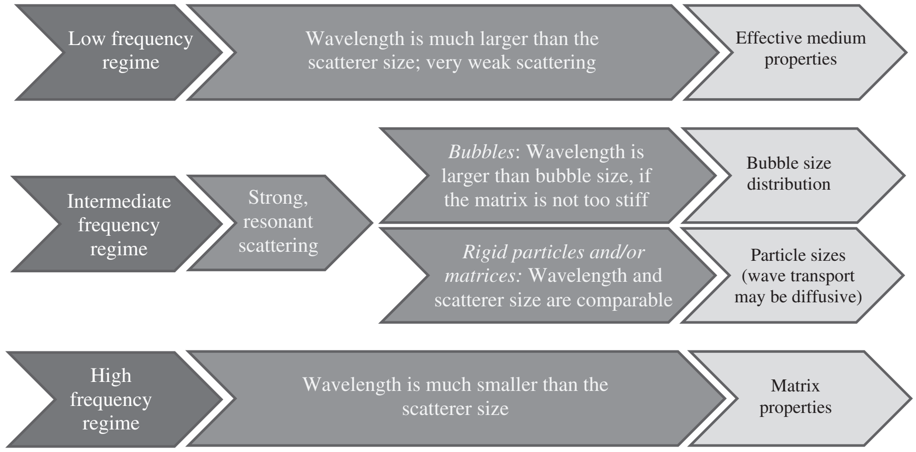


**Fig. 3.** Ultrasonic spectrum pertaining to scatterer size and wavelength. Material properties that can be determined from ultrasound measurements are indicated on the right.

medium predictions for the velocity of sound ($v$) in multi-phase media are (Javanaud, 1988):

$$v = 1/\sqrt{\kappa_S \rho}, \text{with } \kappa_S = \text{ and } \rho = \sum_j \phi_j \kappa_{Sj} \sum_j \phi_j \rho_j \tag{7}$$

Here $\kappa_S$ is the effective compressibility of the dough, $\rho$ is its effective density, $\phi_j$ is the volume fraction of each phase and subscript $j$ represents different phases (Povey, 1997). Eq. (7) should only be applied to bread dough on the assumption that its shear modulus is negligible. Then, if we consider a population of gas bubbles occluded into bread dough during mixing, with gas volume fraction $\phi$, the velocity of sound in the bread dough is related to the dough's gas volume fraction as:

$$v = 1/\sqrt{[(1-\phi)\kappa_{S,m} + (\phi\kappa_{S,g})]\left[(1-\phi)\rho_m + \left(\phi\rho_g\right)\right]} \tag{8}$$

where subscripts $m$ and $g$ denote the dough matrix and gas, respectively (Mehta et al., 2009). This equation considers that the multi-phase medium such as dough behaves like an effective non-scattering homogeneous medium and does not consider the interaction of the individual gas bubbles with the ultrasound wave (Urick, 1947). Calculation of an 'effective' attenuation coefficient for a multi-phase medium is somewhat more complex (Javanaud, 1988).

#### 2.2.2. Intermediate frequency regime

In the intermediate frequency regime, ultrasonic waves are scattered strongly due to resonances. For rigid particles and/or matrices (Fig. 3), resonances occur when the wavelength and scatterer size are comparable [for example, see Strybulevych et al. (2007)]. However, the resonances are unusual for bubbles and occur at lower frequencies. At intermediate frequencies, the general mathematical theory used for describing wave propagation is more complex, and the attenuation coefficient and velocity are scatterer size dependent (McClements, 1991). In this review of literature, ultrasound propagation in the bubble resonance region is discussed and scatterers are bubbles unless stated otherwise.

As concentrations of scatterers in a system increase, the scatterers become more closely packed. Then the probability of a wave scattered at a discontinuity propagating as far as another discontinuity increases (unless the wave is completely absorbed) so that the scattered wave can be re-scattered. As a result, the number of scattering occurrences can become very high leading to a very complex scattering behavior for the system (McClements, 1991). This process, known as multiple scattering, depends on many factors, including how strongly ultrasound is scattered, the concentration of the scatterers, and how strongly the scattered waves are absorbed (Povey, 1997).

Every time a scattering event occurs, acoustic energy is redirected and the scattered wave becomes slightly out of phase with the wave it arises from (Povey, 1997). This makes the interpretation of ultrasonic measurements very challenging in highly concentrated systems because the solution to the wave propagation equation usually is a series equation composed of terms that are a function of powers of the volume fraction of the scatterers, restricting the application of such solutions to dilute systems (Javanaud, 1988).

Sound propagation in highly scattering systems can be simplified by first calculating the scattering attributes for a single scatterer, and then for a collection of scatterers by combination of scattering from the individual scatterers (McClements, 1991). One of the traditional approximations used to describe wave propagation in inhomogeneous media containing randomly distributed scatterers is the method developed by Foldy (1945). According to Foldy's (1945) model, the incident wave and the waves scattered to the forward direction are assumed to form a different new wave which travels without scattering and with a velocity different than that of the incident or the scattered waves. Foldy's model assumes that the scattering events are not affected by neighboring scatterers (the independent scattering approximation), making it one of the simplest models with satisfactory predictions of wave propagation in inhomogeneous media (Leroy, Strybulevych et al., 2011). Thus, Foldy's model is only expected to give accurate predictions for systems with a low concentration of scatterers, and therefore, it is not always applicable when the concentration of scatterers is high or when the scattering is strong. For bread dough at the end of mixing, despite its high bubble concentration (approximately >10%), Foldy's model was shown to describe ultrasonic velocity and attenuation coefficient reasonably well (Leroy, Fan et al., 2008). However, when the multiple scattering problem becomes complicated because the amplitudes of the scattered waves are significant, coupling between scatterers or the coupling between the scattered waves can no longer be neglected (McClements & Gunasekaran, 1997). When that is the case, positional correlations of the scatterers and multiple scattering patterns may have a significant influence on wave propagation (Leroy et al., 2011), and when the bubble concentration is very high, the nature of the bubble resonances may be modified (Pierre, Elias, & Leroy, 2013).

*2.2.2.1. Scattering from bubbles and the resonance phenomenon.* The presence of bubbles can substantially affect sound propagation in a system due to a phenomenon called resonant scattering (McClements, 1991). Air bubbles in water scatter sound significantly even at low concentrations (as little as 0.1% by volume) (Ben Salem et al., 2013), and hence bubbly water can become highly dispersive, *i.e.*, ultrasonic attenuation coefficient and phase velocity become strongly frequency dependent

(Leighton, 1997). For example, the low frequency sound velocity in a mixture of 1% air in water is approximately 120 m/s, lower than that in air (~330 m/s) and that in pure water (~1500 m/s), whereas at high frequencies above the bubble resonance the velocity approaches that of water (Povey, 1997). The depression of sound velocity with small volume fractions of bubbles is also observed for bubbly gels (Leroy, Fan et al., 2008; Leroy et al., 2011; Leroy, Strybulevych et al., 2008; Strybulevych et al., 2007) and bread dough (Koksel et al., 2014; Leroy, Fan et al., 2008; Scanlon, Page, Leroy, Elmehdi et al., 2011).

When sound waves propagate through a bubbly medium, bubbles start to oscillate and the dynamics of a bubble can be explained using a damped harmonic oscillator model (Leroy, Devaud, & Bacri, 2002). For a bubble to oscillate at its natural frequency, energy input is required (Leighton, 1997). For ultrasonic experiments, the bubbles are excited into oscillation by an incident ultrasound wave. After an initial displacement, the bubble pulsates as the bubble wall oscillates. This is like a frictionless bob-on-a-spring system where the height of the bob changes harmonically after the initial displacement. In the bob-on-a-spring system, the spring provides the restoring force whereas in the bubble in a liquid system, the restoring force is due to the compressibility of the gas in the bubble. As the wave propagates, the gas in the bubble expands and contracts as a result of its large compressibility compared to the medium surrounding it (Leighton, 1997). In both the spring and bubble systems, the oscillation is harmonic, occurring at a natural resonance frequency (Leighton, 1997).

The natural resonance frequency of a gas bubble in a liquid experiencing low amplitude periodic oscillations was first explained by Minnaert (1933). Resonance of a spherical gas bubble in a liquid occurs specifically at a low frequency which is known as the Minnaert resonance frequency, $\omega_M$ (Leroy et al., 2011):

$$\omega_M = \frac{v_g\sqrt{3\rho_g/\rho_l}}{R} \tag{9}$$

where $R$ is the bubble radius, $v_g$ is the velocity of ultrasound in the gas, and $\rho_g$ and $\rho_l$ are the densities of the gas and the liquid, respectively. The density of gas and the ultrasonic velocity in the gas are small compared to those of the liquid (Povey & McClements, 1988), so the Minnaert resonance occurs at a very low frequency (Leroy et al., 2011):

$$\frac{\omega_M R}{v_l} = \frac{v_g\sqrt{3\rho_g/\rho_l}}{v_l} << 1 \tag{10}$$

where $v_l$ is the velocity of ultrasound in the liquid. Since $v = f\lambda$, Eq. (10) implies that, at resonance, the bubble size is very small compared to the wavelength ($2\pi R << \lambda$). This observation was confirmed by Strybulevych et al. (2007) who compared the wavelength at the resonance frequency to the size of the bubble, and showed that resonance occurs when $R$ is much smaller than the wavelength for a 0.75% volume fraction of air bubbles trapped in a model food gel (Fig. 4a). They reported that for a median bubble radius of about 0.1 mm (Fig. 4b) (resonance frequency ~ 100 kHz) the corresponding wavelength was 15 mm while the sound velocity in the gel was the plateau velocity of 1500 m/s, evident at the high frequency end of Fig. 4d. The same conclusion can be deduced from the nonyeasted bread dough results of Koksel et al. (2014) where the resonance frequency was approximately 2.5 MHz; the corresponding ultrasonic wavelength at resonance is much larger than the median bubble size in nonyeasted bread doughs (in the range of 100 to 110 μm, as measured by bench-top X-ray microtomography) (Bellido et al., 2006).

For a single bubble oscillating freely in a liquid, the frequency of these oscillations, *i.e.*, the Minnaert resonance frequency, is derived by equating the potential energy built up during the compression of the oscillating bubble until the bubble reaches its smallest volume and the kinetic energy built up by the surrounding liquid when the bubble regains its equilibrium volume (Minnaert, 1933). Accordingly, the Minnaert resonance frequency can be written as:

$$\omega_M = \sqrt{\frac{3\kappa p_0}{\rho_l R^2}} \tag{11}$$

where $\kappa$ is the polytropic index of the gas in the bubble - a number that describes a thermodynamic process following the relation $pV^\kappa = C$ where $p$ is pressure, $V$ is volume and $C$ is a constant - and $p_0$ is the static pressure of the gas in the bubble (Leroy et al., 2011). This formula relates $\omega_M$ of a bubble to its size as well as to the physical properties of the gas in the bubble and the density of the surrounding medium (Carstensen & Foldy, 1947).

For the Minnaert equation, it is assumed that the bubble surface is completely clean. In practice, the bubble surface always carries some contamination or surface active materials. This is especially true for bread dough, considering that proteins, lipids and other bread ingredients compete for the bubble-dough matrix interface (Kokelaar & Prins, 1995), and these surface-active materials will affect $\omega_M$. For instance, the $\omega_M$ of a 10 μm bubble at 10 cm depth in seawater increased about 8% when a film of aliphatic alcohol was adsorbed on to the surface of the bubble (Glazman, 1983).

Below $\omega_M$, ultrasonic velocity is significantly smaller than that in the surrounding medium (in the low frequency regime). Around $\omega_M$, the velocity increases significantly, while at frequencies much higher than $\omega_M$ it approaches the velocity of the surrounding medium (Fig. 4d) (Leroy, Fan et al., 2008; Strybulevych et al., 2007). Near $\omega_M$, the attenuation increases sharply (Fig. 4c) (Povey & McClements, 1988; Strybulevych et al., 2007). The attenuation peaks at the $\omega_M$ of the bubbles if the bubbles are of a single size (Carstensen & Foldy, 1947), while for a polydisperse system - as in the case of bread dough - the peak in attenuation is spread out over a wide frequency range, extending to frequencies where the corresponding wavelength is similar to the mean size of the separation between bubbles (Leroy, Fan et al., 2008). Consequently, as bubble concentration increases, *i.e.*, as the mean separation between bubbles decreases, the attenuation peak broadens.

*2.2.2.2. Theoretical models describing the propagation of ultrasound near the bubble resonance frequency in systems containing bubbles.* As stated by Foldy (1945), if the scatterers are bubbles, the total scattering is equivalent to the scattering from one single bubble times the number of bubbles, and these scattering events are independent of the positions of the bubbles. According to Foldy's model, the wavenumber of the medium with a monodisperse population of bubbles is:

$$k^2 = \left(\frac{\omega}{v} + i\frac{\alpha}{2}\right)^2 = k_0^2 + 4\pi n f(\omega) \tag{12}$$

where $k_0$ ($=\omega/v_0$) and $v_0$ are the wavenumber and the velocity, respectively, of waves in the scatterer-free medium, $n$ is the bubble concentration, and $f(\omega)$ is the scattering function at angular frequency, $\omega$. For a polydisperse population of bubbles, Eq. (12) can be modified as:

$$k^2 = k_0^2 + \int [4\pi n(R)dRf(\omega, R)] \tag{13}$$

where $n(R)dR$ is the number of bubbles per unit volume (for bubbles with a radii between $R$ and $R + dR$), and $f(\omega,R)$ is the scattering function, a function of angular frequency, $\omega$, and bubble radius, $R$.

It has been shown that bubbles occurring naturally are well characterized by functions within the logarithmic family (commonly the lognormal distribution function) (Limpert, Stahel, & Abbt, 2001; Proussevitch, Sahagian, & Tsentalovich, 2007). The bubble size distribution in dough at the end of mixing has also been characterized as lognormal (Bellido et al., 2006; Shimiya & Nakamura, 1997), due to the random repetitive subdivision of bubbles into smaller sizes. Accordingly, it is reasonable to

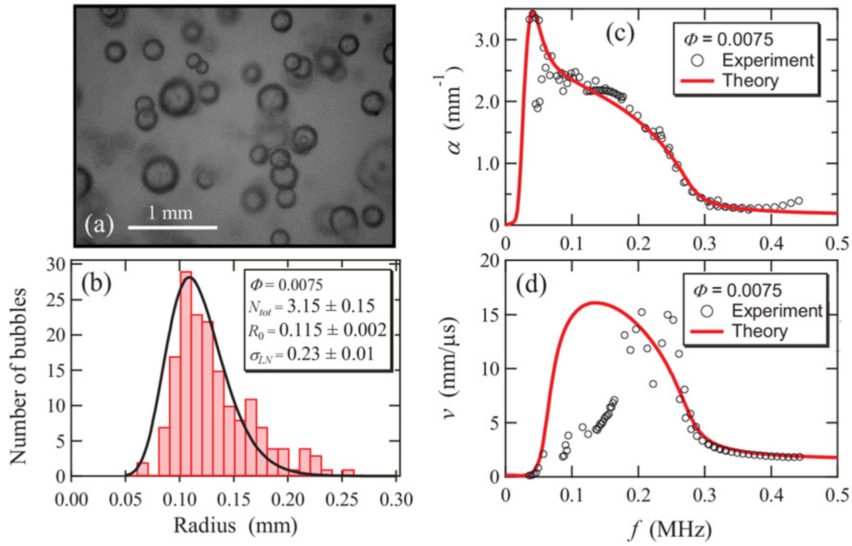


**Fig. 4.** (a) Sample image for bubbles in a 2% agar gel, (b) size distribution of the bubbles (volume fraction of bubbles = 0.75%, bubbles are lognormally distributed with a median of 0.115 mm), (c) attenuation coefficient, and (d) phase velocity as a function of frequency (Republished with permission of Royal Society of Chemistry, from Characterizing a model food gel containing bubbles and solid inclusions using ultrasound, Strybulevych, A., Leroy, V., Scanlon, M.G., Page, J.H., Vol 3, 2007; permission conveyed through Copyright Clearance Center, Inc.).

expect that the bubble size distribution in dough is given by the lognormal distribution, which is one of the most commonly used probability distribution functions:

$$n(R) = \frac{n}{\sqrt{2\pi}\varepsilon R}\exp\left\{-\frac{(\ln R/R_0)^2}{2\varepsilon^2}\right\} \tag{14}$$

Here $n$ is the bubble concentration, $\varepsilon$ is the width of the distribution, and $R_0$ is the median bubble radius of the lognormal bubble size distribution (BSD). When $R << \lambda$, the scattering function in Eq. (13) can be represented as (Leroy, Strybulevych et al., 2008):

$$f(\omega, R) = \frac{R}{(\omega_M/\omega)^2 - 1 + i\left(\delta^{thermal} + \delta^{viscous} + \delta^{radiative}\right)} \tag{15}$$

where $\omega_M$ is the Minnaert frequency and $\delta^{thermal}$, $\delta^{viscous}$ and $\delta^{radiative}$ represent the damping constants of the bubble due to thermal (Eq. (16), viscous (Eq. (17), and radiative (Eq. (18) energy losses, respectively.

$$\delta^{thermal} = \frac{3\Im(\kappa)p_0}{\rho R^2\omega^2} \tag{16}$$

$$\delta^{viscous} = \frac{4G''}{\rho R^2\omega^2} \tag{17}$$

$$\delta^{radiative} = Rk_0 \tag{18}$$

where $\Im$ designates the imaginary part, $\rho$ is the density of the medium surrounding the bubbles, *i.e.*, the density of the dough matrix for a bread dough system, $G''$ is the imaginary part of the complex shear modulus of the medium surrounding the bubbles, and $\kappa$ is the complex polytropic index of the gas. At resonance, the oscillating bubble's inertia depends on the properties of the medium surrounding it (Eq. (17), because the bubble's movements will be damped due to the work done against viscous forces at the interface between the bubble and the medium (Leighton, 1997). As the bubble oscillates and as its volume changes, the temperature and the pressure of the gas inside it will polytropically change, *i.e.*, the process will be adiabatic if there is no heat transfer between the bubble and the medium surrounding it, whereas the process will be isothermal if as much heat flows into the bubble upon expansion as flows out upon compression (Leighton, 1997).

Prosperetti (1977) modeled the thermodynamic behavior of the gas in the bubbles to describe the thermal interaction of the bubble with its surroundings and defined the complex polytropic index, $\kappa(\omega,R)$, as a function of angular frequency and bubble size:

$$\kappa(\omega, R) = \frac{\gamma}{1 - 3(\gamma-1)i\dfrac{D_{th}}{\omega R^2}\zeta} \tag{19}$$

with

$$\zeta = 1 - \sqrt{i}R\sqrt{\frac{\omega}{D_{th}}}\coth\left(\sqrt{i}R\sqrt{\frac{\omega}{D_{th}}}\right) \tag{20}$$

where the real part of the complex polytropic index is equal to the ratio of the heat capacities of the gas in the bubble, $\Re(\kappa) = \gamma$ where $\Re$ designates the real part, $D_{th}$ is the thermal diffusivity of the gas in the bubble and $\sqrt{i}$ stands for $e^{i(3\pi/4)}$. The oscillation of an air bubble in water approaches the adiabatic limit ($\kappa = \gamma = 1.4$) when $\sqrt{D_{th}/\omega} < R$ (Eq. (19)) (Prosperetti, 1977), which has also been reported to hold for polydisperse air bubbles (median bubble radius of the order of 80 μm) in an elastic medium (at frequencies up to 0.4 MHz) (Leroy et al., 2011).

If the wave is propagating in viscoelastic media, as is the case for bread dough, $\omega_M$ is modified by the shear modulus ($G$) of the dough matrix, and moves to higher frequencies (Strybulevych et al., 2007). Attenuation is enhanced as well (Strybulevych et al., 2007), *e.g.*, see Eq. (17) for the effect of $G''$ on the viscous energy losses. A good approximation of $\omega_M$ in viscoelastic media is (Alekseev & Rybak, 1999):

$$\omega_M = \sqrt{\frac{3\Re(\kappa)(p_0 + 2T/R) + 4G'}{\rho R^2} - \frac{2T}{\rho R^3}} \tag{21}$$

where $G'$, for bread dough, is the real part of the complex shear modulus of the dough matrix, and $T$ is the surface tension of the interface between the gas bubble and the dough matrix. For an aqueous yield stress fluid containing bubbles of similar size range as in nonyeasted bread dough (Bellido et al., 2006), Leroy, Strybulevych et al. (2008) showed that the first part of Eq. (21), $[(3\Re(\kappa)(p_0 + 2T/R) + 4G')/\rho R^2]$, is approximately two orders of magnitude greater than the second part, $[2T/\rho R^3]$.

After measuring $v$ and $\alpha$, the size distribution of scatterers, *i.e.*, bubbles, is determined, provided that a satisfactory correspondence between the experimental results and theory predictions is obtained. The angular frequency divided by the real part of the wave vector gives the theory prediction for $v$ and twice the imaginary part of the wave vector gives the theory prediction for $\alpha$ (Strybulevych et al., 2007):

$$v = \omega / \Re(k) \tag{22}$$

$$\alpha = 2\Im(k) \tag{23}$$

The approach in determining the size distribution assumes that the BSD conforms to a common probability density function with a simple mathematical equation, *e.g.*, lognormal distribution (Leroy, Fan et al., 2008) (Eq. (14)). However, problems may arise with this approach if the assumed probability density function does not represent the size distribution of scatterers reasonably well (McClements, 1996). Then it becomes more difficult to determine the bubble size distribution from experimental data. A complete and precise approach to check model predictions against the experimental results is to feed the discrete bubble size distribution, if it can be measured independently, into the ultrasonic model and then compare theory and experiment.

Given that Foldy's model assumes a low concentration of scatterers and weak scattering, there has been considerable effort to modify this model to be applicable to concentrated systems or systems where strong scattering is present. These efforts addressed the scattering function as a whole (Leroy, Strybulevych, Scanlon & Page, 2009) (Eq. (15)) or a part of it (Henyey, 1999) (Eq. (18)) so that energy losses due to the interactions between the scatterers are accounted for.

One of the modifications to address a drawback of Foldy's model was proposed by Henyey (1999), whose correction accounted for the radiative damping occurring in the effective medium instead of in the scatterer-free medium. Accordingly, Eq. (18) was modified to:

$$\delta^{radiative} = Rk \tag{24}$$

Leroy, Strybulevych et al. (2008) measured $v$ and $\alpha$ in monodisperse bubbly gels (gas volume fractions of 0.15 and 1%) by a transmission technique over a wide frequency range, and showed that $\alpha$ was well predicted whereas $v$ was overestimated at high frequencies by Foldy's model. Leroy, Strybulevych et al.'s work showed that the agreement between the experimental data and Henyey's model was worse compared to that of Foldy's model and that Foldy's model provides an “imperfect but satisfactory” characterization of $v$ and $\alpha$ as long as bubble concentration is below 1%. Notwithstanding the higher bubble concentrations, Foldy's model was used in preference to Henyey's for describing wave propagation in both nonyeasted (Koksel et al., 2014) and yeasted (Strybulevych et al., 2012) bread doughs.

For more concentrated systems (as high as >10% bubbles by volume), Leroy, Strybulevych et al. (2008) stated that when the resonance is strong enough that the interactions between bubbles become significant, $f(\omega,R)$ should be modified and a “collective” scattering function should be used. By stating so, they attempted to address one of the drawbacks of Foldy's model - that it does not take the interaction between scatterers into account. Leroy et al. (2009) studied ultrasound transmission through a model system with a single layer of bubbles in order to understand the effect of interaction between bubbles on the transmission of ultrasound. They showed both experimentally and theoretically that transmission through this model system was lowest at a frequency different than the resonance frequency of the individual bubbles. This means that a shift in $\omega_M$ to higher values was observed which was attributed to the interaction between bubbles. They reported an increase in $\omega_M$ by a factor of 1.7 for a bubble concentration of 10% and a larger increase in $\omega_M$ for a higher concentration of bubbles. These results suggest that a greater shift in $\omega_M$ is to be expected for bread doughs, and especially so during fermentation.

Another one of the drawbacks of Foldy's model is that it does not account for the possible interaction of the scatterer positions *i.e.*, how closely packed adjacent bubbles are. Leroy et al. (2011) addressed this issue by investigating wave propagation in a model system (a system made of polydisperse bubbles in an elastic matrix) with low-frequency resonances. They showed that Foldy's model was not a perfect fit for the results they obtained for $v$ and $\alpha$, and stated that the distance between adjacent bubbles affects the ultrasonic parameters. Leroy et al. (2011) proposed “a self-consistent approach” by considering how far or close adjacent bubbles are to each other. According to this new approach that they developed, the magnitude of the correction for the effect of the positions of the bubbles was found to be proportional to the bubble volume fraction. They concluded that their new approach characterized the experimental data better than Foldy's model.

### 2.2.3. High frequency regime

In the high frequency (short wavelength) regime, the ultrasonic frequencies are higher than the bubble resonance frequencies. Even though the attenuation coefficient is lower compared to that in the resonance region, it is substantially higher than that in the low frequency region (Scanlon & Page, 2015). Accordingly, for a randomly distributed scatterer population, the penetration depth of sound is short (Kytomaa, 1995). However, in this regime, the velocity and attenuation coefficient offer additional information on the matrix properties and the frequency dependence of the velocity and attenuation can be used to extract information about molecular relaxations occurring in the dough matrix (Fan et al., 2013). Ultrasound propagation in the high frequency regime is outside the scope of this review and therefore will not be discussed further here.

## 3. Research on bubble size distribution and mechanical properties of dough investigated by ultrasound

Studying bubbly systems using ultrasound may be challenging because of the practical difficulties associated with bubble-laden food systems, such as high attenuation (Povey, 1997). However, for wheat flour dough systems, it has been shown that the effect of both the bubbles and the properties of the dough matrix on the overall rheology of the dough can be interrogated when ultrasound of different frequencies is used (Leroy, Fan et al., 2008; Scanlon et al., 2008) since the ultrasonic frequencies can be divided into zones connected to different structural levels (Scanlon, 2013). At low frequencies (below approximately 500 kHz), dough properties are measured as a “composite” (Scanlon, 2013). Following this frequency regime (approximately from 500 kHz to 5 MHz), frequency-dependent peaks in phase velocity and attenuation coefficient are observed (Koksel et al., 2014; Leroy, Strybulevych et al., 2008; Scanlon, 2013; Scanlon et al., 2008; Strybulevych et al., 2012). Information from this region can be used to extract parameters defining the BSD in the dough (Leroy, Fan et al., 2008; Scanlon, 2013). At higher frequencies (approximately >5 MHz), velocity of ultrasound in the dough approaches the velocity in the bubble-free dough, *i.e.*, the dough matrix, and the attenuation coefficient is substantially smaller than it is near resonance (Leroy, Fan et al., 2008; Leroy et al., 2011). At frequencies greater than the resonance frequency of bubbles, the direct effects of bread ingredients, *e.g.*, enzymes and dough conditioners, as well as the mechanical properties of dough polymers, *e.g.*, wheat flour proteins, can be investigated (Fan et al., 2013).

The use of ultrasound as a tool to investigate properties of dough goes back as early as the 1970s. Reports that employed ultrasound include investigations of the mechanical properties of dough (Alava et al., 2007; Elmehdi et al., 2003; Elmehdi, Page, & Scanlon, 2004; García-Alvárez et al., 2006, 2011; Kidmose, Pedersen, & Nielsen, 2001; S. Lee, Pyrak-Nolte, & Campanella, 2004; Leroy et al., 2010; Ross et al., 2004), the BSD in dough (Koksel et al., 2014; Leroy, Fan et al., 2008;

Leroy et al., 2009; Scanlon, Page, Leroy, Fan, Elmehdi, Kiefte and Mehta, 2011; Strybulevych et al., 2012) and the effect of ingredients and processing conditions on dough properties (García-Alvárez et al., 2006; Gómez, Oliete, García-Alvárez, Ronda, & Salazar, 2008; Hatcher et al., 2014; Kidmose et al., 2001; H. O. Lee, Luna, & Daut, 1992; Létang et al., 2001; Mehta et al., 2009; Owolabi, Bassim, Page, & Scanlon, 2008; Rosell, Marco, García-Alvárez, & Salazar, 2011; Scanlon, Page, Leroy, Fan et al., 2011; Skaf, Nassar, Lefebvre, & Nongaillard, 2009). A summary of studies of low-intensity ultrasound evaluation of dough properties is presented in Table 1.

### 3.1. Ultrasound as a tool to investigate the mechanical properties of dough

The use of ultrasound as a tool to investigate the mechanical properties of dough can be grouped into two: (1) studies where longitudinal waves were used, (2) studies where shear waves were used. In the first group, both nonyeasted and fermenting doughs were studied, whereas in the second group only nonyeasted doughs were studied, possibly because of the difficulty measuring ultrasonic parameters owing to the very high attenuation of shear waves. According to Létang et al. (2001), the penetration depth of ultrasonic shear waves in bread dough systems is of the same order of magnitude as starch granule size at the frequency range they used (3–4.5 MHz).

Kidmose et al. (2001) investigated the viscoelastic properties of nonyeasted doughs made using flour from hard or soft wheat varieties, sugar, fat and water, using a low frequency (37 kHz) transmission ultrasonic technique. Ultrasonic measurements were performed as a function of time and the ultrasonic results were compared to the results obtained by shear oscillatory rheometry at much lower frequencies. As time progressed the signal velocity from ultrasonic measurements increased, as did $G'$. They reported that dough becomes firmer during aging due to the interactions of dough components after mixing and that the changes in dough structure could be probed by ultrasound. Kidmose et al. (2001) also investigated doughs made from different wheat flours at constant and variable water contents. When different types of flour were mixed at their optimum water absorption, the differences in consistency of doughs was eliminated and ultrasonic velocity, a function of bulk and shear moduli (Eq. (6)), was highly correlated to $G'$ from rheological parameters. When flours of different wheat varieties were mixed at constant water content, the flours from hard wheat varieties produced stiffer doughs (relatively greater bulk moduli) compared to those from soft wheat varieties. Since the bulk modulus of dough is much higher than its shear modulus, the longitudinal ultrasonic properties are dominated by variations in the bulk moduli. Accordingly, when flours of different wheat varieties were mixed at constant water content, not unexpectedly, no correlation between longitudinal ultrasonic parameters and shear rheological parameters ($G'$, $G''$) was found.

Elmehdi et al. (2004) also studied the mechanical properties of nonyeasted lean formula (hard wheat flour, salt, water) doughs at the low frequency regime. They manipulated the dough gas volume fraction (from 1% to 8%) by varying the mixer headspace pressure during mixing and investigated the mechanical properties of the doughs using a 50 kHz transmission ultrasonic technique. They showed that velocity decreased dramatically as gas volume fraction increased (Fig. 5a). They also reported that $\alpha$ increased linearly as the gas volume fraction in the dough increased (Fig. 5b). By evaluating the longitudinal modulus of the dough, determined from $\alpha$ and $v$, they demonstrated that the dough's mechanical properties were strongly affected by the presence of bubbles (Fig. 5c).

Another low frequency (100 kHz) ultrasonic transmission investigation was performed by García-Alvárez et al. (2006), who studied the viscoelastic properties of nonyeasted doughs made from wheat flours of different breadmaking potential and water, using a Brabender Farinograph mixer. Using the ratio of attenuation coefficient and signal velocity, flours with good and poor breadmaking potential were separated based on their ultrasonic properties, *i.e.*, flours with poor breadmaking potential had a tendency to have high attenuation and low velocity. However, the attenuation to velocity ratio of some of the flours with distinctive breadmaking potentials were similar, hence it was concluded that ultrasonic tests alone did not grant sufficient information to distinguish differences in flour quality. Following this study, Alava et al. (2007) manipulated the water content of doughs made from flours of different breadmaking potential (as assessed by differences in the resistance to stretching and extensibility of the dough) and tested these doughs with the same ultrasonic technique as García-Alvárez et al. (2006). The ultrasonic results were compared with the results obtained from the Extensograph (where water content was adjusted to optimal water absorption) and Alveograph (where a fixed water

**Table 1**
Summary of studies of low-intensity ultrasound evaluation of dough properties.

| Purpose | Variables | Dough type | Wave type[a] | Frequency | Authors |
|---|---|---|---|---|---|
| Dough rheological properties | – | Yeasted | L | 2–10 MHz | Lee et al. (2004) |
| | – | Nonyeasted | S | 2–4 MHz | Létang et al. (2001) |
| | – | Nonyeasted | S | 400 kHz | Leroy et al. (2010) |
| | Time after mixing, water concentration | Nonyeasted | L | 37 kHz | Kidmose et al. (2001) |
| | Gas volume fraction | Nonyeasted | L | 50 kHz | Elmehdi et al. (2004) |
| | Water concentration | Nonyeasted | L | 0.2–1.1 MHz | Lee et al. (1992) |
| | Vegetable shortening concentration | Nonyeasted | L | 50 kHz | Mehta et al. (2009) |
| Formulation optimization | Salt concentration | Nonyeasted | L | 1–5 MHz | Koksel et al. (2014) |
| | Vegetable shortening concentration | Nonyeasted | L | 50 kHz | Mehta et al. (2009) |
| | Flour types, yeast concentration | Yeasted | L | 2 kHz | Skaf et al. (2009) |
| | Surface-active bakery ingredients | Nonyeasted | L | 20 kHz–2 MHz | Scanlon, Page, Leroy, Fan et al. (2011) |
| Process optimization | Energy input (extrusion) | Nonyeasted | L | 50 kHz | Owolabi et al. (2008) |
| | Energy input (mixing) | Nonyeasted | L, S | 2–10 MHz, 2–4 MHz | Létang et al. (2001) |
| | Mixing time | Nonyeasted | L | 3–5 MHz | Ross et al. (2004) |
| Fermentation optimization | – | Yeasted | L | 50 kHz | Elmehdi et al. (2003) |
| | – | Yeasted | L | 2–10 MHz | Lee et al. (2004) |
| | Flour types, yeast concentration | Yeasted | L | 2 kHz | Skaf et al. (2009) |
| Microstructure optimization | – | Nonyeasted | L | 50 kHz–5 MHz | Leroy, Fan et al. (2008) |
| | – | Nonyeasted | L | 20 kHz–2 MHz | Scanlon, Page, Leroy, Fan et al. (2011) |
| | – | Nonyeasted and yeasted | L | 0.5–5 MHz | Strybulevych et al. (2012) |
| | Salt concentration | Nonyeasted | L | 1–5 MHz | Koksel et al. (2014) |
| Breadmaking potential of flours | – | Nonyeasted | L | 100 kHz | García-Alvárez et al. (2006) |
| | Water concentration | Nonyeasted | L | 100 kHz | Alava et al. (2007) |
| | Flour protein quality | Nonyeasted | L | 100 kHz | García-Alvárez et al. (2011) |

[a] L: Longitudinal waves, S: Shear waves.

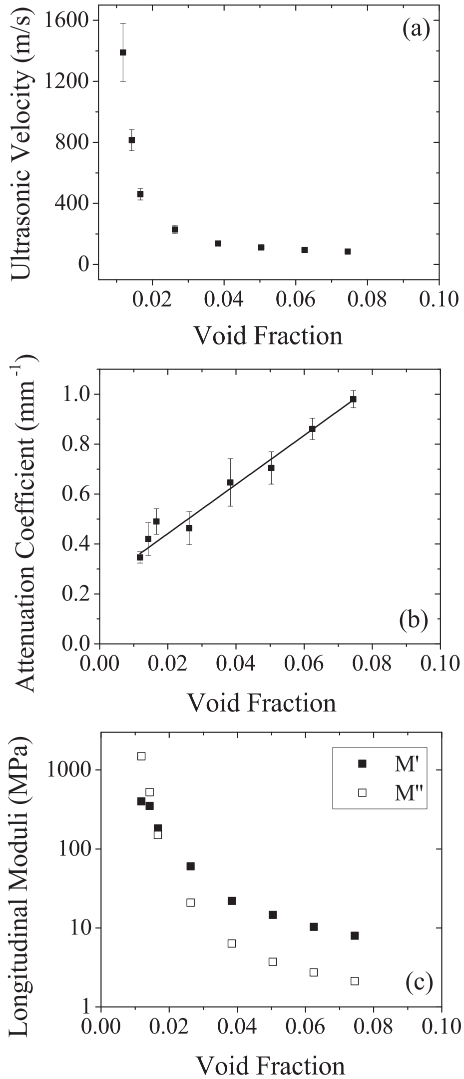


**Fig. 5.** (a) Ultrasonic velocity, (b) attenuation coefficient (and best linear fit to the data), and (c) longitudinal moduli of nonyeasted dough as a function of gas volume fraction (Reproduced, by permission of AACC International, from Elmehdi, H. M., Page, J. H., and Scanlon, M. G. 2004. Ultrasonic investigation of the effect of mixing under reduced pressure on the mechanical properties of bread dough. Cereal Chem. 81:504-510). Measurements were performed at 50 kHz.

content was used). Using the ratio of attenuation coefficient to signal velocity as an indicator of breadmaking quality, Alava et al. (2007) concluded that when there were variations in water content (as in doughs tested by the Extensograph), ultrasonic parameters could not clearly differentiate between flours of different strengths, while if the water content was kept constant (as in doughs tested in the Alveograph) ultrasound provided information on the strength of the flour. They acknowledged that further investigation was needed in order to employ ultrasound as a tool to discriminate the breadmaking potential of different flours. A similar low frequency evaluation of nonyeasted doughs made from flours of different breadmaking potential was conducted by García-Alvárez et al. (2011). They demonstrated that ultrasonic parameters were sensitive to changes in dough consistency caused by differences in flour protein quality. A high correlation between flour protein quality and velocity was reported. Moreover, they showed that velocity was sensitive to softening of the dough brought about by protease activity during flour storage.

Ross et al. (2004) investigated the effect of mixing time (under-, optimum- and over-mixing) on nonyeasted wheat flour dough systems prepared using various flour types (bread, all-purpose and cake flours) with differing protein contents. Empirical (Mixograph) and fundamental (dynamic oscillatory shear) rheology were employed as well as an ultrasonic transmission technique operating in the bubble resonance regime (3–5 MHz). They reported that both velocity and attenuation coefficient peaked at optimum mixing time for all the wheat flour dough systems they tested. Peaks in attenuation coefficient and velocity derived from ultrasound measurements as well as peaks in storage and loss moduli derived from shear rheology measurements at optimum mixing time were reported. These peaks were attributed to the development of highly aligned and fully hydrated glutenin polymers. The gas volume fraction of doughs, which they found to be constant as mixing time was manipulated, was not incorporated in a discussion of their results.

Elmehdi et al. (2003) used a low frequency (50 kHz) ultrasonic transmission technique to investigate the changes in dough structure and dough gas volume fraction during fermentation. Their dough samples, prepared using hard wheat flour, yeast, water and salt, were mechanically developed using a GRL-200 mixer. A significant change in ultrasonic velocity (Fig. 6a) was observed despite minute changes in density (Fig. 6b) at the beginning of fermentation. This substantial change in ultrasonic velocity was attributed to the changes in gluten structure and dough matrix elasticity caused by yeast activity. They concluded that the $CO_2$ concentration in the dough matrix increased with yeast activity, reducing the pH and affecting intermolecular interactions in gluten. To study the evolution of yeasted dough over longer times than could be investigated by Elmehdi et al. (2003), Skaf et al. (2009) developed a novel low frequency transducer that enabled ultrasonic measurements near 2 kHz. Although they did not report velocity and attenuation in their fermenting doughs, they did monitor significant changes in ultrasonic transit time and amplitude. During the initial stages of fermentation, their results can be taken as further evidence of the facultative respiratory capacity of yeast (Scanlon & Page, 2015). Lee et al. (2004) also investigated how dough mechanical properties were affected by fermentation using an ultrasonic transmission technique but at substantially higher frequencies (2–10 MHz). Doughs

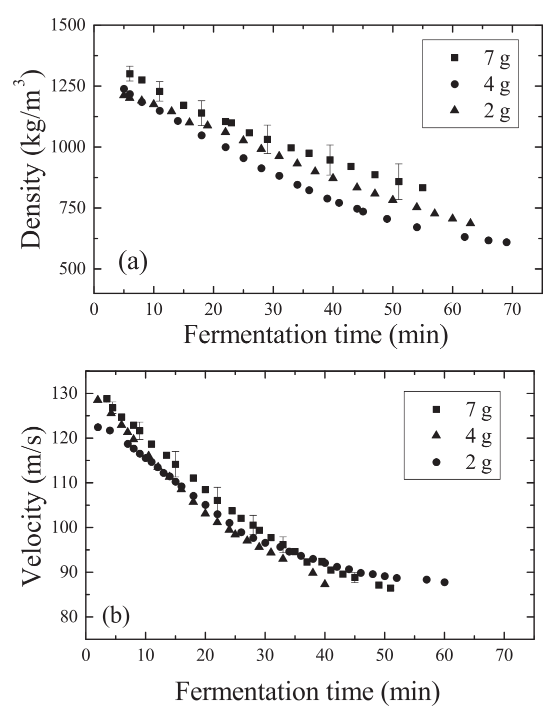


**Fig. 6.** (a) The change in density of fermenting doughs (dough masses of 2, 4 and 7 g) as a function of time. (b) The change in ultrasonic velocity measured for the fermenting dough samples in (a) (Reprinted from Food and Bioproducts Processing, Vol 81, Elmehdi, H.M., Page, J.H., Scanlon, M.G., Monitoring dough fermentation using acoustic waves, p. 217-223, 2003, with permission from Elsevier).

were prepared using all-purpose wheat flour, yeast and water, using a Mixograph mixer. From the ultrasonic velocity and attenuation coefficient, the longitudinal moduli of the dough were calculated. According to Lee et al. (2004), the changes in the longitudinal moduli of the dough with longer fermentation times were due to a decrease in the dough matrix elasticity. Even though the changes in the dough's bubbly structure during fermentation (and how these changes would affect the longitudinal moduli) were not discussed, a decrease in dough elasticity was confirmed by conventional extensional rheology tests.

Leroy et al. (2010) performed ultrasonic shear wave measurements on mechanically developed lean formula (hard wheat flour, water, salt) nonyeasted wheat flour doughs in order to determine the complex shear modulus of the dough. By combining their findings with the results obtained from testing subsamples of the same dough with small strain shear rheometry, they were able to characterize dough's shear modulus over a frequency range of more than eight decades (Fig. 7). They reported that their nonyeasted wheat flour dough could be characterized as a soft viscoelastic solid ($G' > G''$) and that the frequency dependence of shear moduli followed a low-exponent power law model. At higher frequencies (2–4 MHz) than those used by Leroy et al. (2010), little frequency dependence of shear wave properties was also reported for nonyeasted doughs (Létang et al., 2001).

### *3.2. Ultrasound as a tool to investigate bubble size distribution (BSD) in dough*

It has been demonstrated that bubbles play a very important role in ultrasonic experiments; however, experiments are often limited to frequencies below or above the resonance frequency of bubbles and for materials with low concentrations of bubbles. This limitation is due to the very high attenuation of sound in bubbly media around the resonance frequency of bubbles, which makes ultrasonic measurements challenging (Leroy et al., 2009).

Leroy, Fan, et al. (2008) investigated the BSD in nonyeasted dough by using an ultrasonic transmission technique (50 kHz–5 MHz) and employing Foldy's model to describe wave propagation in two well-characterized bubbly gels. By using Foldy's model, they estimated the BSD from the ultrasonic results on the assumption that the distribution was lognormal. According to their results, for a nonyeasted wheat flour

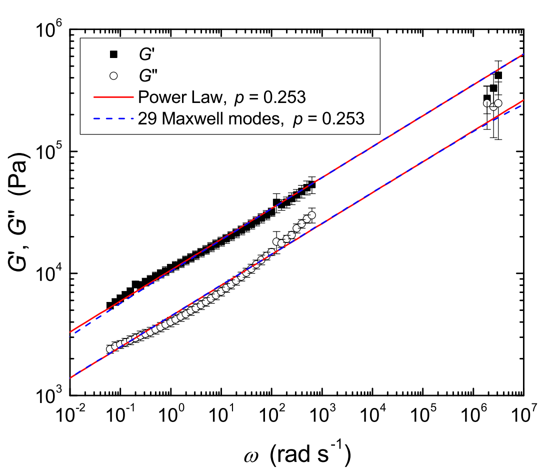


**Fig. 7.** Complex shear modulus of a strong wheat flour dough determined by rheometry and ultrasound (Reprinted from Journal of Non-Newtonian Fluid Mechanics, Vol 165, Leroy, V., Pitura, K.M., Scanlon, M.G., Page, J.H., The complex shear modulus of dough over a wide frequency range, p. 475-478, 2010, with permission from Elsevier). Solid and open symbols represent the real and imaginary parts of the complex shear modulus, respectively. Solid red lines and dashed blue lines are fits that represent power-law behavior and a spectrum of discrete Maxwell relaxation modes, respectively.

dough (hard wheat flour, water and salt), an increase in the median bubble radius (from 14 μm at 53 min after mixing to 18 μm at 96 min after mixing) and a narrowing of the BSD ($\varepsilon = 0.46$ to $\varepsilon = 0.44$) were observed (Fig. 8). They concluded that these time-dependent changes were due to the effects of disproportionation occurring in the dough. Disproportionation is a mechanism resulting in an increase in the median bubble size with time due to the higher Laplace pressures in smaller bubbles compared to those in larger bubbles (Kokelaar et al., 1996; Murray & Ettelaie, 2004; Shimiya & Nakamura, 1997; Shimiya & Yano, 1988).

Around the bubble resonance frequencies, Scanlon, Page, Leroy, Elmehdi et al. (2011) studied the dynamics of bubbles in mechanically developed nonyeasted doughs (hard wheat flour, water and salt) using an ultrasonic transmission technique. Around these frequencies, ultrasonic velocity and attenuation coefficient were substantially affected by the changes in the bubble sizes as a result of disproportionation. A decrease in the peak in $\alpha$ with time as well as a shift in the peak frequency to lower values were observed, consistent with changes in bubble sizes associated with disproportionation. The same trend was observed when mechanically developed nonyeasted doughs with different salt concentrations (0.8–2.4% salt on flour weight basis) were monitored 3 to 5 h after mixing (Koksel et al., 2014). In Fig. 9, the time evolution of typical bubble radius (40–50% increase in bubble radii from 3 to 5 h) in these doughs is presented.

Strybulevych et al. (2012) used an ultrasonic reflection technique to examine the dynamics of bubbles in both nonyeasted and yeasted doughs as a function of time. For mechanically developed nonyeasted doughs made from hard wheat flour, water and salt, both the width and the normalized mean bubble size of the lognormal BSD increased with time, owing to disproportionation. For yeasted doughs, prepared using a sponge and dough procedure, at the initial stage of the fermentation process, the normalized mean bubble size decreased which was attributed to loss of oxygen due to its consumption by yeast in the dough matrix. After this initial period, the normalized mean bubble size increased which was due to the growth of bubbles as $CO_2$ diffused into them as a result of the activity of the yeast.

### *3.3. Ultrasound as a tool to evaluate dough ingredients and dough processing conditions*

Production of consistent and high quality baked goods depends on the manipulation and control of dough ingredients and dough processing conditions (Campbell & Martin, 2012). Accordingly, optimization of dough ingredients and dough processing conditions has been the focus of many studies. Since conventional techniques for dough testing can take relatively long times and do not offer fundamental rheological information, and since ultrasonic techniques are relatively less expensive (Awad et al., 2012) and provide non-destructive and rapid results (Elfawakhry, Hussein, & Becker, 2013; García-Álvarez et al., 2006), the use of ultrasonic techniques for the measurement of dough properties as affected by ingredients and processing conditions is increasing.

Lee et al. (1992) studied the rheological properties of nonyeasted dough, made from all-purpose wheat flour and water, with an ultrasonic transmission technique (0.2–1.1 MHz). They showed that both the $v$ and $\alpha$ were affected by dough moisture content. They reported that maximum $\alpha$ and minimum $v$ were obtained for doughs with the lowest moisture content. Owolabi et al. (2008) investigated the physical properties of extruded nonyeasted doughs as affected by water content using an ultrasonic transmission technique (50 kHz). They reported that above a certain specific mechanical energy input (for developing the dough in the extruder), as the water content decreased, velocity increased (Fig. 10a). The results of Létang et al. (2001) supported the finding that acoustic properties of doughs were sensitive to energy input (Fig. 10b) during dough development, pointing to the ability of ultrasonic techniques to discriminate wheat flour-water systems based on differences in how they were processed.

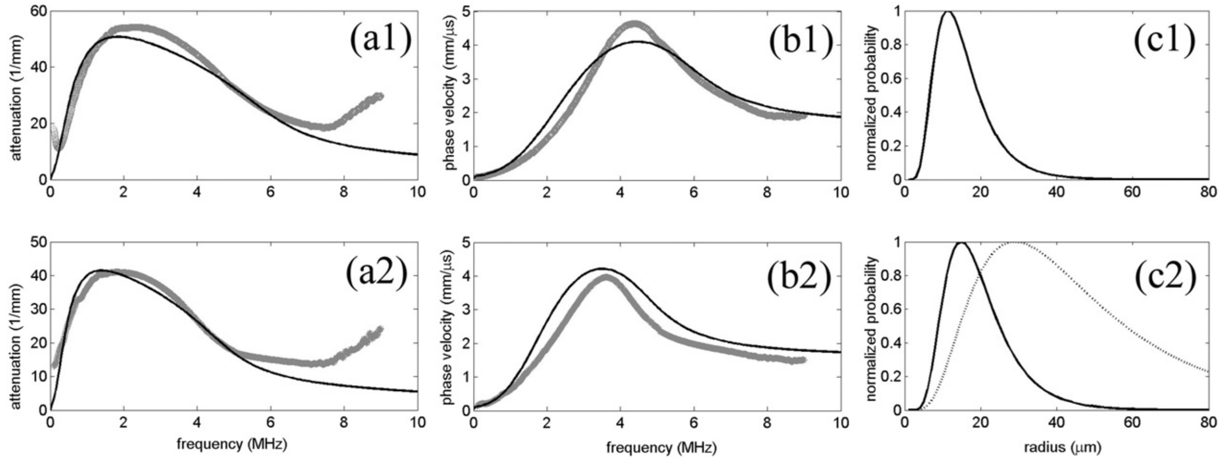


**Fig. 8.** Experimental results (gray symbols) and theory predictions (the solid black lines) for the frequency dependence of attenuation coefficient (a), phase velocity (b) and the bubble size distribution extracted using Foldy's model (c) obtained from a nonyeasted dough subsample analyzed 53 min after mixing (1) and 96 min after mixing (2) (Reproduced, by permission of AACC International, from Leroy, V., Fan, Y., Stybulevych, A. L., Bellido, G. G., Page, J. H., and Scanlon, M. G. 2008. Investigating the bubble size distribution in dough using ultrasound. Pages 51-60 in: Bubbles in Food 2: Novelty, Health and Luxury. G. M. Campbell, M. G. Scanlon, and D. L. Pyle, eds. AACC International, St. Paul, MN). The light gray curve in (c2) is the bubble size distribution in a nonyeasted dough sample measured by X-ray microtomography, 90 min after the end of mixing (Bellido et al., 2006).

In addition to water, the effects of different bakery ingredients on ultrasonic parameters have been investigated. Mehta et al. (2009) investigated the effect of vegetable shortening on dough mechanical properties using an ultrasonic transmission technique (50 kHz). Doughs were prepared from hard wheat flour, water, salt and vegetable shortening (2% to 8% on flour weight basis). They concluded that ultrasound was able to probe the changes in the dough matrix arising from the modification of gluten polymers brought about by shortening. Scanlon, Page, Leroy, Fan et al. (2011) studied the effect of surface-active bakery ingredients on nonyeasted wheat flour dough properties at low frequencies and showed that low frequency ultrasound was a sensitive probe of concentration changes in distilled monoglycerides (up to 2% on flour weight basis) and shortening (up to 8% on flour weight basis), but these two surface-active ingredients had contrasting effects on dough properties as assessed by ultrasound. Skaf et al. (2009) monitored the evolution of bread dough (flour, water, salt and yeast) during a one hour fermentation period using an ultrasonic transmission technique (<50 kHz) and successfully followed the changes in dough properties during fermentation as affected by different flour types and variation in yeast content. Rosell et al. (2011) studied the rheological

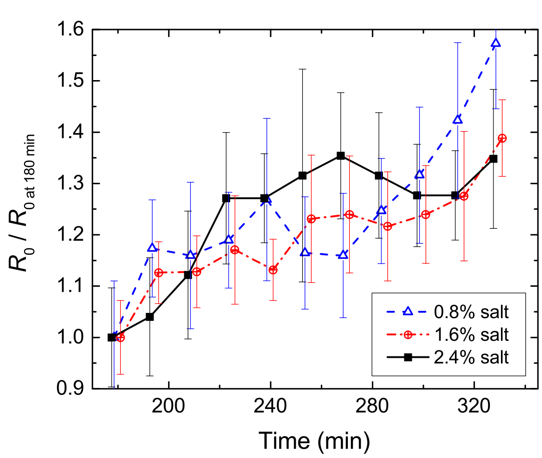


**Fig. 9.** Time-dependent changes in the normalized bubble radius in nonyeasted bread doughs as a function of salt concentration (Reproduced, by permission of AACC International, from Koksel, F., Strybulevych, A., Page, J. H., Scanlon, M. G., 2014. Ultrasonic characterization of unyeasted bread dough of different sodium chloride concentrations. Cereal Chem. 91:327-332).

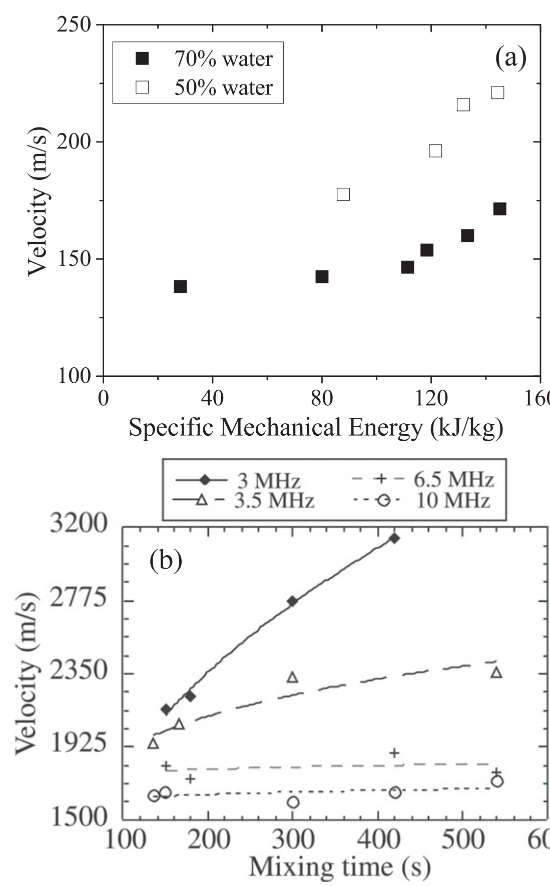


**Fig. 10.** (a) Ultrasonic longitudinal velocity (50 kHz) as a function of specific mechanical energy for nonyeasted extruded doughs made with 50% (open squares) and 70% (solid squares) water content (Reprinted from Journal of Food Engineering, Vol 86, Owolabi, G.M., Bassim, M.N., Page, J.H., Scanlon, M.G., The influence of specific mechanical energy on the ultrasonic characteristics of extruded dough, p. 202-206, 2008, with permission from Elsevier), (b) ultrasonic longitudinal velocity as a function of mixing time for a nonyeasted dough containing 52% water (Reprinted from Ultrasonics, Vol 39, Létang, C., Piau, M., Verdier,C., Lefebvre, L., Characterization of wheat-flour–water doughs: a new method using ultrasound, p.133-141, 2001, with permission from Elsevier).

properties of composite (rice-soybean) gluten-free flour doughs with the addition of a cross-linking enzyme (transglutaminase) using ultrasound. They demonstrated that as the ratio of soybean to rice flour increased, the consistency of the dough increased and this was reflected in an increase in velocity. For doughs with a higher concentration of the cross-linking enzyme, an increase in $v$ and a decrease in $\alpha$ were observed, once again, pointing to the sensitivity of ultrasound to changes in dough properties brought about by ingredients (Rosell et al., 2011). Ultrasound's sensitivity to changes in dough properties as a result of manipulating dough formulation and/or breadmaking process indicate its great potential for identifying the structural changes occurring in dough during the breadmaking process.

## 4. Conclusions

This overview of ultrasound as a research tool to investigate the bubble size distribution in dough and dough mechanical properties has shown that the cereal food industry can significantly benefit from a good understanding of ultrasonic techniques and advances in their applications. With tuning of the frequency of ultrasound, identifying differences in the breadmaking potential of different wheat varieties, determining the effect of dough ingredients on dough properties, quantifying bubbles at the end of dough mixing and characterizing how they evolve afterwards, are entirely possible. There is no doubt that the use of ultrasound offline as a tool to investigate the structure and physicochemical properties of cereal based foods, and online as a means of controlling the quality of intermediate and final products during manufacturing, will continue to stimulate further research in the near future.

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