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Ultrasonic Characterization of Foods and Drinks: Principles, Methods, and Applications

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ABSTRACT: The use of ultrasonics as an analytical technique for providing information about the physicochemical properties of foods is becoming increasingly popular in the food industry. Ultrasound can be used to rapidly and nondestructively measure properties such as food composition, structure, flow rate, physical state, and molecular properties. As well as being used as an analytical instrument in the laboratory, ultrasound can also be used for continuous monitoring of food properties on-line during processing. This article critically reviews the current status of ultrasound in the food industry, presents the basic principles of ultrasonic testing of foods, and highlights areas where ultrasound will prove to be most useful in the future.

KEY WORDS: ultrasound, acoustics, analysis, on-line, physical properties.

I. INTRODUCTION

The manufacture of food products is a highly dynamic and intensely competitive industry. To succeed, food manufacturers must continue to provide safe, nutritious, and desirable foods, as well as being able to rapidly respond to changing preferences of consumers for cheaper, healthier, more convenient, and exotic foods. To do this they must improve the quality of existing products, develop new products, reduce waste, and lower manufacturing costs. There are a number of ways these objectives can be achieved. Two of particular importance have recently been highlighted:

1. Development of on-line sensors for monitoring the properties of foods during production and storage^{1,2}
2. An improved understanding of the relationship between the bulk physicochemical properties of foods and their molecular/structural properties³

It is widely recognized that there is a lack of suitable sensors for providing information about the physicochemical properties of foods, espe-

cially for the continuous monitoring of foods during processing.¹ This is holding back the implementation of a number of advanced process control strategies that would benefit the food manufacturing industry (e.g., model-based, fuzzy logic, and artificial neural networks).² One of the major problems in developing analytical techniques for use in the food industry is the diversity and complexity (both compositional and structural) of foods. Many traditional "wet-chemistry" techniques have limited application because they are destructive, time consuming, and labor intensive. Consequently, there has been a drive to develop new technologies, or to apply techniques currently used in other areas, for the analysis of foods.^{4,5}

There have been major advances in the application of Fourier transform infrared and nuclear magnetic resonance (NMR) techniques for rapid and nondestructive characterization of foods.⁶⁻⁸ NMR can provide information about the concentration, molecular structure, diffusion, particle size, and physical state of foods.⁹ Once calibrated, infrared can be used to simultaneously determine the concentration of a number of major components in foods, including moisture, protein, lipids,

and carbohydrates.¹⁰ Nevertheless, both technologies are still fairly expensive and have limited application to certain food materials.

Over the last decade or so there has been increasing interest in the use of ultrasound for characterizing food materials,^{11–15} and a wide range of applications have already been developed (Table 1). In this technique, a high-frequency sound wave is propagated through the material being tested. Information about the properties of a material are then obtained by measuring the type and degree of interaction between the sound wave and the material. Ultrasound has major advantages over many other analytical methods because it is nondestructive, rapid, precise, relatively inexpensive, and can be applied to concen-

trated and optically opaque foods. The author therefore expects that ultrasound will find increasing use in the food industry, as both a basic research tool and as an on-line sensor.

This article critically assesses current and potential applications of ultrasound in the food industry, highlighting its advantages and disadvantages over alternative technologies. An appreciation of the value of ultrasound as an analytical technique relies on some understanding of its operation. For this reason, this article reviews the basic principles of ultrasonic propagation in materials, experimental techniques for carrying out ultrasonic measurements, methods of data interpretation, and practical applications of the technique in the food industry.

TABLE 1
Examples of the Use of Ultrasound for Characterizing the Physicochemical Properties of Food Materials

Application	Food material	Property measured	Ref.
Composition	Sugar concentration of aqueous solution	c	47, 48, 59–65
	Salt concentration of brine	c	49, 50
	Triglycerides in oils	c, α	66, 67, 85–93
	Droplet concentration of emulsions	c, α	13, 23, 37, 57, 95–100
	Alcohol content of beverages	c	62–64
	Air bubbles in aerated foods	c, α , Z	103–105
	Composition of milk	c, α	134–135
	Ratio of fat to lean in meats	c, α	71, 106–120
Phase transitions	Biopolymer concentration in gels	c, α	42–44
	Milk fat globules	c, α	
	Triglycerides in fatty foods	c, α	71–78
	Water in meat	c, α	116
Particle size	Oil droplets in salad cream	c, α	37, 101
	Milk fat globules	c, α	57
	Casein micelles	c, α	56
Miscellaneous	Air bubbles in aerated foods	c, α , Z	103–105
	Quality of eggs	c, α	154–157
	Ripeness and quality of fruits	c, α , Z	130–133
	Texture of biscuits	c, α	174, 175
	Gelation of gels	c, α	180
	Cracks in cheese	c, α , Z	148, 149
	Flow rate of liquids in pipes	t	179
	Level of liquids in tanks	t	179
	Detection of extraneous matter	t, Z	179
	Monitoring enzymatic reactions	c, α	43
	Molecular interactions of solutes	c, α	43
	Structure of biopolymers	c, α	40–44, 51–54
	Creaming profiles of emulsion droplets	c, α	13, 100
	Temperature of foods	t	179
	Imaging of microbial growth	t, Z	141–143

To avoid possible confusion, it is helpful to point out that there are two distinctly different types of application of ultrasound in the food industry: *high-* and *low-*intensity applications. This review is only concerned with the use of low-intensity ultrasound as an analytical technique for providing information about the physical and chemical properties of food materials. The power levels used are so low (typically less than 1 W/cm²) that they cause no alteration in the physical or chemical properties of a food once the ultrasonic wave is removed (i.e., they are nondestructive).¹⁶ On the other hand, high-intensity applications use power levels that are so high (typically between 10 and 1000 W/cm²) that the properties of a food material are changed, often permanently. High-intensity ultrasound is used in the food industry as a means of altering the physical and chemical properties of foods^{17,18} (e.g., sonochemistry, homogenization, cleaning, cell disruption, sterilization, crystal modification, enzyme inhibition and tenderization of meat). High-intensity applications have been reviewed by Roberts.¹⁸

II. BASIC PRINCIPLES

A. Description of an Ultrasonic Wave

To understand how ultrasound can be used to provide information about the physicochemical properties of foods one must be reasonably familiar with the basic principles of ultrasonic propagation in materials. Consider a material to consist of a series of imaginary layers that are in contact with one another (Figure 1A). When an oscillating stress wave is applied to the surface, it acts across the length of the material because of the restoring forces between the layers. In the presence of a stress wave, the layers oscillate around their equilibrium positions at the same frequency as the wave. Once the wave is removed, the energy stored as ultrasound is dissipated, and the layers return back to their equilibrium positions, that is, there is no net movement of the layers (Figure 1A). This is an important attribute of ultrasound because it means that the technique is nondestructive.

Ultrasonic waves can be applied to materials in a variety of ways. When an oscillating stress wave is applied perpendicular to the surface of a material, the layers move in the same direction as the propagating wave, and a *compression wave* is generated (Figure 1B). Conversely, when the oscillating force is applied parallel to the surface of the material, the movement of the layers is perpendicular to the direction of the propagating wave and a *shear wave* is generated (Figure 1C). Compressional and shear waves are the most commonly used in the food industry. Even so, there are other types of ultrasonic wave that may prove more useful for certain applications (e.g., surface waves).¹⁹

The propagation of an ultrasonic wave through a material is represented graphically by Figure 2, which shows the variation of the amplitude of the wave with time and distance travelled. The amplitude can be expressed in terms of a number of different physical properties of the material that vary in the presence of an ultrasonic wave, such as the displacement of the layers from their equilibrium position, the velocity or acceleration of the layers, or the local energy, pressure, density, or temperature within the material.¹⁹ An ultrasonic wave is characterized by its amplitude (A) and frequency (f), which are chosen by the investigator, and its wavelength (λ) and attenuation coefficient (α), which are (frequency-dependent) characteristics of a material. The attenuation coefficient is a measure of how rapidly the amplitude of a wave decreases as it travels through a material: the higher α , the more rapid the reduction in amplitude. The ultrasonic velocity (c) is related to the wavelength and frequency ($c = \lambda f$), and so it is also a characteristic property of a material. Measurements of the ultrasonic velocity and attenuation coefficient are the basis of most ultrasonic techniques used to evaluate the properties of foods.

B. Ultrasonic Properties of Materials

Materials are normally characterized in terms of fundamental physical properties, such as density and elastic moduli. They may also be characterized in terms of their ultrasonic properties:

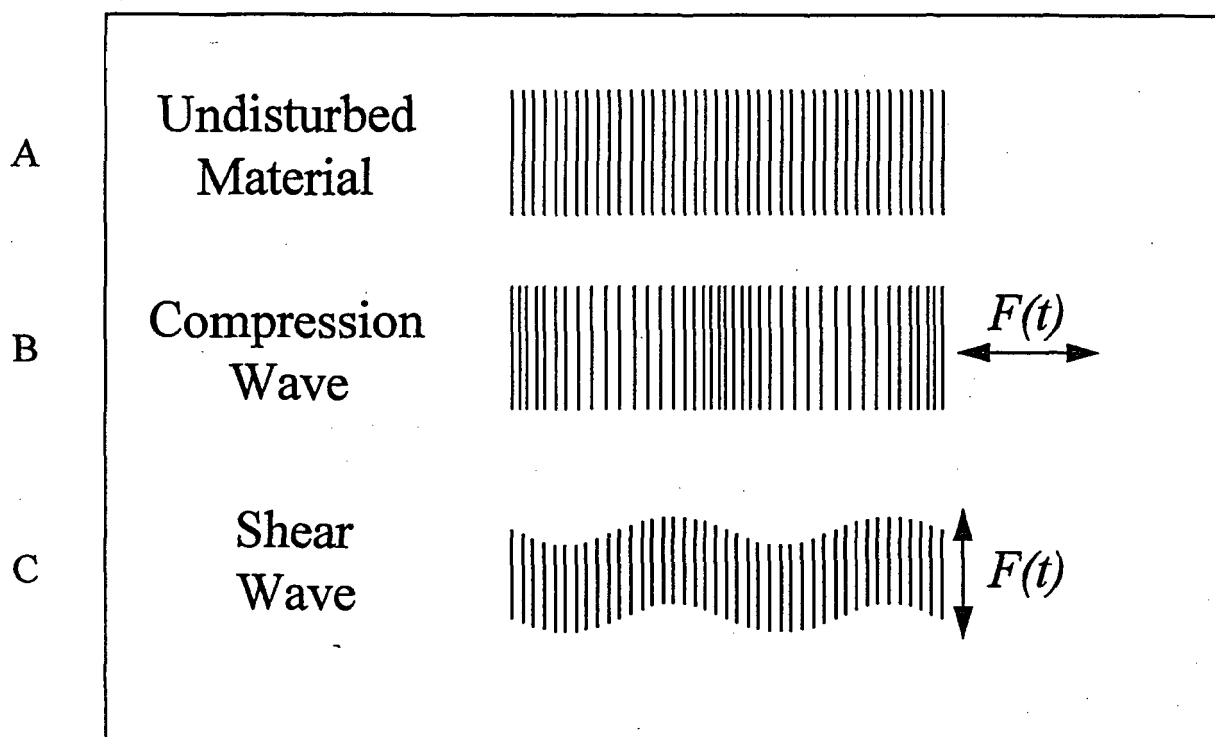


FIGURE 1. Ultrasonic waves can propagate through materials as either compression or shear waves, depending on the way the acoustic pressure $F(t)$ is applied to the surface of the material.

ultrasonic velocity, attenuation coefficient, and acoustic impedance. In this section, the meaning of these ultrasonic properties is explained, and their relationship to the fundamental physical properties of food materials is discussed.

The relationship between the ultrasonic properties of a material and its physical properties has been derived by a mathematical analysis of the propagation of an ultrasonic wave through a material.^{20,21}

$$\left(\frac{k}{\omega}\right)^2 = \frac{\rho}{E} \quad (1)$$

Here, k is the complex wave number of the material ($=\omega/c+i\alpha$), ω is the angular frequency ($=2\pi f$), E is the elastic modulus, ρ is the density, and $i = \sqrt{-1}$. This simple equation is of central importance for the interpretation of ultrasonic measurements. It clearly illustrates the fundamental relationship between the measurable ultrasonic properties (c and α) and the bulk physical proper-

ties (E and ρ) of a material. However, it should be stressed that the elastic modulus and density measured in an ultrasonic experiment are complex and frequency-dependent properties, and may have values that are different from the same quantities measured in a static experiment. This is particularly true for materials that strongly absorb or scatter ultrasound (e.g., bubbly liquids, foams, and some crystallizing systems). For materials where the attenuation is not large (i.e., $\alpha \ll \omega/c$), the difference between the static and dynamic values is negligible and can usually be ignored. This is true for many homogeneous materials encountered in the food industry (e.g., water, oils, and aqueous solutions).

1. Ultrasonic Velocity

Most ultrasonic instruments currently used in the food industry are based on measurements of the ultrasonic velocity (i.e., the distance traveled

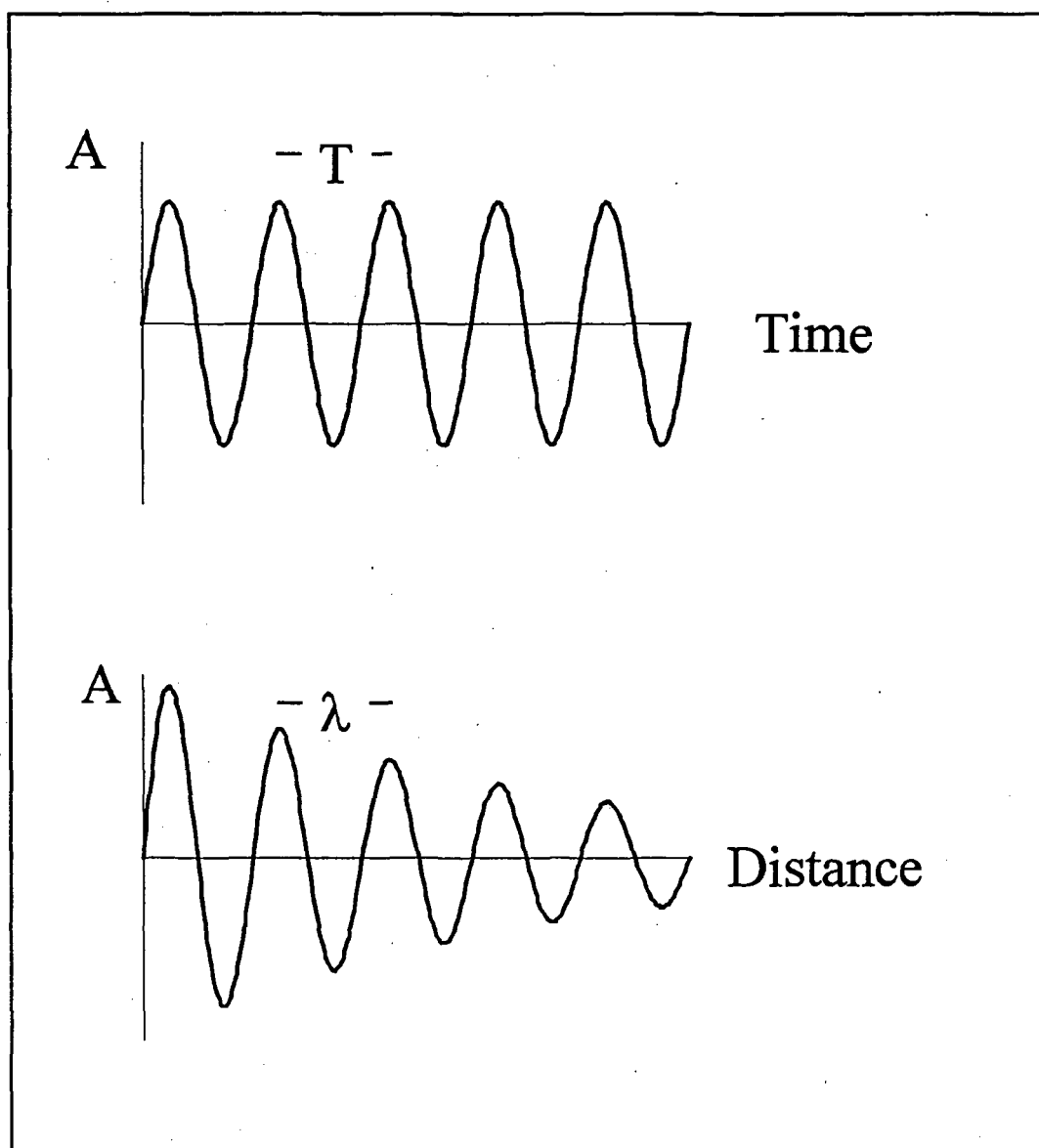


FIGURE 2. The propagation of an ultrasonic wave in a material can be represented by plotting the variation of the amplitude of the displacement (A) of a layer from its equilibrium position as a function of time or distance. Here, T is the period ($=1/f$) and λ is the wavelength of the wave.

by an ultrasonic wave in unit time). It is normally determined in one of two ways: either the wavelength of ultrasound is measured at a known frequency ($c = \lambda f$) or the time t taken for a wave to travel a known distance d is measured ($c = d/t$). Each of these methods is described in some detail in Section III.

The velocity at which an ultrasonic wave propagates through a material depends on its physi-

cal properties. For low-attenuating materials (i.e., $\alpha \ll \omega/c$), Equation 1 can be simplified:

$$\frac{1}{c^2} = \frac{\rho}{E} \quad (2)$$

The ultrasonic velocity of a material is therefore simply related to two of its most basic physical properties: the elastic modulus and density.

The less dense a material or the more resistant it is to deformation, the faster an ultrasonic wave propagates. Usually, differences in the moduli of materials are greater than those in density, and so the ultrasonic velocity is determined more by the elastic moduli than by the density. This explains why the ultrasonic velocity in solids is greater than that in fluids, even though fluids are less dense (Table 2).

The modulus used in Equation 2 depends on the physical state and dimensions of the material being tested, as well as the type of ultrasonic wave used (compressional or shear). For bulk solids, the appropriate modulus is $K + 4G/3$, where K is the bulk modulus and G is the shear modulus. For liquids and gases, the appropriate modulus is the bulk modulus, which is the reciprocal of the adiabatic compressibility, κ . Shear waves can be propagated through solids because of the relatively strong and permanent bonds between the molecules ($E = G$), but they are highly attenuated in fluids because the molecules easily slip past one another. Thus, shear waves are commonly used to test the properties of solids, but are rarely used to analyze liquids and gases.

For viscoelastic materials, the moduli are complex and frequency dependent, for example, $E = E' + iE''$, where E is the storage modulus and E'' is the loss modulus. In this respect, ultrasonic characterization of foods is similar to dynamic rheological testing. Both techniques utilize oscillating compressional or shear waves that cause small deformations in a material. Nevertheless, there are also important differences. First, the frequencies used in ultrasonic testing (typically 0.1 to 100 MHz) are orders of magnitude higher than those used in dynamic rheological tests (typically <1 kHz), which means that the two techniques are sensitive to relaxation mechanisms that occur on greatly different time scales. Second, rheological tests directly measure the relationship between the applied stress and the resulting strain (or vice versa), whereas ultrasonic techniques measure the effect that the rheological properties have on the velocity or attenuation of an ultrasonic wave passing through the material.

The ultrasonic properties of a number of common food and nonfood materials are compiled in Table 2. The velocity of ultrasound in solids is

usually greater than that in liquids, which is in turn greater than that in gases. This means that ultrasonic velocity measurements can be used to monitor phase transitions (e.g., crystallization or melting), where the material transforms from a solid to a liquid, or vice versa. The fact that different components in a food have different ultrasonic velocities means that ultrasound can also be used to determine the composition of foods or to identify the material being tested (see later).

2. Attenuation Coefficient

As an ultrasonic wave travels through a material, its amplitude decreases due to attenuation (Figure 2B). All materials attenuate ultrasound to some extent, the major sources of attenuation being adsorption and scattering. Adsorption is important in both homogeneous and heterogeneous materials and is caused by molecular processes that convert some of the energy stored as ultrasound into other forms, and ultimately into heat.²² Scattering is only important in heterogeneous materials, such as emulsions, suspensions, and foams, and occurs when an ultrasonic wave is incident on a discontinuity (e.g., a particle) and is scattered in directions that are different from that of the incident wave.²³ Unlike absorption, the energy is still stored as ultrasound, but the direction of propagation is altered so that the wave is not detected by a receiver in the forward direction. Nevertheless, there are often absorption mechanisms associated with scattering in heterogeneous materials, and these can dominate the overall attenuation (e.g., thermal or viscous loss mechanisms).²³ Measurements of the absorption and scattering of ultrasound can be used to provide valuable information about the physicochemical properties of food materials (see later).

The attenuation coefficient of a material has units of Nepers per meter (Np/m) when defined by the following equation:

$$A = A_0 e^{-\alpha x} \quad (3)$$

Here A_0 is the initial amplitude of the wave and x is the distance it has traveled through the material.

TABLE 2
Ultrasonic Properties of Selected Food and Nonfood Materials. All
measurements are at 20°C unless stated otherwise

Material	Frequency (MHz)	Velocity (m/s)	Impedance ($\times 10^8$ kg m/s)	Attenuation (Np/m)
Gases				
Air	0.4	330	4.3×10^{-4}	0.14
Hydrogen	1	1300		0.01
Nitrogen	1	330		0.24
Oxygen	1	310		0.17
Aqueous Solutions				
Distilled water	5	1482.3	1.48	0.7
Water + 10% NaCl				
Water + 10% glucose	2.25	1508.8	1.53	
Water + 10% fructose	2.25	1519.8	1.55	
Water + 10% sucrose	2.25	1506.5	1.53	
Water + 10% protein				
Ice (-20°C)		3840	3.5	
Fats and Oils				
Corn oil	1.25	1469.5	1.35	
Grapeseed oil	1.25	1470.7	1.35	
Groundnut oil	1.25	1465.9	1.35	
Olive oil	1.25	1465	1.35	
Palm oil	1.25	1459.3	1.35	
Rapeseed oil	1.25	1469.4	1.35	
Safflower oil	1.25	1471.4	1.35	
Soybean oil	1.25	1469.8	1.35	
Sunflower oil	1.25	1471.6	1.35	
Solid animal fat (31°C)		2000–2070	2.0–2.1	
Miscellaneous Solids				
Glass	20	5660	12	2
Quartz	1	5700	15	
Perspex (lucite)	1	2700	32	
Aluminum	1	6400	17	
Brass	1	3500	3	
Copper	1	4700	42	
Steel	1	6000	47	
Fruits and Vegetables				
Potato	0.5	700–850	0.7–0.85	
Tomato pericarp	0.5	1300	1.3	
Tomato placenta	0.5	900–1000	0.9–1.0	
Orange rind	0.5	1320–1360		
Lemon rind	0.5	900–1600		

TABLE 2 (continued)
Ultrasonic Properties of Selected Food and Nonfood Materials. All
Measurements Are At 20°C Unless Stated Otherwise

Material	Frequency (MHz)	Velocity (m/s)	Impedance ($\times 10^6$ kg m/s)	Attenuation (Np/m)
Aerated Foods				
Unyeasted bread dough	0.5	114	0.14	
Proving bread dough (39°C)	0.5	≈ 250	0.1–0.3	3.5
Proving pastry	0.5	500–700		
Aerated chocolate (25°C)	0.5	900–1000	0.9–1	
Meat and Fish Products				
Distilled water (39°C)	5	1528	1.53	
Blood (39°C)	5	1534	1.62	
Bone (39°C)		4000	3.75–7.38	
Fat (39°C)	5	1440	1.35	
Liver (39°C)		1590	1.64–1.68	
Muscle (39°C)		1590	1.65–1.74	
Homogenized milk	1		1.5	
Skim milk (28°C)	1	1522	1.5	23
Cheese (10°C)	0.05–0.93	1365–1645	1.3–1.6	
White fish flesh	1	1550	1.6	7

The attenuation coefficient is determined by measuring the amplitude of an ultrasonic wave as a function of the distance it has traveled through a material, and fitting the measurements to the above equation. The attenuation coefficient is sometimes reported in units of decibels per meter (dB/m), where $1 \text{ Np} = 8.686 \text{ dB}$.

3. Acoustic Impedance

The acoustic impedance of a material is important because it determines the fraction of an ultrasonic wave that is reflected from its surface. The *specific acoustic impedance* (Z) is defined as the ratio of the acoustic excess pressure (P) and the particle velocity (U):

$$Z = \frac{P}{U} = \frac{\omega p}{k} \quad (4)$$

In general, Z is complex and can be divided into a real and imaginary part: $Z = R_Z + iX_Z$, where R_Z is the *resistive* component and X_Z is the *reactive*

component. For materials where the attenuation of ultrasound is small (i.e., $\alpha \ll \omega/c$) the imaginary part can be ignored, so that $Z = R_Z = \rho c$, which is called the *characteristic impedance*. The acoustic impedance of a material is usually determined by measuring the fraction of an ultrasonic wave that is reflected from its surface (see Section II.C.1).

C. Interaction of Ultrasound with Matter

1. Reflection from Surfaces

When an ultrasonic wave encounters a boundary between two different materials it is partly reflected and partly transmitted (Figure 3). The fraction of ultrasound reflected or transmitted depends on the acoustic impedances of the two materials. The reflection of ultrasonic waves from surfaces is analogous to that of light waves, but in optics it is differences in refractive indices, rather than acoustic impedances, that determine the fraction of ultrasound reflected or transmitted. The

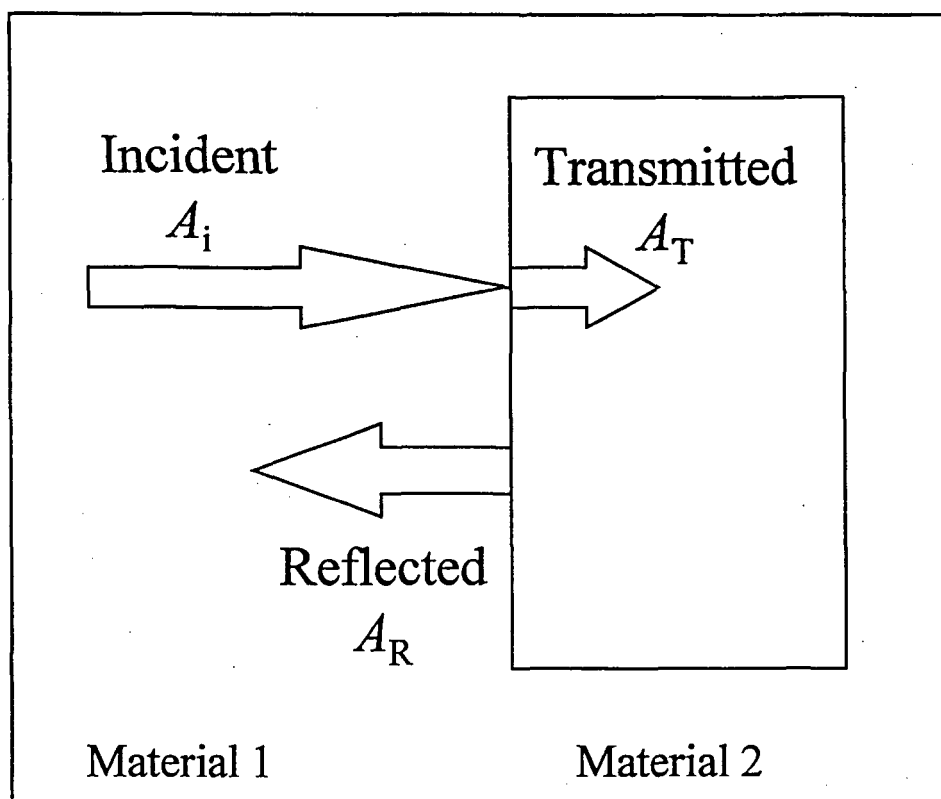


FIGURE 3. When an ultrasonic wave encounters a boundary between two materials, it is partly reflected and partly transmitted, the amount depending on the difference in acoustic impedances of the materials.

ratios of the amplitudes of the transmitted (A_t) and reflected (A_r) waves to that of the incident wave (A_i) are called the transmission (T) and reflection coefficients (R), respectively. The transmission and reflection coefficients for a plane wave incident upon a plane surface between two materials (Figure 3) are given by the following expressions:

$$T = \frac{A_t}{A_i} = \frac{2Z_1}{Z_1 + Z_2} \quad (5)$$

$$R = \frac{A_r}{A_i} = \frac{Z_1 - Z_2}{Z_1 + Z_2} \quad (6)$$

These equations indicate that only a small fraction of the ultrasound incident on a boundary between two materials of very similar acoustic impedance is reflected (e.g., oil and water), but that a large fraction is reflected when the two materials have very different acoustic impedances

(e.g., air and water). The extent of reflection at boundaries has important practical implications for many applications of ultrasound in the food industry (e.g., imaging, detection of foreign bodies, and design of measurement cells).

In many food applications, ultrasonic waves are incident on curved surfaces, rather than planar ones (e.g., the surface of an apple or the inside of a pipe). In this case, the fraction of ultrasound reflected and transmitted (refracted) depends on the angle of incidence as well as the acoustic impedances of the two materials.^{20,21} The relationship between the angles of the incident, reflected, and refracted waves is governed by the same laws as for optics, although the solutions are somewhat more complex, because both compressional and shear waves are generated for solids. Reflection of ultrasound from real food materials may also be complicated by the fact that many foods have surfaces that are not smooth (e.g., orange peel). Reflection of ultrasound from rough

or irregular surfaces leads to constructive and destructive interference between waves reflected from different points. Analysis of this type of reflected wave could be used to provide valuable information about surface structure.

2. Ultrasonic Adsorption and Relaxation

As an ultrasonic wave passes through a material its amplitude decreases because energy is absorbed by the material. The principal forms of adsorption of ultrasound in gases and liquids are thermal conduction, viscosity, and molecular relaxation.²² In solids, a number of other physical mechanisms may also contribute to the overall adsorption. By characterizing the extent and nature of ultrasonic absorption by a material it is possible to obtain valuable information about physical properties (e.g., thermal conductivity, viscosity, and various molecular relaxation processes).

When an ultrasonic wave propagates through a material it causes periodic fluctuations in the local temperature and pressure. Adsorption occurs because the mechanical energy stored in the ultrasonic wave can be diverted into other dynamic physicochemical mechanisms that exist within materials. Because these mechanisms are not totally efficient, some of the ultrasonic energy is lost as heat. For example, energy is lost because heat flows between regions of high and low temperature (thermal conduction) or because of friction caused by the movement of molecules relative to one another (viscosity). In liquids, the sum of the contributions from thermal conduction and viscosity to the overall adsorption is called the *classical adsorption*.

$$\frac{\alpha_{\text{classical}}}{f^2} = \frac{4\pi^2}{\rho c} \left(\frac{4\eta}{3} + \frac{(1-\gamma)\tau}{C_p} \right) \quad (7)$$

Here, η is the viscosity, τ is the thermal conductivity, γ is the ratio of specific heat capacities, and C_p is the specific heat capacity at constant pressure. The first term in the brackets is the contribution of viscous losses, and the second term is the contribution of heat conduction losses.

In systems where losses due to relaxation are insignificant, Equation 7 can be used to obtain information about changes in the thermal properties or viscosity of liquids from ultrasonic absorption measurements.

Relaxation occurs when energy associated with an ultrasonic wave perturbs a system that is in dynamic equilibrium. Consider a system in which there is a dynamic equilibrium between two states, A and B, with first-order rate constants for both the forward (k_1) and backward reactions (k_{-1}):



When an ultrasonic wave is applied to the system, the local pressure and temperature are altered, and so the system attempts to take up a new equilibrium position with new rate constants, k_1 and k_{-1} . The time taken for the concentrations of A and B to change to their new equilibrium values is characterized by a relaxation time t_R , which is related to the rate constants ($1/t_R = k_1 + k_{-1}$). The ultrasonic velocity and attenuation coefficient of a system with a single relaxation mechanism are frequency dependent:

$$\frac{\alpha_{\text{relaxation}}}{f^2} = \frac{A f_R^2}{f_R^2 + f^2} \quad (9)$$

$$c_{\text{relaxation}} = c_0 + (c_\infty - c_0) \left(\frac{f^2}{f_R^2 + f^2} \right) \quad (10)$$

Here, c_0 and c_∞ are the ultrasonic velocities of the system at zero and infinite frequency, respectively, f_R is the relaxation frequency ($=1/t_R$), and A is a parameter related to the magnitude of the relaxation mechanism. The frequency dependence of the velocity and attenuation coefficient in a system that has a single relaxation mechanism is illustrated in Figure 4. The ultrasonic velocity increases from a constant value at low frequency to another constant value at high frequencies. The $\alpha_{\text{relaxation}}/f^2$ contribution to absorption decreases with frequency, while the $\alpha_{\text{classical}}/f^2$ contribution remains constant (Figure 4). Measurements of the frequency depen-

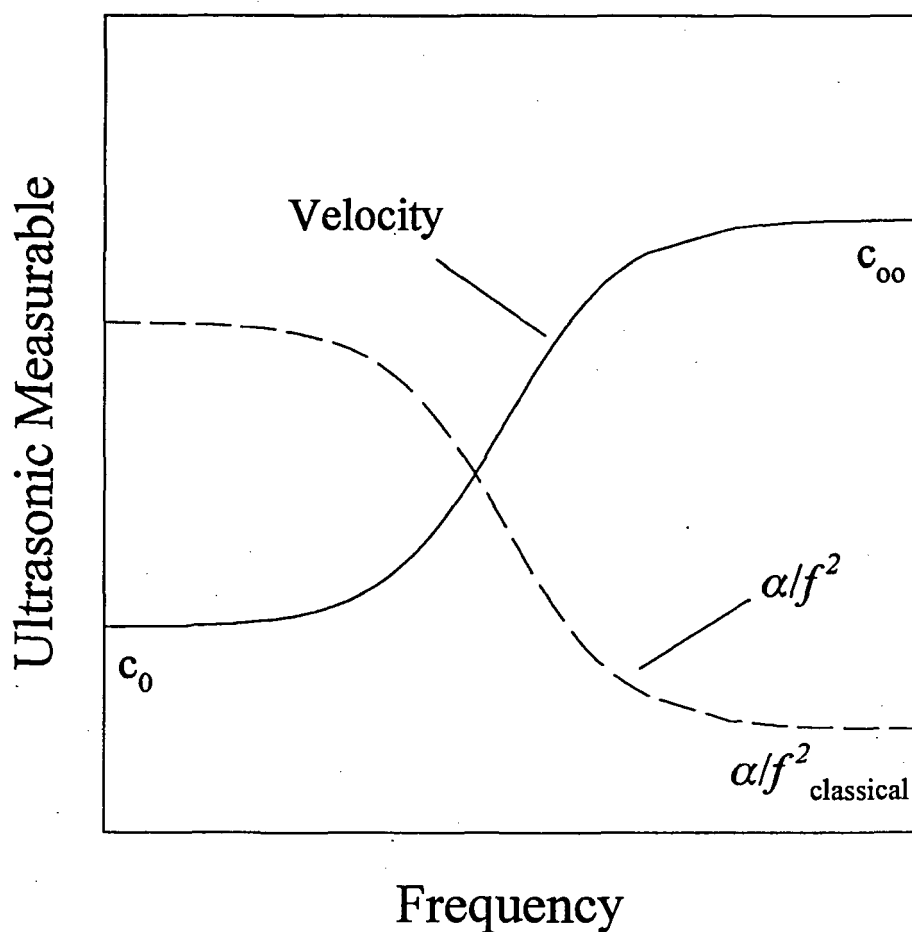


FIGURE 4. Molecular relaxation mechanisms within a material cause the ultrasonic velocity and attenuation coefficient to be frequency dependent. By measuring the ultrasonic properties of a material as a function of frequency, it is possible to obtain information about molecular relaxation phenomena.

dence of the velocity and attenuation coefficient of a liquid can therefore be used to study relaxation phenomena in materials. In many systems, more than one relaxation mechanism contributes to the overall signal, or the relaxation process is more complex than that represented by Equation 8. In these systems, more sophisticated data analysis is required to extract the relevant information. The frequencies typically used in ultrasonic analysis of materials range from about 100 kHz to 100 MHz, and so ultrasound can be used to study equilibria with relaxation times between about 10 ns and 10 ms. Among other things, this range includes conformational changes of proteins (helix-coil) and triglycerides, proton exchange, and hydration equilibria (see later).

3. Scattering of Ultrasound by Particles

The particles typically found in foods vary enormously in size, ranging from a few nanometers (e.g., protein aggregates) to a few centimeters (e.g., fruit pieces). It is often important for food scientists to establish the number, size, and location of these particles within a food. This can often be achieved by using ultrasonic techniques that utilize the scattering of ultrasound by particles. Scattering occurs when an ultrasonic wave incident upon some heterogeneity is redirected into directions that are different from that of the incident wave. One of the most important physical characteristics that determines the extent of scattering is the relationship between the wavelength (λ) of the ultrasound used and the dimen-

sions (d) of the particle. Just as in optics, the scattering of ultrasound by a particle can be divided into three regions.

1. The long wavelength limit (LWL), where the particle size is much smaller than the ultrasonic wavelength ($d < \lambda/10$)
2. The intermediate wavelength regime (IWR), where the particle size is approximately similar to ultrasonic wavelength ($\lambda/10 < d < 50\lambda$)
3. The short wavelength limit (SWL), where the particle size is much greater than the ultrasonic wavelength ($d > 50\lambda$)

The wavelength of ultrasound in water is about 15 mm at 0.1 MHz, 1.5 mm at 1 MHz, 150 μ m at 10 MHz, and 15 μ m at 100 MHz. The particles in foods may therefore fall into any one of the three regions, depending on the frequency of ultrasound used to probe the system being studied. Interpretation of ultrasonic measurements is much simpler when one is in either the SWL or the LWL region, and so the frequency used to analyze a particular food is often chosen so that this situation occurs. Theories used to interpret ultrasonic measurements in scattering systems have been reviewed by a number of workers.²³⁻²⁵

Measurements of the scattering of ultrasound by particles can be used to determine particle concentrations and size distributions in a fashion similar to light scattering techniques.²³ One can either measure the velocity and/or the attenuation of ultrasound as a function of frequency²³ or the angular dependence of the intensity of the scattered ultrasound.²⁵ An appropriate mathematical theory is then used to relate the ultrasonic measurements to the physical parameters of interest. Potentially, one of the most important applications of ultrasound in food systems is the characterization of particles in fluid colloidal dispersions, such as bubbly liquids (gas particles), emulsions (liquid particles), and suspensions (solid particles). The velocity and attenuation of ultrasound in fluid colloidal dispersions depends on the size, shape, and concentration of the dispersed particles, the frequency used, and the thermophysical properties of the component phases.²³ In the LWL, the following equation is used to relate

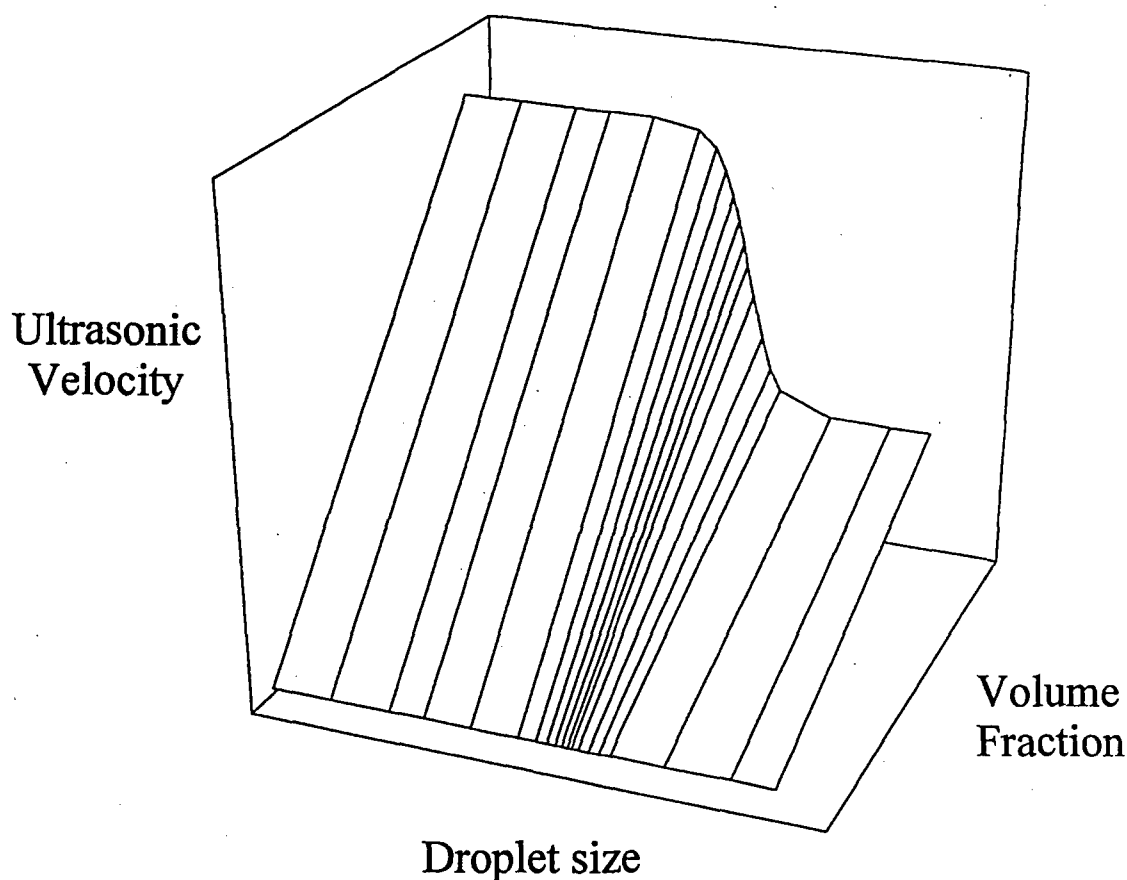
the measurable ultrasonic properties (velocity and attenuation) to the physical properties of interest:

$$\left(\frac{K}{k}\right)^2 = \left[1 - \frac{4i\pi n A_0}{k^3}\right] \left[1 - \frac{12i\pi n A_1}{k^3}\right] \quad (11)$$

Here, K and k are the complex wave numbers of the scattering medium and the continuous phase, respectively, n is the number of scatterers per unit volume, and A_0 and A_1 are the single particle-scattering amplitudes.²³ Values of A_0 and A_1 have been calculated for emulsions, suspensions, and bubbly liquids.²⁶ A similar equation can also be used to describe scattering in systems where the continuous phase is solid, but different values of A_0 and A_1 are needed, and an A_2 term must also be included.²⁶

The predominant sources of scattering in emulsions and suspensions are caused by visco-inertial and thermal transport processes that occur at the interface between the droplet and the surrounding medium.²³ The dependence of the ultrasonic properties of a typical emulsion on particle size in the LWL is shown in concentration in Figure 5. The velocity increases with increasing particle size (Figure 5A), while the attenuation coefficient multiplied by the wavelength ($\alpha\lambda$) has a maximum value (Figure 5B). Thus, it is possible to deduce the size of the particles in an emulsion by measuring its ultrasonic velocity or attenuation coefficient. In fact, a number of instrument manufacturers have recently developed particle-sizing equipment based on this principle.

It is worth discussing the ultrasonic properties of liquids containing gas bubbles in some detail because this has important practical consequences in a number of applications of ultrasound in the food industry. In some foods, we want to characterize the properties of the bubbles themselves (e.g., beer, carbonated drinks, whipped cream, and aerated desserts), whereas in other foods the gas bubbles may interfere with the property that we are trying to measure (e.g., the presence of air bubbles in mayonnaise can interfere with the ultrasonic determination of oil content or droplet size). The ultrasonic properties of bubbly liquids are dominated by a phenomenon known as *resonant* scattering.²⁷ In the presence of an ultrasonic wave, a gas bubble pulsates violently at



A

FIGURE 5. The ultrasonic velocity and attenuation coefficient of emulsions and suspensions depends on the size and concentration of the particles they contain, as well as the frequency of ultrasound used. By measuring the ultrasonic properties as a function of frequency, it is possible to determine both the size and concentration of particles in these systems.

a particular frequency (the resonance frequency, f_R) because of the large difference in compressibility of the bubble compared with that of the surrounding liquid. This causes most of the ultrasonic energy that is incident on the bubble to be scattered in all directions. The frequency at which resonance occurs depends on the size of the particles: the smaller the size, the higher the resonant frequency:

$$f_R^2 = \frac{3\rho_b c_b^2}{4\pi^2 r^2 \rho} \quad (12)$$

Here, ρ is the density of the liquid, ρ_b and c_b are the density and ultrasonic velocity of the

bubble, respectively, and r is the bubble radius. This equation is applicable to fairly large isolated bubbles ($r > 3 \mu\text{m}$); for smaller bubbles, the effects of viscosity, heat conduction, and surface tension have to be included.²⁷ The frequency dependence of the ultrasonic velocity and attenuation coefficient of a dilute bubbly liquid are shown in Figure 6. At frequencies well below the resonant frequency, the ultrasonic velocity is much lower than that of the continuous phase, and may even be lower than that of the gas phase. At resonance, the velocity is highly sensitive to frequency, while at frequencies well above resonance, it tends to the value for the continuous phase. The attenuation coefficient increases dra-

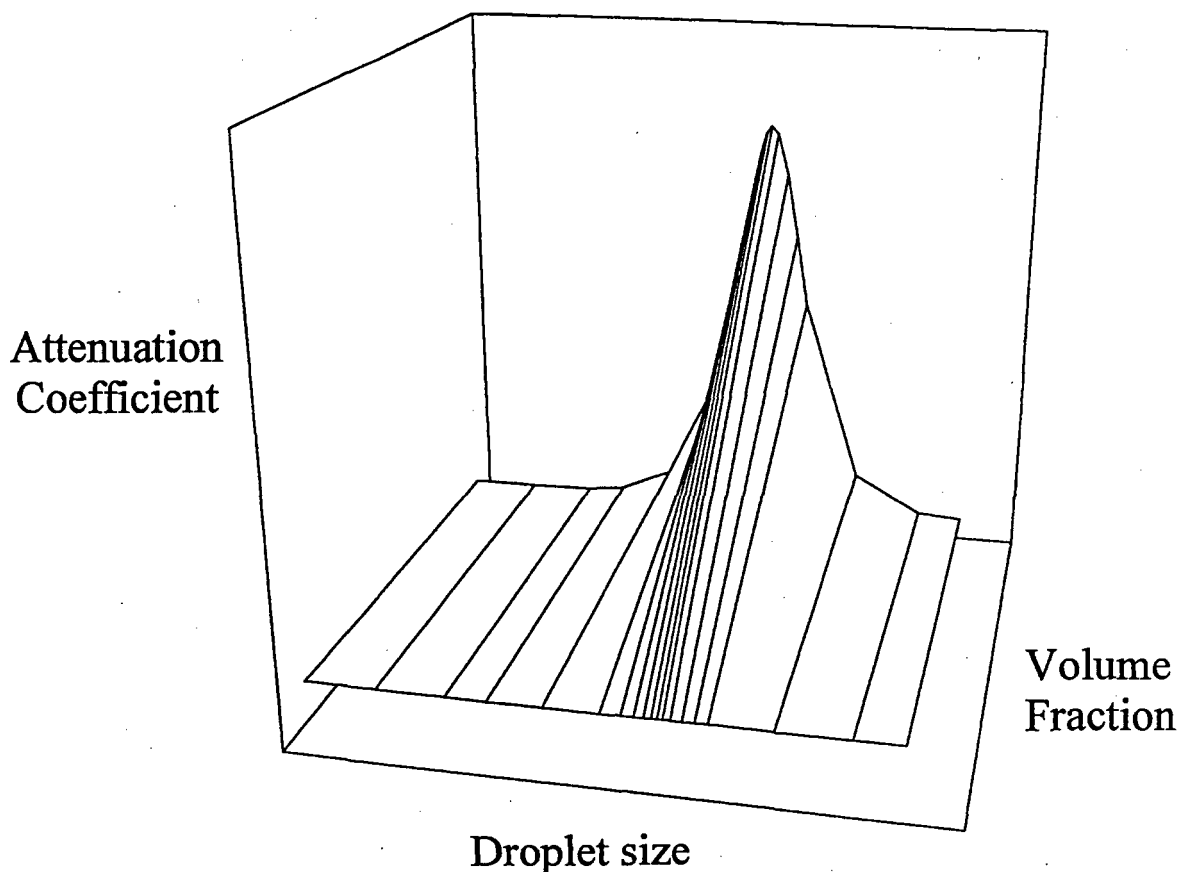


FIGURE 5B

matically at the resonance frequency. By measuring the velocity or attenuation coefficient as a function of frequency, it is possible to determine the concentration and size of bubbles in a dilute bubbly liquid. Nevertheless, the attenuation of ultrasound in bubbly materials can be so large that it is practically impossible to transmit enough ultrasound through them to make a measurement. Methods for overcoming this problem are discussed later (Section V.E).

Theories that describe ultrasonic propagation in systems where the overall scattering of ultrasound is weak (e.g., emulsions) are in good agreement with experimental measurements up to fairly high particle concentrations (>30%), but there are no theories that adequately describe ultrasonic propagation in systems where the scattering is strong (e.g., concentrated bubbly liquids). This is because of the difficulties of including multiple scattering and particle inter-

action effects in mathematical treatments of scattering.²³

D. Interpretation of Ultrasonic Measurements

Food scientists are not usually interested in the ultrasonic properties of materials per se, but in more practical properties, such as composition, structure, or physical state. An important stage in ultrasonic analysis is therefore the conversion of the measurable ultrasonic parameters into the physical properties of interest. Relationships between physical and ultrasonic properties of materials can be established in a number of different ways, but can be roughly divided into two different categories: those that use an empirical approach and those that use a theoretical approach. In the empirical approach, the relationship be-

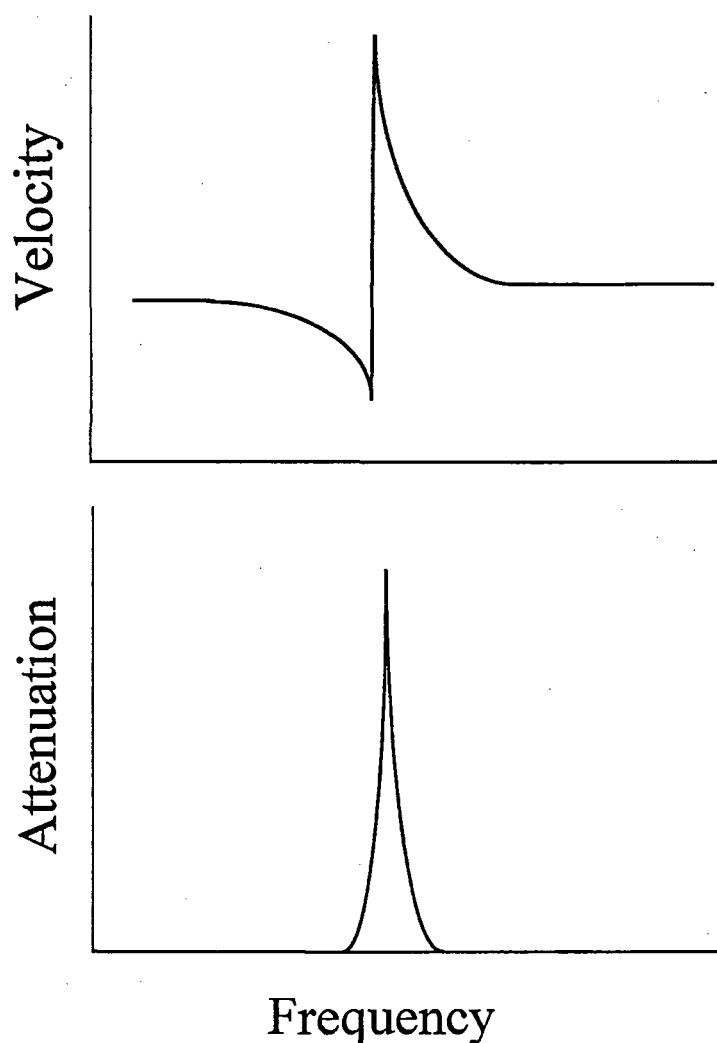


FIGURE 6. The ultrasonic properties of a liquid that contains small air bubbles are largely determined by resonant scattering. There is a large increase in the attenuation coefficient around the resonant frequency of the bubbles, as well as appreciable velocity dispersion.

tween the ultrasonic and physical properties is established **purely from experimental data**. A series of calibration experiments is carried out by measuring the ultrasonic properties of a number of samples of known physical properties, and a best-fit empirical equation is fitted to the data. This equation is then used to relate measurements of the ultrasonic properties of unknown samples to their physical properties. An example of this approach is the determination of the sugar content of fruit juices. A series of glucose solutions is prepared with different sugar concentrations, and their ultrasonic velocities are measured (Figure 7). This calibration curve is

then used to determine the sugar content of unknown samples from measurements of their ultrasonic velocity. The empirical approach has proved successful for many food materials, but has limited use when applied to structurally or compositionally complex materials. This is because a large number of factors contribute to the observed signal, and it is difficult to isolate or control the contribution of each component. Another problem is that an empirical relationship is only strictly applicable over the range of experimental constraints under which it was constructed. Serious errors may result if some experimental variable in the system being ana-

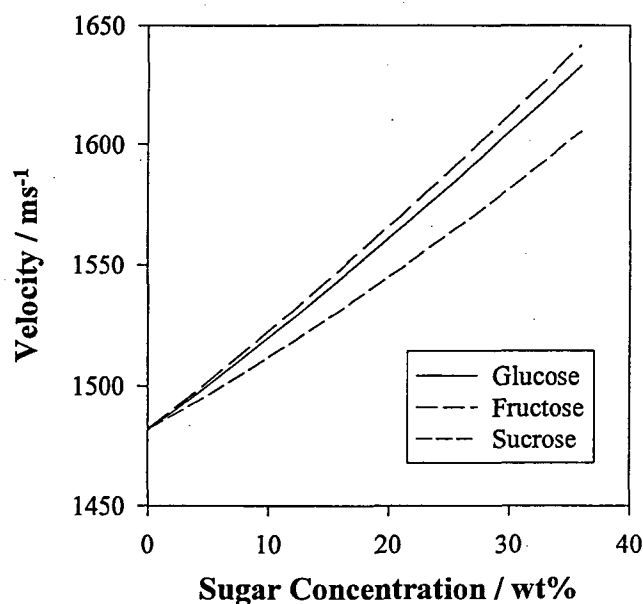


FIGURE 7. Variation of the ultrasonic velocity with sugar concentration for different saccharides dissolved in distilled water at 20°C.

lyzed was not taken into account in the preparation of the calibration curve.

The **theoretical approach** is often favored because it leads to a better understanding of how an ultrasonic wave interacts with a material. It also has the advantage that **predictions** about how a particular experimental parameter affects the ultrasonic properties of a material can be assessed without carrying out any actual experiments. In this approach, **mathematical theories derived to describe the propagation of ultrasound through materials are used to relate measurable ultrasonic parameters to the structure and physical properties of the various components within a material.** For example, Equation 11 can be used to relate the attenuation of ultrasound by a food emulsion to the size of droplets that it contains. The complexity and diversity of food materials means that no single theory can be used to interpret ultrasonic measurements in all foods. Nevertheless, there are a number of foods for which ultrasonic theories do exist for interpreting ultrasonic measurements.

1. Homogeneous Materials

The ultrasonic velocity of a homogeneous material is simply related to its physical proper-

ties (density and elastic moduli) by Equation 2. This equation can be used to provide information about the dynamic rheological properties of materials, from measurements of their ultrasonic velocity and density. The attenuation of ultrasound in homogeneous materials is due to the various absorption mechanisms described earlier. Measurements of the frequency dependence of the ultrasonic velocity and attenuation coefficient of a liquid can be used to provide valuable information about the viscosity, thermal conductivity, or molecular relaxation of liquids (Section II.C.2).

Many homogeneous foods are multicomponent systems, and their ultrasonic properties depend on the concentration and interactions of the various components. For ideal binary liquid mixtures and solutions, there is a simple relationship between the ultrasonic properties and the composition:

$$c^2 = \frac{1}{\kappa_E \rho_E} \quad (13)$$

$$\kappa_E = \phi \kappa_2 + (1 - \phi) \kappa_1 \quad (14)$$

$$\rho_E = \phi \rho_2 + (1 - \phi) \rho_1 \quad (15)$$

$$\alpha_E = \phi \alpha_2 + (1 - \phi) \alpha_1 \quad (16)$$

Equation 13 is basically the same as Equation 2, but is given in terms of the adiabatic compressibility ($\kappa = 1/K$) rather than the bulk modulus (K). In addition, the adiabatic compressibility and density are *effective* values (hence the subscript E), which depend on the composition of the system. The subscripts 1 and 2 refer to the two components, and ϕ is the volume fraction of component 2. Similar relationships can also be derived for solids, but a different value for the modulus is needed because both shear and compressional waves can propagate in solids. The above equations highlight the simple relationship between the ultrasonic properties of a material and its composition.

In practice, there are often significant deviations between the measured ultrasonic properties of real mixtures and solutions and predictions made using the above equations. This is because of nonideal behavior, the presence of chemical or molecular equilibria, or changes in the properties of materials on dissolution. The magnitude of the deviations between measured and predicted ultrasonic properties may provide useful insight into the nonideal behavior of solutions and mixtures.

2. Heterogeneous Materials

Many foods are heterogeneous materials that consist of a number of different components separated from each other by more or less distinct boundaries. The ultrasonic properties of these systems depend on reflection, scattering, refraction, and diffraction of ultrasound, in addition to the absorption mechanisms mentioned earlier. Ultrasonically, it is convenient to categorize foods according to the relationship between the dimensions of the heterogeneities (d) and the ultrasonic wavelength (λ). Foods in which $d \ll \lambda$ are called microheterogeneous, whereas those in which $d \gg \lambda$ are called macroheterogeneous. Obviously, there are some instances when the wavelength of ultrasound is similar to the dimensions of the objects in a food. In addition, it may be possible to move from one extreme to another simply by varying the ultrasonic frequency. Nevertheless, this categorization is useful because it determines how ultrasonic measurements on a particular material are interpreted.

In macroheterogeneous materials, we are mainly concerned with the reflection of ultrasonic waves from relatively large objects. As mentioned earlier, the fraction of ultrasound reflected from the surface of a material is governed by the acoustic impedances of the different components, the angle of incidence, and the smoothness of the interface (Section II.B.3). Information about the internal structures of macroheterogeneous foods can be obtained using techniques similar to those used in medical imaging or materials testing. The time-of-flight and amplitude of ultrasonic pulses that are transmitted through, or reflected from, internal structures are measured. The time-of-flight is related to the distance the ultrasonic pulse travels before it is reflected, whereas the amplitude is a measure of the attenuation within the material and the efficiency of reflection from the objects. It is possible to obtain one-, two-, or three-dimensional images of internal structures by measuring the ultrasonic properties at different locations on the surface of a material (Section III.A.3).

Ultrasound can also be used to provide information about the structure and concentration of particles in microheterogeneous materials, such as emulsions, suspensions, and bubbly liquids (see Section II.C.3). Mathematical theories available for interpreting measurements in these systems have recently been reviewed.²³⁻²⁵

III. EXPERIMENTAL TECHNIQUES

One of the major reasons that ultrasound has not been used more widely in the food industry has been the lack of commercial ultrasonic instrumentation specifically designed to characterize foods. Thus, food scientists who wanted to use ultrasound had to design and set up their own ultrasonic instruments, which requires a fairly thorough understanding of the basic physical principles of ultrasound. This situation is changing, largely because instrument manufacturers realize the huge potential market for ultrasonic techniques in the food industry. Consequently, a number of instrument manufacturers have developed ultrasonic instruments for analyzing foods that are now commercially available. As with any analytical technique, whether self-made or purchased commercially, it is important to understand the

basic principles of its operation. This knowledge aids in the selection of appropriate equipment, as well as minimizing the possibility of making inaccurate or imprecise measurements.

A. Measurement Systems

The ultrasonic properties of materials can be measured using a wide range of different types of measurement system. The choice of a particular measurement system depends on a number of factors, including the nature of the material being tested, the property being measured, the equipment available, the initial expense of purchasing the equipment, maintenance costs, the desired speed and accuracy, and whether the measurements are to be made on-line or in a laboratory. Despite the variety of ultrasonic techniques available, most of them can be categorized according to the nature of the ultrasonic waves propagated into the sample being analyzed (Figure 8), that is, *pulsed* or *continuous wave* (cw). Pulse techniques are by far the most widely used for measuring the ultrasonic properties of materials because they are easy to oper-

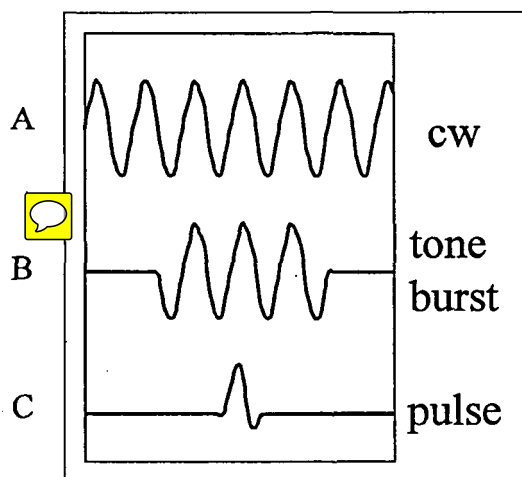


FIGURE 8. Typical signals used in ultrasonic testing of materials. (A) In a *continuous wave*, all the ultrasonic energy is focused at a single frequency. (B) In a *tone-burst pulse*, the ultrasonic energy is spread out over a *narrow range around a particular frequency*. (C) In a *broad-band pulse*, the ultrasonic energy is spread out over a wide range around a particular frequency.

ate, measurements are rapid and noninvasive, and the technique can easily be automated. Continuous wave techniques are used when highly accurate measurements are needed and tend to be found in specialized research laboratories. Even though pulse techniques are less accurate than cw techniques, they are accurate enough for most applications involving foods.

1. Pulsed Techniques

The simplest and most widely used technique for making ultrasonic measurements is called the *pulse-echo technique*. More sophisticated pulsed methods have been developed to improve the accuracy of measurements; however, the operating principles are basically the same as those of the pulse-echo technique.^{28,29} For this reason, only the pulse-echo technique is described, along with some of its most important modifications. A typical experimental configuration consists of a *measurement cell* that contains the sample, a *signal generator*, an *ultrasonic transducer*, and an *oscilloscope* (Figure 9A). The signal generator produces an *electrical pulse of an appropriate frequency, amplitude, and duration* (Figure 8). This pulse is converted into an ultrasonic pulse by the transducer, which then propagates through the sample until it reaches the far wall of the measurement cell, where it is reflected back to the transducer (Figure 9B). The transducer now acts as a receiver and converts the ultrasonic pulse back into an electrical pulse that is displayed on the oscilloscope. Because each pulse is only partly transmitted and partly reflected at the cell walls, a series of echoes is observed on the oscilloscope (Figure 9C). The velocity, attenuation coefficient, and impedance of a sample can be determined by analyzing these echoes.

Each echo has traveled a distance twice the cell length d than the previous echo, and so the velocity can easily be calculated by measuring the time delay t between successive echoes: $c = 2d/t$. The cell length is determined accurately by measuring the time delay for a material of known ultrasonic velocity, such as distilled water ($2d = c_w t_w$, where the subscript w refers to water). The attenuation coefficient is determined by measuring the decrease in amplitude of the successive

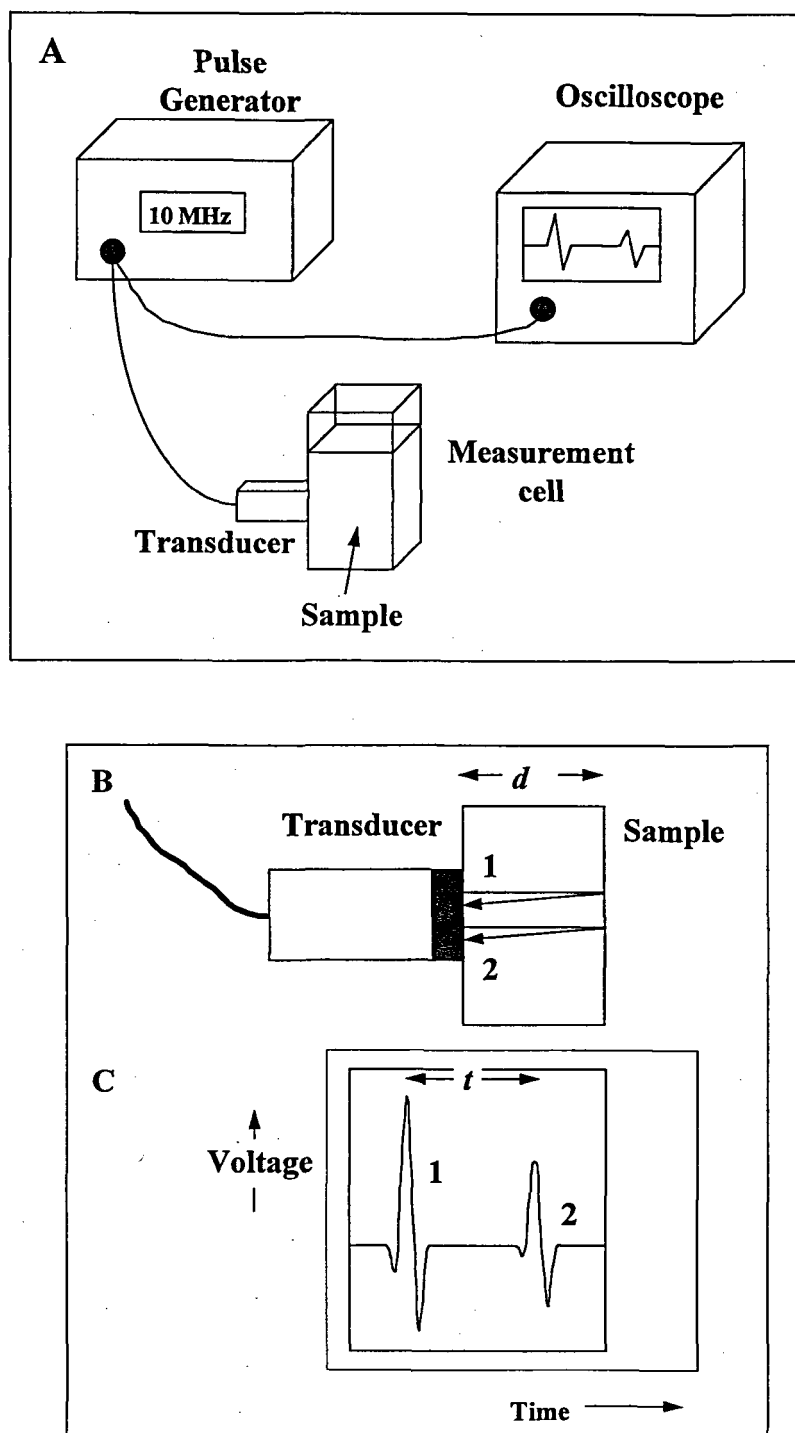


FIGURE 9. (A) A typical experimental configuration for carrying out ultrasonic pulse-echo experiments consists of a signal generator to generate electrical pulses, an ultrasonic transducer to convert electrical pulses into ultrasonic pulses, a measurement cell to contain the sample, and an oscilloscope to display the received echoes. (B) The transducer converts the electrical pulse to an ultrasonic pulse. This pulse travels across the sample, is reflected from the inside wall of the measurement cell, and returns to the transducer where it is detected. (C) Because the pulse is partly reflected and partly transmitted, a series of echoes is observed on the oscilloscope. The ultrasonic velocity and attenuation coefficient are determined from the time between successive echoes and the decrease in the amplitude of the echoes with distance traveled, respectively (see text for details).

echoes: $A = A_0 e^{-\alpha d}$. The specific acoustic impedance of a sample is determined by measuring the fraction of ultrasound that is reflected from the boundary between the measurement cell and the sample.³⁰ Some of the most important factors that must be considered when designing a measurement system for making accurate ultrasonic measurements are discussed in Section III.B.

A commonly used alternative to the pulse-echo technique is the **through transmission** technique. This technique utilizes two ultrasonic transducers, one as a generator and the other as a receiver. The sample is placed between the transducers, and the time-of-flight and amplitude of an ultrasonic pulse that is propagated across the sample is measured. The ultrasonic velocity ($c = d/t$) and attenuation coefficient ($A = A_0 e^{-\alpha d}$) are determined from these measurements. **The through transmission technique is used to analyze samples that highly attenuate ultrasound because the small pulse received from the sample can be isolated from the large pulse used to excite the transducer.**

For many food applications, it is useful to measure the frequency dependence of the ultrasonic properties of the food being tested because more information about its properties can be obtained. **Frequency scanning** can be achieved in two different ways using pulsed techniques. In the first approach, a transducer is **"tuned" to a particular frequency by using a tone-burst pulse from a signal generator**¹³ (i.e., a pulse that contains a number of cycles of a given frequency) (Figure 8B). The ultrasonic velocity and attenuation coefficient are then determined by measuring the **time-of-flight and amplitude of the echoes at each successive frequency**. In the **second approach**, a high-voltage, short-duration electrical pulse is used to excite the transducer to **simultaneously generate a wide range of frequencies**^{30,31} (Figure 8C). Fourier transform analysis of the echoes is then used to obtain information about the frequency dependence of the ultrasonic properties. The advantage of the Fourier transform technique is that a range of frequencies can be accessed using a single pulse of ultrasound, and so measurements are extremely rapid.

2. Continuous-Wave Techniques

Continuous wave ultrasound is utilized in a number of ultrasonic measurement devices, but the most commonly used is the **interferometer**.²⁹ A simple interferometer is illustrated in Figure 10. The same basic components are used for a cw experiment as for a pulsed experiment: a signal generator, a transducer, a measurement cell, and an oscilloscope. Nevertheless, their function and arrangement are slightly different. The sample is contained in the measurement cell, between an ultrasonic transducer and a reflector plate that is capable of moving vertically through the sample. The signal generator applies a sine wave of suitable frequency and amplitude to the transducer. The transducer converts this into an ultrasonic sine wave that propagates into the sample and is reflected back and forth between the reflector plate and the front face of the transducer. **Standing waves are produced in the sample**, and the amplitude of the signal received by the transducer goes through a series of maxima and minima as the reflector plate is moved vertically because of destructive and constructive interference. The velocity and attenuation coefficient are determined by analyzing the resulting signal.

The distance between successive maxima is equal to half the ultrasonic wavelength (λ) of the material, and so the velocity can be calculated: $c = f\lambda$. The amplitude of the maxima decrease as the distance between the reflector plate and transducer is increased because of attenuation by the sample, reflection at the boundaries and diffraction. The attenuation coefficient is determined by measuring the amplitude of the maxima and minima as a function of distance for the sample and for a calibration material. The accuracy of the measurements can be improved by measuring the amplitude and distance between a large number of successive maxima.

The frequency of the measurement **is determined by the resonant frequency of the crystal in the transducer (see below)**. As well as being operated at its resonance frequency, a transducer can also be operated at odd harmonics of its resonant frequency (i.e., $f, 3f, 5f, 7f \dots$), although the amplitude of the signals generated decreases with increasing frequency. Frequency scanning can

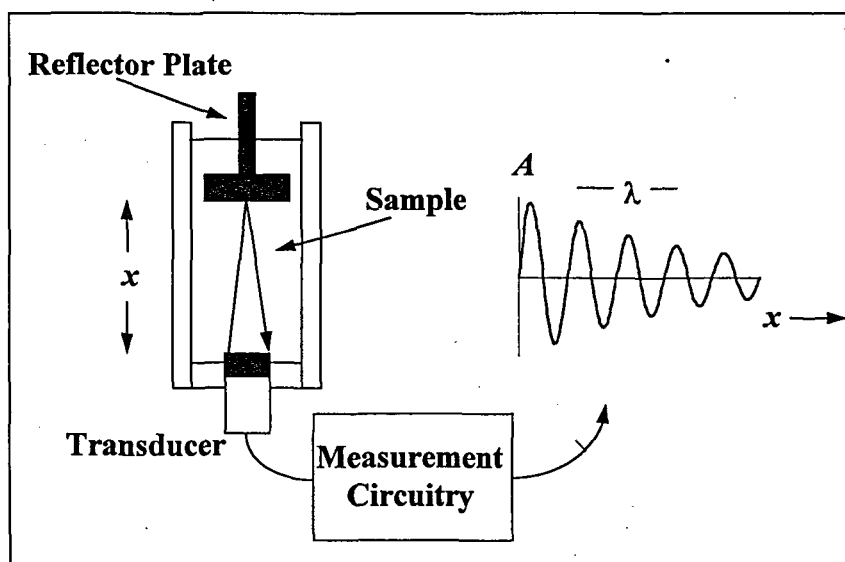


FIGURE 10. Experimental configuration for carrying out ultrasonic interferometry experiments. The sample is placed in a measurement cell, between an ultrasonic transducer and a reflector plate. A continuous wave is applied to the transducer, and a series of standing waves are set up in the sample. Moving the reflector plate through the sample leads to the observation of a number of peaks (constructive interference) and nulls (destructive interference). The wavelength of ultrasound in the sample (which is related to the ultrasonic velocity) is equal to twice the distance between successive maxima.

therefore be carried out using a cw technique by making a measurement at the resonant frequency, and then at each of the odd harmonics, by using electrical input signals of appropriate frequency to excite the transducer. This technique is more time consuming and laborious than the Fourier transform technique described earlier because measurements have to be carried out separately at each frequency.

3. Ultrasonic Imaging

Ultrasonic imaging is routinely used in materials testing and medical physics to provide one-, two-, and three-dimensional images of the internal structures of biological and nonbiological materials.^{32,33} It is not surprising therefore that it can also be used to provide useful information about many food systems. Imaging techniques use short-duration ultrasonic pulses so that reflections from objects within a sample can be distinguished from one another. An ultrasonic pulse is reflected when it encounters a boundary between

two materials of different acoustic impedance (Section II.C.1). The time delay between the transmission of a pulse and the return of its echo depends on the distance the pulse has traveled through the sample before it was reflected. If the velocity of ultrasound (c) in the material is known, it is simple to convert time-of-flight measurements to distances: $d = ct$. Thus, it is possible to determine the location of objects within materials. Measurements of the amplitude of echoes provides information about the attenuation coefficients and impedances of the different components.

A number of methods have been developed to generate and display ultrasonic images, the three most common being the A-scan, B-scan, and C-scan. An A-scan is simply a plot of signal amplitude vs. time, and is the signal usually observed on an oscilloscope (or similar instrument) when an ultrasonic pulse is transmitted into a sample and reflected from any internal structures (Figure 11). Because the time-of-flight of an echo is simply related to the distance it has traveled, an A-scan provides a one-dimensional image along

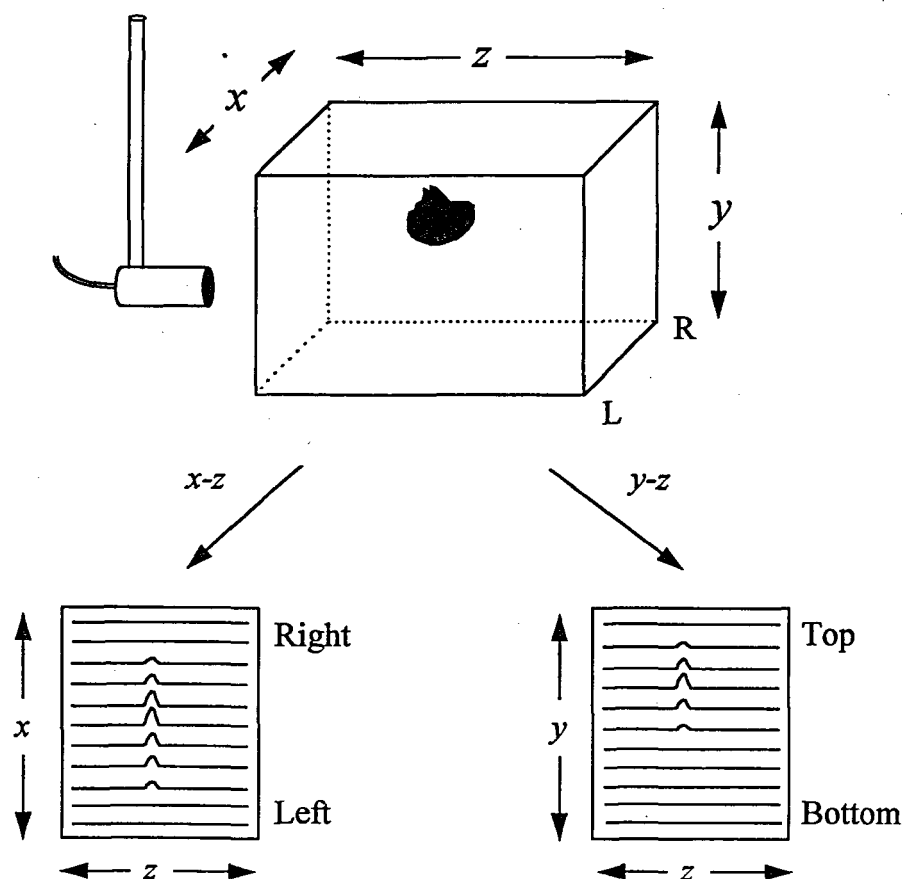


FIGURE 11. Ultrasound can be used to create two- and three-dimensional images of food materials by physically moving the transducer across the surface of the sample and measuring the time delay and amplitude of the returning echoes.

the z -axis, from which the location of objects can be deduced. More detailed information about the internal structure of samples is obtained by physically moving the transducer in the x - y plane and acquiring an A-scan at each location. A transducer is fixed to a mechanical device whose position can be varied using a series of stepper motors (Figure 11). The operator defines the area over which the transducer is moved and the distance between each location at which a measurement is taken. Thus, an array of data is generated that corresponds to an A-scan at each position in the x - y plane. The problem is to represent this information as a meaningful image that can be displayed on a computer screen. The B-scan and C-scan are two methods commonly used to achieve this. In a B-scan, one selects a particular time-of-flight (e.g., 10 μ s) along the z -axis and displays

the amplitude of the signal on the screen as a grey scale (or color scale; e.g., white corresponding to a low amplitude and black to a high one). This procedure is carried out for each A-scan acquired in the x - y plane, thus providing a two-dimensional image of the sample at a particular slice across the z -axis. In a C-scan, one focuses on a certain feature within an A-scan (e.g., a particular echo) and measures the change in its time-of-flight or amplitude when the transducer is moved in the x - y plane. Again, a grey scale is used to represent the time-of-flight or amplitude.

The time taken to obtain a complete image depends on the sophistication of the imaging equipment, as well as the number of measurements per sample that the operator decides to carry out. Typically, an image containing 100 by 100 A-scans would take a few seconds to a few minutes to

acquire. A number of ultrasonic imaging instruments have been developed for examining systems that undergo more rapid changes³³ (e.g., beats of the heart). These instruments use an array of transducers, rather than a single one, to obtain an image. There may be certain dynamic processes in the food industry where this type of imaging instrument is more suitable.

The resolution of any imaging device depends on the minimum distance between two objects that can be clearly distinguished. The resolution in the z-direction depends on the duration of the ultrasonic pulse (e.g., a 1- μ s pulse can distinguish between two objects in water that are greater than 1.5 mm apart). Pulses as short as 10 ns in duration can be generated to enhance resolution in the z-direction. The minimum distance that can be resolved in the x-y plane depends on the size of the steps selected by the operator between which each A-scan is acquired. In practice, the resolution is often worse than this minimum distance because the area of a material that is imaged depends on the diameter of the ultrasonic transducer used (typically between 5 and 25 mm) and the tendency for beam spreading to occur (see Section III.B.1). This means that even though it is possible to observe changes in the properties of a sample over a small distance, the details of the

objects within the sample cannot be clearly distinguished.

4. On-Line Sensors

It is widely recognized that there is a lack of suitable sensors for providing information about the properties of foods on-line during processing.^{1,2} One of the most promising applications of ultrasound in the immediate future is therefore as an on-line sensor.¹⁴ There are a number of important attributes that any on-line sensor must have. It should be capable of rapid and precise measurements, it should be nondestructive and noninvasive, it should be robust and relatively low cost, and it should be hygienic and capable of withstanding the pressures and temperatures normally encountered during food manufacturing and cleaning-in-place. In addition, it should have a computer interface so that the information can be used directly by a process control unit. Ultrasound has all of these attributes and is therefore ideally suitable for application in the food industry.

The simplest on-line ultrasonic sensor consists of an ultrasonic transducer fixed into a piece of pipe through which the food flows (Figure 12). A pulse of ultrasound is generated by the trans-

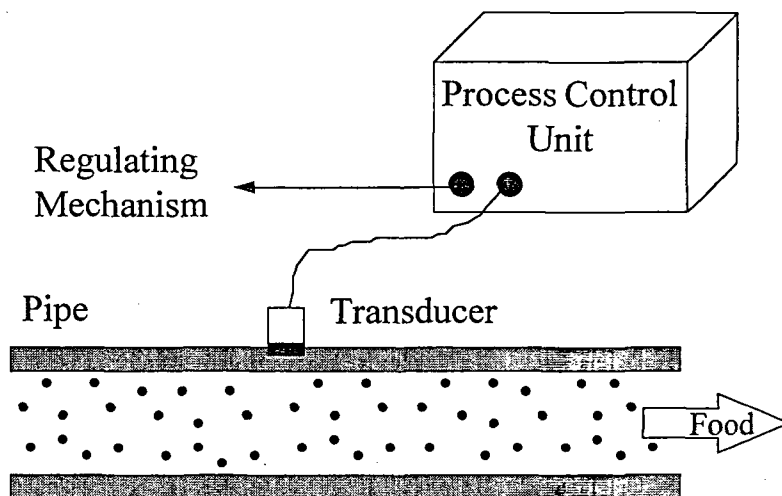


FIGURE 12. The properties of a food can be measured in real time during processing by having an ultrasonic sensor fixed into the pipe through which the food flows.

ducer and travels through the pipe and across the food before being reflected back to the transducer by the back wall of the pipe. The ultrasonic velocity and attenuation coefficient of the food are determined by measuring the time-of-flight and amplitude of the received echo (Section III.A.1). Appropriate empirical or theoretical equations are then used to relate the measured ultrasonic properties to the physical properties of interest to the food manufacturer (e.g., solid fat content, composition, or droplet size). It is possible to have a series of ultrasonic sensors that monitor the properties of a food at various critical locations within the process, thus giving manufacturers greater control over the properties of the final product.

There are already a number of commercial on-line sensors based on ultrasound that can be used to characterize the properties of food materials. Nearly all of these use ultrasonic velocity measurements at a single frequency to obtain information about the composition of binary liquids and solutions. For more complex applications, such as particle sizing or concentration determinations in emulsions, it is often necessary to measure the frequency dependence of the ultrasonic properties. A number of instrument manufacturers are currently developing on-line sensors for carrying out frequency-scanning ultrasonic measurements.

B. Components of Measurement Systems

A number of key components are common to most ultrasonic measurement systems: transducer, signal generator, digitizer, display, and measurement cell. An understanding of how each of these components operates is an important aid to the design of measurement systems and the interpretation of ultrasonic measurements.

1. Transducer

Transducer selection is one of the most crucial considerations when designing any ultrasonic experiment. An ultrasonic transducer is a device that converts electrical energy into mechanical

energy and vice versa. The most common type of transducers used in ultrasonic materials testing contain a single *piezoelectric* crystal (usually made from either quartz or a ceramic such as lithium niobate). Piezoelectric materials generate an electrical potential when they are deformed along a certain axis, and change their dimensions when an electrical potential is applied across them, and so they can be used as both generators and receivers of ultrasound.¹⁹ The ultrasonic transducers most commonly used to test foods in the food industry consist of a piezoelectric crystal within a protective metal casing (Figure 13). The crystal is usually bonded to a front plate that protects it from damage, and may also be bonded to a backing material to dampen its oscillations. The acoustic impedance and thickness of the front plate is chosen so as to optimize the energy output of the transducer. Three important factors must be considered when choosing a suitable transducer: the range of frequencies it is capable of generating, its diameter, and its acoustic matching.

The frequency of ultrasound chosen for a particular experiment depends on the type of information required from the material being tested (e.g., composition, structure, and flow rate). A number of experimental factors that affect the accuracy of ultrasonic measurements also depend on the frequency of ultrasound used (e.g., resolution of successive echoes, diffraction, interfer-

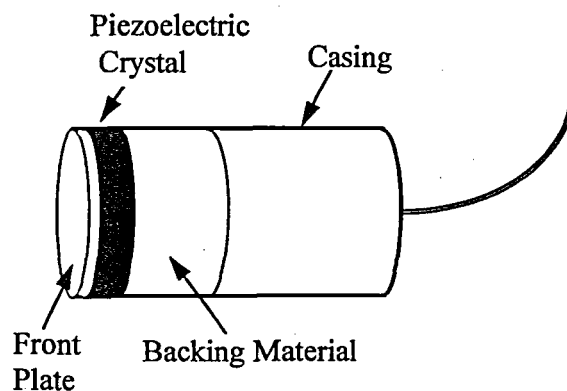


FIGURE 13. Design of a typical piezoelectric transducer used to test foods.

ence, and highly attenuative samples; see below). Most experiments utilize ultrasonic waves with frequencies ranging from about 0.1 to 100 MHz.

The range of frequencies that a given transducer can generate depends on two factors: the resonant frequency and the degree of damping of the piezoelectric crystal. The resonant frequency of an ultrasonic transducer depends on the thickness of the piezoelectric crystal: the thicker the crystal, the lower the resonant frequency. The degree of damping determines the range of frequencies produced. An undamped transducer “rings” for a long time when it is excited and produces most of its energy over a very narrow range of frequencies centered at the resonant frequency. This type of transducer is commonly used for continuous wave applications.

A transducer that contains a highly damped crystal is called a *broad-band* transducer. This is because the very short duration pulse produced by the crystal when it is excited generates a wide range of frequencies centered near the resonant frequency (Figure 8C). This type of transducer is used when very narrow pulses are essential (e.g., to distinguish between echoes reflected from objects that are close together) or when carrying out a frequency-scanning experiment using a Fourier transform technique. The difference between a damped and undamped transducer can be appreciated by a simple analogy. If one takes a hammer and hits a bell (which is normally undamped), it rings for a long time at a particular frequency, but if one takes hold of the edge of the bell, the ringing quickly dies away because the acoustic energy is dissipated.

The diameter of a transducer determines the region within a material that is analyzed in an ultrasonic experiment. It is often assumed that an ultrasonic wave travels through a material with a diameter equal to that of the transducer that generated it. In practice, the finite size of the transducer relative to the ultrasonic wave means that diffraction (beam spreading) occurs³³ (Figure 14). The field produced by an ultrasonic transducer can be divided into two regions: the near field and the far field. The near field extends a distance x from the front of the transducer and depends on the diameter of the transducer and the wavelength of ultrasound: $x = D^2/4\lambda$. In this region, the diameter of the ultrasonic beam is approximately equal

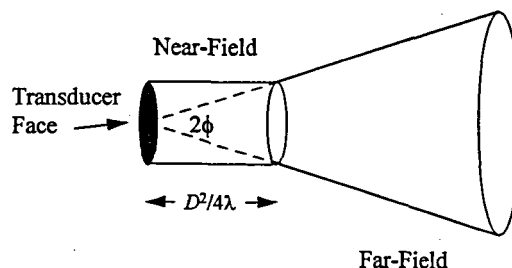


FIGURE 14. An ultrasonic wave spreads out when it leaves an ultrasonic transducer because of diffraction effects.

to the diameter of the transducer. In the far field, the ultrasonic beam diverges (Figure 14). Diffraction effects can cause serious errors in both time and amplitude measurements and should be corrected for if accurate ultrasonic measurements are essential.^{34,35} Another problem associated with the diffraction of ultrasonic waves is side wall reflections. If the angle of beam spreading is large enough, some of the ultrasonic wave is reflected from the side walls of a sample, or the cell that contains it, and interferes with the wave passing directly through the sample. This effect can also lead to appreciable errors in ultrasonic measurements, and experiments should be carefully designed to avoid it.

Another important experimental factor that should be considered when selecting a transducer for a particular application is the *acoustic matching*. Ultrasonic transducers are usually manufactured with a thin epoxy or ceramic wear-plate in front of the piezoelectric crystal (Figure 13). By varying the type of material used to manufacture the wear plate and its thickness, it is possible to acoustically “match” the transducer to a particular type of material (i.e., optimize the transmission of ultrasound from the transducer to the material). Most commercial transducers are acoustically matched to either steel or water. If the transducer is going to be in direct contact with a food sample, it is usually better to use a transducer that is matched to water, but if the sample is contained in some kind of measurement cell (e.g., a glass cuvette), transducers matched to steel are often better. It is sometimes possible to have transducer manufacturers match them to a particular material.

2. Signal Generator

The electrical input to an ultrasonic transducer is produced by a *signal generator*. The type of electrical input used depends on the particular application, and may be continuous wave or pulsed. A series of different types of electrical inputs commonly used in ultrasonic experiments is illustrated in Figure 8. **The experimental parameters that one must select are the frequency, amplitude, and duration of the input signal.** The frequency content of the electrical input should correspond to the range of frequencies that the transducer is capable of generating. For cw experiments, a sine wave (which has a single frequency and an infinite duration) is used (Figure 8A); for pulsed Fourier transform experiments, a short-duration pulse (which contains a wide range of frequencies) is used (Figure 8C); and for tone-burst experiments, a pulse containing a predetermined number of cycles is used (Figure 8B). The amplitude of the pulse should be large enough so that an observable signal is obtained, but not so large as to damage the sample.

3. Signal Digitization, Display, and Processing

Once an ultrasonic wave has been generated and propagated into a sample, it is necessary to capture and analyze the resulting signal. The signal that returns from the ultrasonic transducer is an analog electrical signal, **which can be displayed directly on an analog oscilloscope.** Nevertheless, it is usually more convenient to digitize the signal prior to display. **Digitization is carried out using an analog-to-digital converter,** which may be part of a digital storage oscilloscope, a card in a personal computer, or some other measurement device. The two most important features to consider when choosing an analog-to-digital converter are the **digitization rate and the amplitude resolution.** The digitization rate is the number of points along a signal that are sampled every second. To represent a sine wave accurately, **it is necessary to sample at a rate that is at least twice the highest frequency component of the signal being analyzed.** Typically, the highest frequency used in an ultrasonic experiment is

about 100 MHz, and so sampling rates of at least 200 MHz should be used. The digitization rate also determines the shortest time interval that can be measured between two points: the faster the rate, the shorter the time that can be measured. The amplitude resolution of a digitizer determines how precisely the amplitude of the analog signal can be reproduced. Modern digital storage oscilloscopes typically have amplitude resolutions of 8 bits.

Once the signal from a transducer has been digitized, it can be displayed on the screen of an oscilloscope or a personal computer and then analyzed using various signal-processing techniques, or it can be analyzed directly. A few of the most commonly used signal-processing techniques are mentioned below. **Averaging** of a number of signals can be used to improve the signal-to-noise ratio. High-frequency noise can be removed by **digital smoothing**, which works by replacing each point of the received signal with an average of itself and its nearest neighbors (two, three, or more). The signal can be **filtered above or below certain frequency cutoff points** to remove frequency components not associated with the signal of interest. **Cross-correlation techniques can be used to determine the time interval between two echoes.**

Ultrasonic experiments normally produce a signal that is in the **time domain (i.e., amplitude vs. time).** In many applications it is useful to know the frequency content of a signal. A mathematical technique known as the *Fourier transformation* has been developed to convert signals from the time domain into the **frequency domain.** This method works on the principle that **a signal in the time domain can be represented as a series of sine waves of different frequency, magnitude, and phase.** The frequency dependence of the ultrasonic velocity and attenuation coefficient of a material can be calculated from the measured phase and magnitude at each frequency.^{30,31} This technique is particularly useful for determining the frequency content of broad-band ultrasonic pulses. The ultrasonic properties of a material can thus be measured over a range of frequencies using a single ultrasonic pulse, which is much faster than using the tone-burst or cw ultrasonic techniques mentioned earlier.

4. Measurement Cell

Careful design of the cell used to contain the sample can make the difference between a successful and unsuccessful application of ultrasound.²³ The measurement cell should be manufactured so that it has smooth and parallel walls of an appropriate thickness and separation. Parallel walls are necessary to prevent phase cancellation of waves across the face of a transducer, which can lead to constructive or destructive interference of the signal. The cell walls should be thick enough so that any ultrasound that reverberates within them does not interfere with the signal being analyzed. The cell should be constructed from materials that are chemically inert and capable of withstanding any temperature or pressure variations that the cell may be subjected to. The ultrasonic properties of materials are temperature dependent, and so it is important to thermostat the measurement cell to control the temperature, or simultaneously measure the temperature and ultrasonic properties, and make an appropriate temperature correction.

IV. FOOD PROPERTIES MEASURED BY ULTRASOUND

During the past 50 years, ultrasound has been used to measure a wide variety of different properties of foods (Table 1). In this section, we discuss the most important types of application of ultrasound in the food industry. In the following section, we discuss applications of ultrasound to specific types of food.

A. Distance/Level Detection

Ultrasonic devices are commercially available that can be used to accurately measure the thickness of materials.³⁶ An ultrasonic transducer is pressed against one side of the material, and a narrow pulse of ultrasound is transmitted into the material. The time taken (t) for the pulse to travel through the sample and be reflected back to the transducer is measured. The distance (d) that the pulse has traveled is calculated from a knowledge of the velocity of ultrasound (c) in the material:

$d = ct/2$. Values of the velocity of ultrasound for many materials are tabulated in the literature, or can be measured in the laboratory. The advantage of using ultrasound over other more conventional methods of distance measurement is that it is only necessary to have access to one side of the material being tested. Thus, it is possible to determine the thickness of materials that are difficult to measure using conventional techniques (e.g., fouling of pipes, chocolate layers on confectionery, layers of fat or lean tissue in meat, liquids in a can, and egg shells). The thickness of materials can be determined to within 1% for samples ranging from 0.02 mm to 1 m.³⁶

A related application is the determination of the level of liquids in tanks or other containers. Two types of ultrasonic technique can be used for this purpose.¹⁷⁹ In the first technique, an ultrasonic transducer located at the bottom of the tank generates a pulse of ultrasound that travels vertically through the liquid until it reaches its surface, where it is reflected back to the transducer. The time-of-flight of the pulse is directly proportional to the height of the liquid. In the second technique, an ultrasonic transducer is used to determine whether a liquid is above or below a certain height. The transducer is fixed to the outside wall of a tank and generates a pulse of ultrasound that travels across the wall, until it is reflected back to the transducer from the wall/sample interface. If there is no liquid present, the returned echo has a high amplitude because of the large acoustic mismatch between the wall and air; however, if liquid is present in the tank, the amplitude of the returned echo decreases because the acoustic impedances of the sample and wall are more similar. Thus, an ultrasonic transducer can be used to detect the presence or absence of a liquid in a tank at a particular position. The advantage of this type of sensor is that it does not require that the ultrasonic pulse travel through the liquid, which can be a problem for highly attenuating materials.

B. Extraneous Matter

Extraneous matter, such as pieces of metal, glass, or wood, sometimes find their way into foods before or during processing. The presence of these materials is undesirable, and therefore it

is important to have analytical methods to detect them so that the extraneous matter can be removed or the sample rejected. Most foods are optically opaque, and so methods that utilize light cannot be used. In contrast, many foods are acoustically transparent and are therefore amenable to study by ultrasound. When an ultrasonic pulse is transmitted into a sample, it is reflected from any surfaces that it encounters, providing that there is a large enough difference in the acoustic impedance of the object and that of the surrounding material (Section II.C.1). Materials such as glass, metal, and wood have much larger acoustic impedances than other food components, and so can easily be detected by ultrasound. The presence of extraneous matter in a sample can be determined by measuring the time interval between the ultrasonic pulse reflected from it and that reflected from the back of the sample (Figure 11). By moving an ultrasonic transducer around the sample, it is possible to determine both the size and location of an object. This application is a simple example of the imaging techniques mentioned earlier.

C. Temperature

Ultrasonic thermometers are based on the principle that the velocity of ultrasound in a material is dependent on the temperature.¹⁷⁹ Ultrasonic thermometers have been developed that consist of a cylindrical rod of one material (the "delay line") bonded to a short cylinder of another material (the "end piece"). An ultrasonic pulse travels along the length of the delay line, and is partly transmitted and partly reflected at the delay line/end piece interface (Figure 15). The reflected portion travels directly back to the transducer, while the transmitted portion travels through the end piece and is then reflected back to the transducer. The time interval between the two pulses of ultrasound is given by the relationship $\Delta t = 2d/c$, where d and c are the thickness and ultrasonic velocity in the end piece, respectively. Materials tend to expand or contract when they are heated or cooled, and therefore both d and c change with temperature. Thus, it is usually necessary to prepare a calibration curve of Δt vs. temperature for each ultrasonic thermometer. Ul-

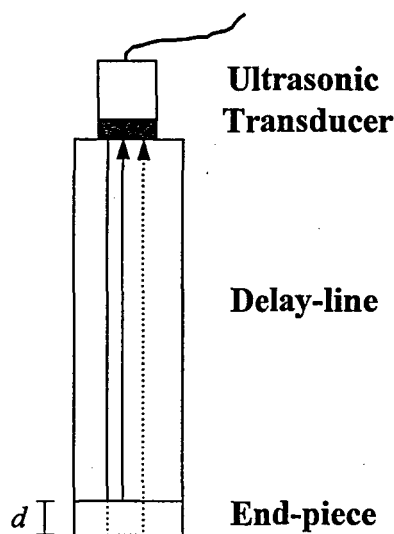


FIGURE 15. An ultrasonic thermometer consists of a cylindrical rod of material bonded to a small cylindrical end piece. The time-of-flight of the pulse in the end piece is related to the temperature.

trasonic thermometers must be carefully designed to avoid interference from sidewall reflections, or overlap of pulses due to reverberations in the end piece. This type of thermometer is useful in situations where it is inconvenient to use conventional temperature sensors (e.g., in a microwave environment, at very high temperatures, or in highly corrosive materials).

D. Flow Rate

The rate at which a food material flows through a pipe is important in many food-processing operations. A number of ultrasonic devices have been developed that measure the speed at which foods flow through pipes (e.g., *doppler*, *transit time*, and *cross-correlation* flow meters).^{36,179} Ultrasonic flow meters are suitable for determining flow rates of up to a few meters per second on systems ranging from a few millimeters (e.g., the flow of blood in a vein) to greater than a kilometer (e.g., the flow of water in rivers or oceans). The design of various types of ultrasonic flow meters has been considered by Lynworth.³⁶ Most of these measure the average flow velocity of a liquid across a pipe; however, some more sophisticated flow meters allow measurements of the flow profile across a pipe. It

should also be noted that many ultrasonic flow meters actually measure the velocity of the inhomogeneities in a liquid rather than the velocity of the liquid itself.

E. Composition

The composition of foods plays an important role in determining their overall quality and cost, and so it is important to have analytical techniques to measure the concentration of the various components in foods. The possibility of using ultrasound to determine the composition of a variety of foods has already been established (Table 1). This application relies on there being a significant change in the ultrasonic properties (e.g., velocity, attenuation, or impedance) of a food as its composition changes. The greater the magnitude of the change, the more accurately the composition can be determined. For example, the ultrasonic velocity of a sugar solution increases by about 4 m/s per 1% increase in sugar concentration at 20°C (Figure 7). It is simple to measure the ultrasonic velocity of a solution to better than 0.4 m/s using existing commercial ultrasonic instruments, and so the sugar concentration can be determined to within 0.1%. This technique has been used successfully to determine the sugar concentration of various fruit juices and drinks (see Section V).

There are a number of possible drawbacks to using ultrasound for measuring the composition of foods. First, many foods are multicomponent systems, and each component contributes to the overall ultrasonic signal in a different way. Thus, information about the component of interest may be obscured by changes in the concentration of one or more of the other components present. In some cases, it is possible to circumnavigate this problem by making measurements as a function of frequency or temperature (because different components have different dependencies on these parameters). Nevertheless, it means that ultrasound often has limited application to complex multicomponent foods. Second, the ultrasonic properties of some foods are sensitive to the structure and interaction of their components, as well as to their concentration. Thus, two foods may have the same composition but different ultra-

sonic properties because of changes in the structure or interactions of the components. For example, the ultrasonic velocity and attenuation coefficient of two emulsions that have the same overall composition but different droplets sizes are very different.²³ This problem can often be overcome by measuring the frequency dependence of the ultrasonic properties of a material, because at certain frequencies the ultrasonic properties are independent of structure, but do depend on composition (e.g., the velocity of ultrasound in an emulsion is independent of droplet size at high frequencies).²³

F. Particle Size

The size of the particles in dispersed systems, such as emulsions, suspensions, and foams, has a pronounced effect on their overall physicochemical properties (e.g., stability, appearance, rheology, taste, and microbiology). It is therefore advantageous for food scientists to have techniques to measure particle size. Conventional methods, such as electron microscopy, light microscopy, and laser diffraction, often require extensive sample preparation or optically transparent systems, and are therefore unsuitable for most food applications. Ultrasound has important advantages over these techniques because no sample preparation is necessary and measurements can be made in systems that are optically opaque. In addition, measurements of particle size can be made rapidly and nondestructively on-line during the processing of foods. Thus ultrasound could be used to continuously monitor the size of particles produced by a homogenizer, colloid mill or mixing unit. This information could then be input into a computer that automatically controls the operation of the processing line, thus improving the efficiency of the process.

Ultrasound can be used to determine particle sizes in emulsions or suspensions in a manner analogous to light scattering.²³ An ultrasonic wave incident upon an ensemble of particles is scattered by an amount that depends on the size and concentration of the particles. The ultrasonic velocity and attenuation coefficient depend on the degree of scattering, and so can be used to provide information about particle size (Figure 5). In

fact, it is possible to determine both the size and concentration of droplets in an emulsion or suspension by measuring the frequency dependence of the ultrasonic velocity and/or attenuation coefficient, and then finding the particle size distribution that gives the best fit between the measured ultrasonic properties and those predicted by theoretical equations that describe the propagation of ultrasound in emulsions (Section II.C.3). For some emulsions,³⁷ there is good agreement between theory and experiment up to concentrations of 30 to 40%. By making ultrasonic measurements over a wide range of frequencies, it is possible to analyze particles as small as $0.01\ \mu\text{m}$ and as large as a few millimeters, which covers most of the range of particles important in foods. As well as the applications of ultrasound shown in Table 1, there are many other food systems where ultrasound would be useful for particle sizing (e.g., mayonnaise, cream liqueurs, soups, sauces, butter, cheese, and margarine).

G. Creaming and Sedimentation Profiles

Ultrasound is finding increasing use in the food industry for monitoring the stability of colloidal dispersions to creaming or sedimentation.

Particles that have a lower density than the surrounding fluid move upward due to gravitational forces ("creaming"), whereas those that have a higher density move downward ("sedimentation"). The gravitationally induced movement of particles affects the appearance and stability of many food systems (e.g., the settling of pulp in orange juices or the creaming of fat globules in milk or salad creams). By measuring the ultrasonic velocity or attenuation coefficient as a function of sample height and time, it is possible to quantify the rate and extent of creaming (Figure 16). This technique can be fully automated, can detect creaming long before it is visible to the eye, and provides information about the detailed creaming profile rather than the movement of a single boundary layer. By measuring the ultrasonic velocity and attenuation coefficient as a function of frequency, it should be possible to determine both the concentration and size of the droplets as a function of sample height. Thus, a detailed analysis of creaming and sedimentation in complex food systems can be monitored noninvasively. Thus, ultrasound offers food scientists an extremely powerful technique for studying the factors that affect the shelf-life stability of many foods.

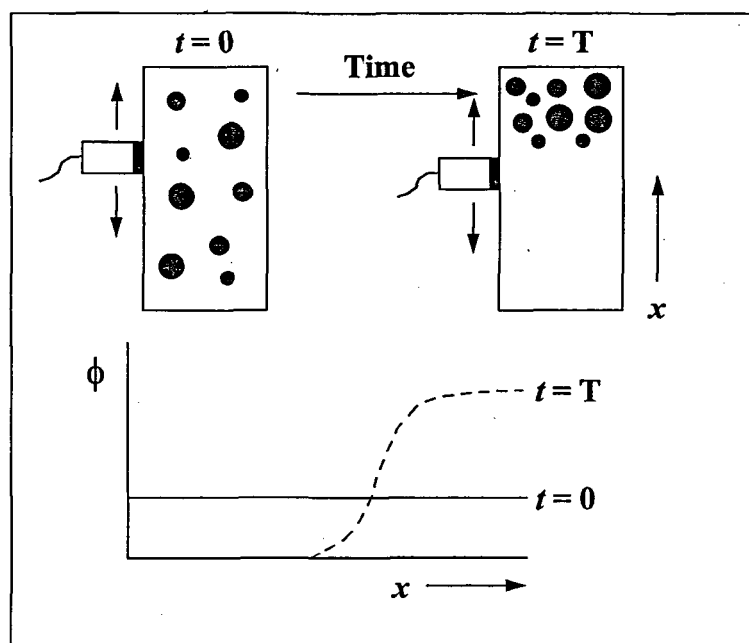


FIGURE 16. Detailed analysis of the creaming or sedimentation of particles in emulsions and suspensions can be determined by measuring the time dependence of the velocity of ultrasound at different heights within a sample.

H. Phase Transitions

Many foods contain components that undergo some form of phase transition during manufacture, storage, or consumption (e.g., melting or crystallization of sugars, oils, or water). The ultrasonic properties of a material change significantly when it melts or crystallizes, and so ultrasound can be used to monitor phase transitions. One of the most commonly used methods is to measure changes in the ultrasonic velocity with time or temperature. The ultrasonic velocity of solids is significantly greater than that of liquids, and so the ultrasonic velocity of a sample increases when a component crystallizes and decreases when it melts (Figure 17). Ultrasonic velocity measurements have been used to monitor crystallization and melting behavior in a variety of bulk and emulsified fats (e.g., margarine, butter, meat, and shortenings) (Table 1).

I. Gelation

The ultrasonic velocity of a material depends on its elastic modulus and density (Equation 2). Ultrasonic velocity measurements are therefore

sensitive to any change in a material that alters these properties. The ultrasonic velocity of a solid depends on its shear modulus, $c = (K + 4G/3)/\rho$, and should increase when a biopolymer solution gels. The shear modulus of food gels is usually less than 1000 N/m^2 , which is about six orders of magnitude smaller than their bulk modulus (typically $2 \times 10^9 \text{ N/m}^2$). For this reason, there is only a small change in the ultrasonic velocity for a sol-gel transition, and ultrasonic velocity measurements are relatively insensitive to gelation. This explains why there have been few studies of gelation using ultrasonic velocity measurements. Nevertheless, ultrasonic velocity measurements could still be used to determine the concentration or hydration of biopolymers in gels or to characterize the properties of emulsion droplets suspended in filled gels. The author believes that measurements of the frequency dependence of the ultrasonic attenuation coefficient may be much more sensitive to gelation because of changes in the viscosity and molecular relaxation of the system (Equations 7 and 9). Indeed, recent experiments have shown that ultrasonic spectroscopy can provide useful insights into the initial stages of gelation of polysaccharides.¹⁸⁰ In addition,

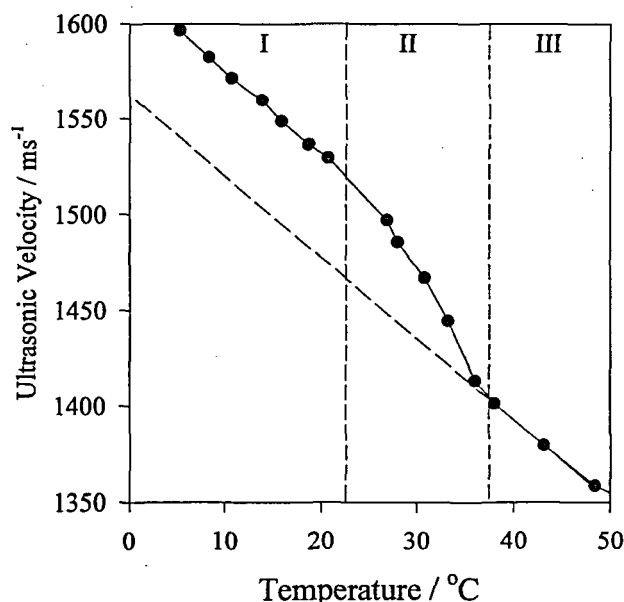


FIGURE 17. The velocity of ultrasound decreases when a fat melts, and so ultrasound can be used to determine the solid fat content of a material.

measurements of the attenuation coefficient may prove useful for characterizing particulate gels (such as those formed from egg white or whey proteins) because of the scattering of ultrasound by the particles.

J. Ultrasonic Imaging

Ultrasound is routinely used in medicine and materials research to provide images of the internal structure of materials.^{32,33} The same techniques can also be applied in food science. Indeed, imaging techniques have already been used to grade the quality of live animals and carcasses in the meat industry, by providing information about the dimensions and location of fat and lean tissue. Other areas where ultrasound imaging would be useful are the monitoring of creaming/sedimentation processes in emulsions and suspensions, detection of extraneous matter, and measurement of crystallization profiles in foods. Ultrasonic imaging techniques can now be purchased at relatively low cost (<\$30,000) and will almost certainly see greater use in the food industry in the near future.

K. Molecular Properties

At present, few food scientists realize that ultrasound is a powerful analytical method for providing information about the fundamental properties of molecules in solution. The two techniques most frequently used are ultrasonic velocimetry and ultrasonic spectroscopy.

1. Ultrasonic Velocimetry

Information about the volume, hydration, and adiabatic compressibility of solute molecules are determined from accurate measurements of the ultrasonic velocity and density of solutions as a function of solute concentration.⁴⁰⁻⁴⁴ The adiabatic compressibility of a solution is determined from its density and velocity: $\kappa = 1/\rho c^2$. A solute molecule is characterized in terms of density $[\rho]$ and compressibility $[\kappa]$ concentrational increments:

$$[\rho] = \frac{\rho - \rho_0}{C\rho_0} \quad (17)$$

$$[\kappa] = \frac{\kappa - \kappa_0}{C\kappa_0} \quad (18)$$

Here, ρ_0 and κ_0 refer to the properties of the pure solvent, and ρ and κ to the properties of the solution. The values of $[\rho]$ and $[\kappa]$ are determined by linear regression analysis of accurate density and compressibility measurements vs. concentration.^{40,41} Concentrational increments depend on:

1. Differences in the physical properties of the solute and water (e.g., proteins are more dense and less compressible than water)
2. Differences between the hydrated water associated with the protein and the bulk water. Hydrated water is more dense and less compressible than bulk water.

Ultrasonic measurements can therefore be used to provide valuable information about the compressibility and hydration of solutes in aqueous solution.⁴²⁻⁴⁴ This technique has shown that random coil proteins have more water associated per gram than globular proteins, and that the intrinsic compressibility of globular proteins depends on the packing of the amino acids in the interior of the protein molecule.⁴⁴ Ultrasound is particularly useful for studying changes in the physicochemical properties of proteins in solution when environmental conditions such as pH, ionic strength, and temperature are altered.⁴⁰⁻⁴⁴ It has also proven useful for studying the properties of small solute molecules, such as amino acids,^{45,46} sugars,^{47,48} and salts,^{49,50} and other large solute molecules, such as DNA and polysaccharides.^{51,52}

2. Ultrasonic Spectroscopy

Ultrasonic *spectroscopy* uses measurements of the ultrasonic velocity and/or attenuation coefficient as a function of frequency to obtain information about relaxation mechanisms.⁵²⁻⁵⁴ Measurements of the frequency dependence of the absorption of ultrasound have been used to study fast chemical reactions and structural rearrangements of biopolymer molecules in solution.⁵³ The

frequencies employed in ultrasonic experiments allow physicochemical equilibria with relaxation times between about 10 ns to 10 ms to be studied. A number of important relaxation phenomena occur in this range and can therefore be studied using ultrasound. The three most important equilibria for biopolymer solutions are

1. Proton transfer equilibria: $\text{R-NH}_3^+ \rightleftharpoons \text{R-NH}_2 + \text{H}_2\text{O}$, and $\text{R-COOH} \rightleftharpoons \text{R-COO}^- + \text{H}^+$
2. Hydration equilibria: $[\text{water}]_{\text{bound}} \rightleftharpoons [\text{water}]_{\text{unbound}}$
3. Conformational equilibria: $\text{helix} \rightleftharpoons \text{random coil}$

For example, there is an increase in the ultrasonic absorption coefficient at pH values corresponding to protonation of the acid and base groups.⁵⁵

Ultrasound can also be used to characterize the aggregation of biopolymers. Individual protein molecules are too small to scatter ultrasound at the frequencies used in most experiments. However, if they aggregate to form large enough particles, ultrasound can be used to study their structure. Ultrasonic absorption measurements as a function of frequency have been used to determine the size of casein micelles,⁵⁶ to detect the isoelectric precipitation of legumin (a globular protein from broad bean),^{40,41} and to study various milk proteins in solution.⁵⁷ The attenuation of ultrasound increases when proteins aggregate because ultrasound is scattered and because of the change in the viscosity and molecular relaxation.

V. APPLICATION TO SPECIFIC FOOD-STUFFS

A. Aqueous Solutions and Gels

A wide variety of foods exist either partly or wholly as solutions, or have been solutions sometime during their production. Fruit juices and drinks are principally solutions of sugars in water. Brine is an aqueous solution of salts. The aqueous phase of milk is a complex solution containing dissolved lactose, salts, and proteins, as well as colloidal protein. A number of experimental techniques have been developed to characterize the

physical properties of solutions, including density measurements, refractive index, polarimetry, NMR, and infrared.⁵⁸ Instruments based on ultrasound can also be used to provide valuable information about solute properties, such as concentration, structure, and interactions. The use of ultrasound for characterizing the molecular properties of solutes in solution was described in Section IV.K. In this section, applications of ultrasound for the determination of solute concentration in some real foods are reviewed.

It is simple to use ultrasonic velocity measurements to determine the composition of two component solutions. Earlier it was mentioned that ultrasonic velocity measurements could be used to determine the sugar concentration of fruit juices and drinks (Section IV.E, Figure 7). Contreras et al.⁵⁹ compared the sugar content determined using an ultrasonic velocity method with that determined using traditional methods such as enzyme analysis, refractive index, and density measurements, and found excellent agreement between the ultrasonic method and the other methods. Despite the simplicity of the ultrasonic technique, there are a number of factors that have to be considered to make accurate measurements. First, the increase in ultrasonic velocity per 1% increase in sugar concentration depends on the type of sugar molecules present,⁴⁸ as well as their concentration: fructose > glucose > sucrose (Figure 7). The type of sugars present varies quite widely from one food to another, and so for accurate ultrasonic measurements of sugar concentration it is necessary to prepare calibration curves for each type of food analyzed. This would not be a problem for a factory that wanted to determine the total sugar content of a product that was being produced routinely (e.g., orange juice), but it does mean that ultrasound has a limited application for accurately determining the sugar concentration of solutions of unknown composition. Nevertheless, it may be possible to use the ultrasonic technique in combination with other techniques, such as density or refractive index measurements, to obtain information about both the concentration and type of sugars present. Another factor that may limit the use of ultrasound is that ultrasonic measurements are particularly sensitive to temperature,⁴⁸ and therefore it is necessary to either control the temperature or to measure and correct for

it (to within about 0.1°C). For these reasons, there are few advantages of using ultrasound, rather than one of the existing techniques, for determining sugar concentrations in the laboratory. Nevertheless, it does have important advantages for on-line applications because measurements are rapid, precise, relatively low cost, nondestructive, and noninvasive.

Ultrasonic determination of the sugar contents of various foods have been described in the literature: measurement of dilution of drinks, sauces, and syrups,^{48,60,61} monitoring of solute concentrations during evaporation processes and estimation of sugar contents of alcoholic beverages,^{48,62–64} and determination of sugar concentrations in sucrose solutions.⁶⁵ The concentration of other simple solutes, such as salts^{49,50} and amino acids,^{45,46} can also be determined using ultrasonic velocity measurements. Ultrasound can also be used to determine the concentration of biopolymers in solution, such as proteins and carbohydrates. For many aqueous biopolymer solutions and gels, there is a linear increase in the ultrasonic velocity and attenuation coefficient with solute concentration up to fairly high concentrations (often >10 wt%). For example, the ultrasonic velocity of many food proteins increases by about 2.7 m/s per 1% increase in protein concentration.^{42–44} The velocity of ultrasound can easily be measured to 0.4 m/s using existing commercial instrumentation, and so it is possible to determine protein concentrations to 0.2% once a suitable calibration curve has been established for the protein of interest. Ultrasound has advantages over optical techniques because the concentration of biopolymers in concentrated or opaque systems can be determined rapidly and nondestructively.

B. Liquid Mixtures

There are many examples of liquid mixtures in the food industry: oils consist of a mixture of liquid triglycerides, and alcoholic drinks are mixtures of water and alcohol. The ultrasonic properties of liquid mixtures depend on their composition, and so ultrasound can be used to measure this property. Simple theoretical equations (Equations 6 to 9) can be used to relate the ultrasonic

velocity of ideal and nearly ideal mixtures to their composition. For example, the triglyceride composition of oils can be related to their ultrasonic velocity using this approach.^{66,67} Empirical equations can be established experimentally to relate the ultrasonic properties of nonideal mixtures to their composition. This approach has been used to determine the alcohol content of many alcoholic beverages.^{62–64} In these systems, it is possible to determine both the alcohol and sugar contents by measuring the ultrasonic properties at two different temperatures.

C. Edible Fats and Oils

Fats and oils play a major role in the human diet because they provide an important source of energy and because of their characteristic heat transfer, organoleptic, and rheological properties.⁶⁸ By definition, a fat is solid (or semisolid) at the temperature of interest, whereas an oil is liquid. Triglycerides are the major constituents of edible fats and oils and largely determine their overall bulk properties, such as viscosity, density, and melting characteristics. The suitability of an oil or fat for a particular food application depends on the type and proportion of triglycerides it contains.

The variation of ultrasonic velocity with temperature for a fatty material can conveniently be divided into three regions (Figure 17). At low temperatures, all the triglycerides are completely solid, and the decrease in velocity with increasing temperature is due solely to the negative temperature coefficient of the ultrasonic velocity of the solid fat (region I). As the fat is heated, some of the triglycerides melt, and the velocity decreases more rapidly because liquid oil has a lower ultrasonic velocity than solid fat. This effect is superimposed on that due to the temperature coefficients of velocity of the separate phases (region II). When a temperature is reached where all the triglycerides have melted, the decrease in velocity with increasing temperature is due to the negative temperature coefficient of the ultrasonic velocity of the liquid oil (region III). The range of temperatures over which region II extends depends on the melting points of the various triglycerides present, and is extremely im-

portant in many applications of fats in the food industry.

1. Ultrasonic Propagation in Solid Fats (Region I)

It is difficult to measure the ultrasonic properties of pure solid triglycerides because of their tendency to form voids on cooling.⁶⁹ These voids are strong scatterers of ultrasound and can often attenuate a signal so much that transmission measurements are practically impossible.⁷⁰ Even when velocity and attenuation measurements can be carried out, it is still difficult to obtain information about the properties of the individual fat crystals because of the interfering effect of the voids. It may, however, be possible to use ultrasound to characterize the size and concentration of voids in solid fats by measuring the frequency dependence of the velocity and attenuation coefficient. Voids do not tend to form so readily in naturally occurring animal and vegetable fats because they contain mixtures of many different triglycerides, which presumably allows the molecules to pack into the crystals without forming voids.⁶⁹ Thus, it is possible to make ultrasonic measurements in these systems. Miles et al.⁷¹ have measured the velocity of ultrasound as a function of temperature in a number of predominantly solid animal fats. They found that the ultrasonic velocity of the solid fats ranged from about 2000 to 2070 m/s at 31°C and decreased by about 5 m/s per °C increase in temperature.

2. Ultrasonic Propagation in Fat/Oil Mixtures (Region II)

Many of the physical properties of fatty materials that are of commercial importance, such as texture, spreadability, and consistency, depend on the ratio of solid to liquid fat over a particular range of temperatures.⁷² For this reason, it is important to have techniques that can be used to measure the *solid fat content* (SFC). The velocity of ultrasound in solid fat is greater than that in liquid oil, and so a measurement of the ultrasonic velocity in a fat/oil mixture can be used to determine the SFC. Ultrasound has certain advantages

over existing techniques used for this purpose (e.g., NMR, dilatometry, and DSC) because it is capable of rapid and precise measurements in samples with low SFC, it is relatively inexpensive, and it can easily be adapted for on-line measurements.^{70,73-77} The following relationship has been derived to relate the ultrasonic velocity of a fatty material to its SFC:⁷¹

$$\text{SFC} = \frac{\frac{1}{c^2} - \frac{1}{c_L^2}}{\frac{1}{c_s^2} - \frac{1}{c_L^2}} \quad (19)$$

Here, SFC is the fraction of total fat that is solid, c is the ultrasonic velocity of the fat/oil mixture and c_L and c_s are the velocities in the material if it were completely liquid or completely solid, respectively. These values are obtained by extrapolating measurements from higher or lower temperatures. This equation assumes that the effects of scattering and absorption of ultrasound are negligible, and that the density of the solid fat and liquid oil phases are not appreciably different. A similar equation has also been derived to predict the ultrasonic velocity of three-phase systems that contain fat, oil, and water, such as margarines or butter.⁷⁰ These equations give SFC predictions that are in good agreement with other methods,⁷⁰ such as NMR. The attenuation of ultrasound in liquid oil and solid fat are often appreciably different, and so attenuation measurements can also be used to determine the SFC of fat/oil mixtures. However, velocity measurements are usually preferred because they can be measured more accurately and are less sensitive to the structure of the fat crystals.

A complicating factor that has to be considered when using ultrasound to determine SFC is the influence of phase equilibria on velocity and attenuation measurements.⁷⁸ When a solid and liquid phase are in equilibrium, the pressure and temperature fluctuations associated with an ultrasonic wave can cause a molecule to flip between the solid and liquid phases: solid \rightleftharpoons liquid. Thus, energy that was stored in the ultrasonic wave is converted to heat, which can lead to a huge increase in the ultrasonic attenuation and a corresponding velocity dispersion.⁷⁸ This effect can

make it difficult to determine the SFC of systems where a significant proportion of the material is in phase equilibria. Nevertheless, frequency-dependent measurements of the velocity and attenuation may prove to be a very valuable means of investigating the kinetics of phase equilibria.^{79,80}

3. Ultrasonic Propagation in Liquid Oils (Region III)

The velocity and attenuation of ultrasound have been measured in a variety of animal, vegetable, and marine oils as a function of temperature and ultrasonic frequency.^{66,67,81-84} The velocity increases with decreasing temperature (ca. -3.3 m/s/°C) and increasing frequency, while the attenuation coefficient increases with decreasing temperature and increasing frequency. Different liquid triglycerides have different ultrasonic velocities that can be related to their chemical structure.^{66,67,85-87} Empirical relationships have been derived to relate the ultrasonic velocities of oils to the chain length and degree of unsaturation of the triglyceride molecules they contain.^{66,67} Thus, ultrasonic velocity measurements can be used for estimating oil composition or determining the adulteration of oils,⁸⁸⁻⁹⁰ particularly when used in conjunction with other techniques, such as density measurements or refractive index.^{86,87}

Measurements of the ultrasonic attenuation as a function of frequency can be used to provide information about relaxation mechanisms that occur in liquid triglycerides. These measurements can often be related to dynamical rheological quantities, such as the shear and bulk viscosity of the system.⁹¹⁻⁹³

D. Food Emulsions and Suspensions

Emulsions and suspensions are particularly common in the food industry⁹⁴ (e.g., milk, salad cream, margarine, mayonnaise, cream liqueurs, soups, desserts, etc.). The bulk properties of these systems are largely influenced by the concentration and size of the particles that they contain.⁹⁴ It is therefore important for food scientists to have analytical techniques to characterize the proper-

ties of suspended particles. Ultrasound can be used to measure disperse-phase volume fraction, particle size, creaming/sedimentation profiles, and phase transitions, as well as particle interactions and phase inversion.²³ It has advantages over many existing techniques because it can be applied to systems that are optically opaque, concentrated, and electrically nonconducting.

The possibility of using ultrasound for characterizing emulsions and suspensions has been clearly demonstrated for a variety of nonfood emulsions and suspensions.⁹⁵⁻¹⁰⁰ Recently, a number of workers have also highlighted its potential for characterizing real food systems. Ultrasonic velocity and attenuation measurements have been used to determine disperse-phase volume fractions in sunflower oil-in-water emulsions,³⁷ salad creams,¹⁰¹ and milk.⁵⁷ Droplet size has been measured ultrasonically for casein micelles,⁵⁶ sunflower oil-in-water emulsions,³⁷ salad creams,¹⁰¹ and milk fat globules.⁵⁷ Ultrasound has been used to monitor the sedimentation of colloidal particles in orange juices¹⁰² and the creaming of oil droplets in emulsions.^{13,100} Given the obvious advantages of ultrasound for rapid on-line measurements, the technique will certainly find increasing application in the food industry in the future.

E. Aerated Food Products

Many foods contains undissolved air in the form of bubbles or cells that range in size from a few micrometers to a few millimeters (e.g., fruit, bread, whipped cream, meringue, and beer foam).^{103,104} Physical properties of these foods that are of commercial importance depend ultimately on the size, concentration, and interactions of the air cells. It is notoriously difficult to characterize the properties of systems that contain gas cells because of their complex structure, physical instability, and delicate nature. Traditional techniques such as light scattering, microscopy, and density measurements are often unreliable, time consuming, and laborious to use, as well as being unsuitable for on-line applications. Ultrasound is extremely sensitive to the presence of gas bubbles: even a fraction of a percent of undissolved gas can cause huge changes in the ultra-

sonic properties of a material (Figure 6). It should therefore be a very useful tool for providing information about the size and concentration of undissolved gas in foods.

Despite its potential, there are few examples of ultrasound being used to characterize aerated food materials. This is because the attenuation of ultrasound caused by resonant scattering (Section II.C.3) is so large that it is practically impossible to transmit ultrasound through aerated materials at the frequencies normally used in ultrasonic analysis (0.1 to 100 MHz). To overcome this problem, the experimental conditions can be altered to increase the magnitude of the signal from a sample, either by lowering the frequency or increasing the power levels used. Nevertheless, both of these alternatives have their own difficulties (see Section V.G). A more elegant method of overcoming the problem of very high attenuation by aerated products is to use a *reflection* method, rather than a transmission method. In this method, the fraction of a pulse of ultrasound *reflected* from the surface of a material is measured. The amplitude of this pulse is related to the size and concentration of the undissolved gas bubbles, and so can be used to provide information about these properties. This technique has already been used to determine the air content of meringue and marshmallow.¹⁰³ Fairley and co-workers have also demonstrated that by measuring the frequency dependence of the amplitude of a reflected pulse, it is possible to determine both the concentration and size of bubbles in whipped cream and yogurts.^{104,105} The relationship between the ultrasonic properties of a concentrated bubbly liquid or foam and its composition and microstructure is complicated because of multiple scattering of the waves and interactions between bubbles.¹⁰⁵ At present, there are no theories available that satisfactorily describe ultrasonic propagation in these systems, and so the data has to be interpreted in a semiempirical fashion. Another drawback of the ultrasonic reflection technique is that the properties of the material at the surface may not be representative of those in the center. Nevertheless, the technique has considerable potential, particularly for use as an on-line sensor for monitoring the properties of aerated foods during processing.

F. Meat and Fish

In the western world, consumers prefer meat that is lean, consistent, flavorful, and tender.¹⁰⁶ To achieve these aims, the meat industry has developed various objective methods to assess the quality and yield of live animals and carcasses. Sophisticated instrumental methods, originally developed for use in the medical field, such as NMR, ultrasonic, and X-ray imaging, have been applied to meat. Nevertheless, the instrumentation is usually too expensive, delicate, and cumbersome for routine testing in the meat industry.¹⁰⁶ For this reason, a number of simpler instruments have been developed that operate on principles similar to these sophisticated instruments but are much cheaper and more robust. One of the most promising of these techniques is ultrasound.¹⁰⁶ In fact, measurement of the composition and back-fat thickness of meat from live animals and carcasses (e.g., fish, cattle, pigs, sheep, and poultry) has been the most popular application of ultrasound in the food industry for the past 30 years or so. There are over 150 papers listed on this subject in the *Food Science and Technology Abstracts* (1969 to 1995). In contrast to most other applications of ultrasound in the food industry, which have never gotten further than use in the laboratory, there are many commercially available ultrasonic instruments for characterizing meat.^{107,108} These instruments have proven to be very valuable in the selection and grading of live animals and carcasses because measurements are rapid, reproducible, and objective.¹⁰⁶

Ultrasound can be used in a variety of different ways to obtain information about the quality of meat. The simplest is to measure the velocity at which an ultrasonic pulse travels through a representative portion of a live animal or carcass.¹⁰⁹ The ultrasonic velocity of lean meat and fat are significantly different, and so it is possible to deduce the fat content from a velocity measurement once a suitable calibration has been carried out.¹⁰⁹ The back-fat thickness is determined by measuring the time-of-flight t of a pulse that has been reflected from the interface between the fat and lean tissue: the longer the t , the thicker the tissue.^{107,110,111} Simple imaging techniques can be used to measure marbling or the area of specific tissues¹¹²⁻¹¹⁵ (e.g., rib-eye area).

The use of ultrasound to characterize meat relies on there being a good understanding of the relationship between the measurable ultrasonic properties (c , α , and Z) and the composition, structure, and physical state of the tissue. For this reason, a number of workers have investigated the various factors that influence the propagation of ultrasound in animal tissues. The dependence of the ultrasonic properties on the physical state of fat and water in meat has been investigated.^{71,116} Measurements of the velocity and attenuation of ultrasound in intact, ground, and homogenized meat samples suggest that muscle structure only has a slight influence on the ultrasonic properties and that the predominant factor is composition.^{117,118} The interpretation of ultrasonic measurements on animal tissue has recently been advanced by the implementation of various signal-processing techniques, such as Fourier transform analysis,¹¹⁹ pattern recognition,¹¹² neural networks,^{112,120} and texture analysis.¹¹⁴

G. Fruits and Vegetables

To use ultrasound to characterize fruits and vegetables, it is necessary to relate some property that is of importance to the food scientist (e.g., ripeness) to some measurable ultrasonic parameter (e.g., velocity, attenuation, or impedance). The property that most influences the ultrasonic properties of fruits and vegetables is the presence of intercellular air spaces that exhibit resonant behavior over a wide range of ultrasonic frequencies. This explains why many fruits and vegetables have very large attenuation coefficients and why their ultrasonic velocities are often lower than that of air (e.g., apples, avocado, banana, carrot, cucumber, melon, papaya, potato, and pumpkin).¹²¹⁻¹²⁷ Ultrasound can be so highly attenuated that it is very difficult to transmit it through fruits and vegetables at the frequencies normally used in the ultrasonic testing of foods (0.1 to 100 MHz). A number of workers have tried to overcome this problem by using higher ultrasonic intensities, lower frequencies (<100 kHz), and small slices of fruit rather than whole fruit, but each of these solutions has its own problems. High-intensity ultrasound can damage the plant tissue.¹²⁸ Low-frequency measurements are

inaccurate due to diffraction, wave-guiding effects, and poor signal resolution.¹²⁹ Obviously, it would be more advantageous to have measurements on whole fruits or vegetables rather than on excised samples. Another problem that is often encountered with fruits and vegetables is the natural variability of their physical properties. This means that a large number of measurements are needed to obtain statistically significant results. Sometimes, natural variations may be so large that they obscure changes in the ultrasonic properties due to some phenomena of interest¹³⁰ (e.g., maturity). Even so, a number of workers have demonstrated that ultrasonic transmission measurements can be used to follow the ripening and quality of apples,¹³¹ melons and avocados,^{130,132} and to detect physiological defects of potatoes.¹³³

A promising solution to the problem of high attenuation in fruits and vegetables is to make use of the reflection technique mentioned in a previous section. The amplitude of the signal reflected from a plant material is related to the size, shape, and concentration of the intercellular air spaces, and so in principle can be used to provide information about these properties. Nevertheless, interpretation of data is often difficult because the complex nature of biological materials means that many factors can influence the measurements. In addition, the properties of the surface of the plant tissue may not be representative of the overall properties, and surface irregularities may make interpretation of reflection measurements more complex. As our understanding of how ultrasound interacts with biological materials improves, ultrasound should become a powerful tool for characterizing these systems.

H. Dairy Products

Milk is a complex material, consisting of a dilute emulsion of fat globules suspended in an aqueous phase that contains colloidal protein, as well as dissolved protein, salts, and lactose.⁹⁴ Its composition is variable and depends on factors such as the type of animal, its health and age, the stage of lactation, its diet, the climate, and the season. Characterization of the physical properties of milk and its products (e.g., butter, cheese,

yogurt, and cream) is important for the design and operation of dairy equipment and the determination of product quality. Ultrasound has proved to be particularly useful for this purpose.

The most popular application of low-intensity ultrasound in the dairy industry has been the determination of the composition of dairy products, that is, the concentration of fat globules, solids-nonfat (SNF), and total solids (TS = fat + SNF). Empirical relationships have been established between ultrasonic properties and the composition of many dairy products.^{134,135} The velocity of ultrasound increases with temperature in water (up to about 73°C) but decreases in fat. At about 13 to 14°C, the velocity of liquid milk fat and water are approximately the same. Any increase in the velocity above this value is therefore due to the SNF (assuming no scattering effects), and so the SNF can be determined. By making simultaneous attenuation measurements or by measuring the velocity at a higher temperature where the fat and water have very different velocities, it is also possible to determine the fat content. This technique can give very precise measurements of the composition of many dairy products.¹³⁴

The velocity and attenuation of dairy products depend on their microstructure as well as their composition. The effects of the size of milk fat droplets⁵⁷ and casein micelles⁵⁶ on the attenuation of ultrasound in milk have been investigated. Ultrasonic attenuation measurements have also been used to nondestructively monitor the coagulation of milk due to the action of chymosin.^{136–138} The attenuation coefficient is measured as a function of time after the enzyme is added. When the milk coagulates, there is a notable decrease in the attenuation coefficient. A number of workers have investigated the possibility of using ultrasound to nondestructively determine the physical properties of packaged milk.^{139–143} It was found that ultrasonics could be used when milk is packaged in plastic containers but not when in paper cartons because of the difficulty of transmitting ultrasound through the highly attenuating paper.¹⁴⁰ Recently, ultrasonic imaging^{141,142} and acoustic streaming¹⁴³ techniques have been developed to monitor the growth of microorganisms in packaged milk. Ultrasonic techniques have also been used to monitor the properties of

ice-cream mix.¹⁴⁴ Although ultrasound has not so far been used to monitor the crystallization of fat globules in milk, it has been demonstrated that it can be used for this purpose in hydrocarbon oil-in-water emulsions.^{145–147} Ultrasound may therefore offer a useful nondestructive method of monitoring fat crystallization in milk and creams.

Ultrasound has also been used for the characterization of various cheeses. Ultrasound has been used to monitor ripeness in cheeses and to detect structural defects or void formation.¹⁴⁸ Measurements of the time-of-flight of ultrasonic echoes reflected from discontinuities in the cheese can be used to detect flaws or air pockets. As a cheese matures its composition and elastic properties change, and these influence its ultrasonic properties (i.e., ultrasonic velocity and attenuation coefficient). A recent study found a good correlation between the ultrasonic and rheological properties of cheeses.¹⁴⁹

Ultrasound has also been used to obtain information about the structure of cows' teats and the flow rate of milk from teats.^{150,151} This information can be used to quantify the responses of cows to physiological changes, such as climate, season, age, state of lactation, etc. Photoacoustic techniques have been used to determine protein content and Maillard reaction products of milk, yogurt, and cheese in ultraviolet and visible light.¹⁵² The possibility of using ultrasonic reflection measurements to estimate size and concentration of bubbles in whipped creams and yogurts has recently been investigated.^{105,153}

I. Eggs

A number of workers have investigated the possibility of using ultrasound to evaluate the quality of eggs. Velocity measurements have been used to determine egg shell thickness, and good agreement was found with conventional methods.^{154,155} A number of workers have tried to relate the ultrasonic properties of whole eggs, egg yolk, and egg white to their composition and structure.^{156,157} The difference in ultrasonic velocity and attenuation between egg yolk and egg white can be attributed to the fact that egg yolk is basically an oil-in-water emulsion containing 16% protein, while egg white is a concentrated aque-

ous protein solution (about 10% protein). Although the ultrasonic properties of eggs are sensitive to composition, it is difficult to use this for estimating their quality. This is because differences in the ultrasonic properties of eggs due to changes in quality are obscured by their natural variability. For this reason, and because of the difficulty in automating the measurement procedure for whole eggs, no commercial ultrasonic instruments have been developed for characterizing eggs.

J. Acoustic Emission

There are an increasing number of applications of acoustic emission in the food industry. One interesting area is to characterize the sounds produced by foods during mastication. These sounds play an important role in an individual's perception of the quality of a food and may also be useful for quantifying the physical properties of food materials.^{158,159} Originally, characterization of the sound produced during the mastication of foods was carried out subjectively; a number of people were asked to listen to a tape recording of a food being eaten and to identify and quantify the various sounds produced (e.g., crispness, crunchiness, and hardness).¹⁶⁰⁻¹⁶³ These tests are often unsatisfactory because of the subjective nature of the responses from different individuals and because they are time consuming and labor intensive.¹⁶⁴ Recently, more emphasis has been given to objective tests where an acoustic receiver is used to detect the sounds and the received signal is then analyzed by Fourier transform.^{164,165} The amplitude-frequency responses obtained from these tests can often be related to the physical mechanisms that generate the sound and the perceived acoustical attributes of the food (e.g., crispness and crunchiness). Innovative signal processing techniques, such as analyzing the power spectrum of the Fourier transform or the apparent fractal dimension of the jaggedness of acoustic signals on crushing, have recently been developed to characterize the crispiness and crunchiness of cereal foods.¹⁶⁶⁻¹⁶⁸ These techniques have been used to monitor the effect of water sorption on the loss of crunchiness and crispiness of cereal foods such as cheese balls and croutons.^{166,167} The

use of these objective approaches is advantageous for the food industry because they are less reliant on individual preferences and are usually quicker, simpler, and cheaper to carry out.

Acoustic emission has also been used to characterize foods flowing along conveyor belts or in pipes. A technique has been developed to measure the moisture content of flowing grain.¹⁶⁹⁻¹⁷¹ The amplitude of the signal produced by grain as it moves along a conveyor belt is increased as the moisture content decreases. For reliable measurements, it is important to take into account factors such as the flow rate and the distance between the receiver and the grain. In a recent study,¹⁷² it was shown that acoustic emission can be used to continuously monitor the efficiency of machines in a tomato-processing plant by measuring the sound they produce. It was found that subtle shifts in the acoustic signal could be used to rapidly identify any mechanical problems. Acoustic emission is in its infancy in the food industry. Its success depends on the ability of workers to establish relationships between the amplitude-frequency response of the acoustic emissions produced by foods and the mechanisms generating them. Potentially, acoustic emission could provide a very useful, low-cost tool for monitoring the properties of foods and the performance of machinery during production.¹⁷³

K. Miscellaneous

Ultrasound has been used to determine the crispness of biscuits.¹⁷⁴ The ultrasonic velocity was measured by transmitting a pulse of ultrasound across a biscuit. A good correlation between the ultrasonic velocity and the elastic modulus determined using an Instron Universal Testing Machine was observed. The ultrasonic technique was capable of more rapid measurements and is nondestructive. Juodiekene et al. used absorption measurements to determine the texture of wafer sheets by measuring the amplitude of an ultrasonic wave transmitted through them.¹⁷⁵ McMaster et al. used velocity measurements to monitor the properties of food materials during extrusion.¹⁷⁶ They established correlations between the ultrasonic properties and the flow rate, composition,

and pressures used. Povey and Rosenthal used ultrasonic attenuation measurements to monitor the breakdown of starch by α -amylase.¹⁷⁷ Sarker and Wolfe used measurements of the amplitude of an ultrasonic pulse reflected from the surface of various fruits and vegetables to detect smoothness, cracks, and defects.¹⁰² An ultrasonic device has been developed to measure the fouling of pipes in UHT processing plants.¹⁷⁸ The time-of-flight of an ultrasonic pulse in the fouling film is directly proportional to the thickness of the film. This device can measure film thicknesses accurately on films between 0.5 and 6 mm and can detect the presence of films as thin as 0.1 mm.

VI. ADVANTAGES AND LIMITATIONS

The main advantages of ultrasound are that it is rapid, nondestructive, noninvasive, and can be applied to systems that are concentrated and optically opaque. In these respects, ultrasound is similar to the NMR techniques recently developed to characterize foods.⁷⁻⁹ Nevertheless, ultrasound has the additional advantages that it is relatively inexpensive and that it can easily be adapted for on-line measurements.

One of the major disadvantages of the ultrasonic technique is that the presence of small gas bubbles can attenuate ultrasound so greatly that a signal cannot be detected. As mentioned earlier, this problem can be overcome by using reflection measurements rather than transmission measurements; however, the signal from the bubbles may obscure that from other components. Alternatively, it is often possible to remove the bubbles prior to analysis by drawing a vacuum or putting the sample under pressure. Another problem is that a lot of information about the thermophysical properties of the component phases (e.g., densities, compressibilities, heat capacities, and thermal conductivities) is often needed to interpret ultrasonic measurements using mathematical theories. This means ultrasound may have limited application to systems that contain components with unknown properties. Nevertheless, if the same system is being tested routinely, this is not usually a problem.

VII. CONCLUSIONS

Ultrasound has already been used to nondestructively test a wide variety of different food materials. These applications have led to a fairly good understanding of the factors that affect ultrasonic propagation in these systems. Nevertheless, the complexity and diversity of food materials means that much more basic research is still needed. The benefits that the food industry can gain from the further development and application of ultrasound to foods are substantial. On-line ultrasonic sensors give manufacturers better control over the composition of their product during processing, which leads to improved product quality and reduced manufacturing costs. In the laboratory, ultrasound can be used to provide valuable information about fundamental physical and chemical properties of food materials that are difficult to obtain using other techniques. The author therefore believes that the application of ultrasound to food materials will continue to grow and that many more useful applications will be developed in the near future.

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