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Exploration of an ultrasonic pulse echo system for comparison of milks, creams, and their dilutions

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ABSTRACT

Rapid measurements are valuable in food authentication. As a first step towards application of sound for fast food authentication, the influence of fat content and dilution of liquid milk/cream products on their ultrasound characteristics was examined. A total of 18 liquid dairy products, ranging in fat content from 0 to 60 g/100 g product, including nine cream samples and nine milk samples, were diluted to four levels, which resulted in 90 samples. Ultrasonic velocity, together with physical properties including viscosity, density, and macronutrient composition of the tested dairy products, was determined. The results show that the ultrasonic velocity in the dairy products increased with a decreasing fat content of the samples. There was a significant positive correlation between the velocity and density, which was further related to the impact of the fat content. Water dilution caused different changes in velocity, depending on the ratio of fat content and continuous phase of the samples. A higher fat content of the samples resulted in a smaller effect of dilution on velocity. This study showed that the ultrasonic system allows rapid and non-destructive analysis for comparison of liquid dairy samples and appears promising for detection of dilution.

1. Introduction

Animals have utilized low intensity ultrasound for millions of years, for instance bats and dolphins use echo location for hunting or navigation. Ultrasound is defined as sound waves with frequencies exceeding 20 kHz, which is above the hearing limit of the human ear (Awad, Moharram, Shaltout, Asker, & Youssef, 2012). Sound propagates through materials as mechanical waves, and its velocity and intensity are impacted by the interaction between the sound wave and materials. Consequently, analysing the velocity and the change of intensity could help to obtain information about the some of the properties of materials. For instance to understand bone density (Lee, Coan, & Boussein, 1997), to evaluate compressive strength of concrete (Demirboğa, Türkmen, & Karakoç, 2004), etc.

In modern times, ultrasonic techniques have also been applied by food industries. These industrial applications for food characterization include, amongst others, fat content analysis of edible oil (Yan et al., 2019), determination of the botanical origin of sugars in juices

(Contreras, Fairley, McClements, & Povey, 1992), and assessment of the composition of fish and poultry (Sigfusson, Decker, & McClements, 2001). As a rapid screening approach without any preparation or destruction of the sample, ultrasonic techniques could be easily fitted in online monitoring systems and allow for real-time adjustments during processing. Among several parameters which could be determined by ultrasound, measurement of the ultrasonic velocity is considered as the basis of the ultrasonic techniques used to evaluate food properties (McClements & Gunasekaran, 1997).

One of the food categories for which ultrasonic techniques have been used before are dairy products. Dairy products are consumed worldwide. There are various types of dairy products being produced, including liquid as well as solid products. Liquid milk and cream are two principal liquid dairy products. The cost of these dairy products is related to their composition properties such as their fat, protein, and lactose contents. It is therefore important for manufacturers to be able to characterize these products and adjust them. One area of characterization is to detect adulteration. As an important food in the human diet

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with a high nutritional value, milk is a constant target of adulteration (Nascimento, Santos, Pereira-Filho, & Rocha, 2017). Despite the various possible types of milk adulteration, dilution is the easiest way to adulterate milk, especially if milk payment is primarily based on the volume of milk. The commonly used techniques for the measurement of milk composition range from cheap and high-throughput techniques like Fourier transformed infrared spectroscopy (FTIR), to more expensive and labour-intensive ones like liquid chromatography and mass spectrometry. Nevertheless, these techniques are either time consuming or destructive. Some scholars reported applications of the ultrasonic technique to dairy products, for instance, for evaluating the fat content and particle size distribution, using ultrasonic attenuation (Dukhin, Goetz, & Travers, 2005), monitoring the crystallization of milk fat (Singh, McClements, & Marangoni, 2004), testing reconstitution of milk powders (Meyer, Rajendram, & Povey, 2006), monitoring the phase change for yogurt fermentation (Ogasawara, Mizutani, Ohbuchi, & Nakamura, 2006), and detection of milk adulterated with melamine (Elvira, Rodríguez, & Lynnworth, 2009). These previous studies indicate that ultrasonic velocity is influenced by the particles and fat content in liquid samples. However, no specific study was found on profiling of various types of dairy emulsions, i.e. milks and creams of different fat contents, using ultrasonic velocity, it is therefore promising to apply this rapid and non-invasive technique in characterization of dairy products.

In this study, we explored the influence of fat content of liquid milk/cream products on their ultrasound characteristics. A pulse-echo ultrasonic system was applied to characterize diverse liquid dairy products with a wide range of fat contents, from double creams to skimmed milks. The range was further extended by dilution of the products to four different levels, to test for the sensitivity towards detecting this common type of adulteration. The study thus aims to evaluate the developed ultrasonic velocimetry system for profiling liquid dairy products, and to examine the underlying causes for the variation in ultrasonic velocity between the various products.

2. Materials and methods

2.1. Samples

A total of 18 liquid dairy product samples of four different brands were purchased from supermarkets in Ireland during summer (June to August 2018). They included six types of dairy products of which three samples were obtained each: double cream samples, normal cream samples, and light cream samples, as well as whole milk samples, semi-skimmed milk samples, and skimmed milk samples. All samples were stored at 4 °C and analysed before their expiry date.

These 18 samples were then mixed with demineralised water in the following water/dairy sample ratios (w/w) to create four dilution levels for each sample, resulting in a water/product ratio 20/80, 40/60, 60/40, and 80/20, based on weight percentage. The mixed solution was stirred for 1 min and was measured right after stirring. The final sample set included 90 samples, including 18 undiluted samples and 72 diluted samples.

2.2. Ultrasonic velocity analysis

A purpose-built pulse-echo system was utilized to measure the ultrasonic velocity. As shown in Fig. 1, the system comprised a sample cell with stainless-steel bottom, an immersion transducer (Panametrics-NDT, Olympus NDT U.K. Ltd, Rotherham, South Yorkshire, UK) with a central frequency of 5 MHz, a pulse generator/receiver (Panametrics-NDT Model 5800, Olympus NDT U.K. Ltd., Rotherham, South Yorkshire, UK), an oscilloscope (Tektronix TDS 210, Tektronix UK Ltd., Bracknell, UK), and a computer (Dell, Texas, US).

The principle of the pulse echo system has been described before (Awad et al., 2012; Yan et al., 2019). The monopolar electrical pulses are generated/received by a computer-controlled pulse generator/receiver,

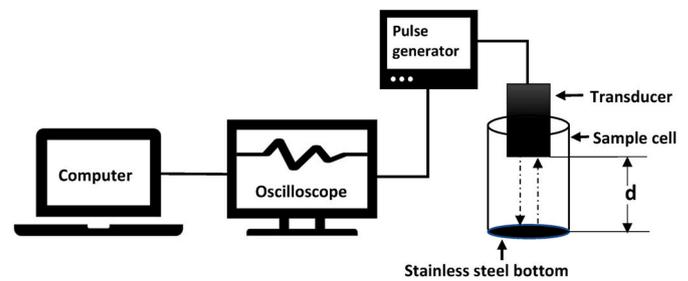


Fig. 1. Schematic diagram of the ultrasonic pulse echo measurement device.

and then transformed to ultrasonic pulses by the transducer. The pulse generator/receiver was operated in pulse echo mode, with a pulse repetition frequency of 1 kHz, 12.5 μ J pulse energy with 20 dB input attenuation, and 20 dB receiver amplification. The ultrasonic pulse was generated from the transducer, propagated through the dairy samples, bounced back from the stainless-steel bottom of the sample cell (solid reflector), and propagated back to the pulse receiver. The transducer then transformed the returned ultrasonic pulses to electrical signals, which were shown on the oscilloscope. Next, the electrical signals were transferred to the computer from the oscilloscope. The measurements were conducted with various distances between the transducer and the bottom of the sample cell. The transducer was mounted on a micrometre stage with a positioning accuracy of ± 0.005 mm. The signals were collected using a MATLAB (R2015b, MathWorks Inc., Natick, MA, USA) program.

The ultrasonic velocity was calculated using the distances and the propagation times of the echoes. Six consecutive distances (10.00 ± 0.005 , 12.00 ± 0.005 , 14.00 ± 0.005 , 16.00 ± 0.005 , 18.00 ± 0.005 , and 20.00 ± 0.005 mm) were used for each sample measurement. The ultrasonic velocity was then determined by two measurements of two consecutive distances using Eq. (1):

$$c = \frac{2 \times (d_2 - d_1)}{t_2 - t_1} \quad \text{Eq. (1)}$$

where c is the ultrasonic propagating velocity (m/s) in the sample; d_1 is the shorter distance of the one-way echo path (m), d_2 is the consecutive distance of the one-way echo path (m); t_1 is the time of receiving the first signal (± 0.1 μ s) under the distance of d_1 , t_2 is the time of receiving the first signal (± 0.1 μ s) under the distance of d_2 . Five velocity values were determined based on the six measurements and averaged. This resulted in a dataset of 18 samples \times five dilution levels \times six measurements is 540 data points. All samples were placed in water bath to maintain the temperature (24.0 ± 0.1 °C) before the measurements. The measurements were performed at 24.0 ± 0.5 °C, which were monitored by an electronic thermometer. The device was calibrated by measuring the ultrasonic velocity in water (1494 ± 3 m/s at 24.0 °C) (Engineering ToolBox, 2004).

2.3. Viscosity analysis

A stress-controlled rheometer (MCR 301, Anton Paar, Austria) with a stainless-steel double gap geometry (DG26.7/TI-SN21833) was used to measure the viscosity. The viscosity was measured by increasing the shear rate from 0.1 to 200/s in 3 min. A total of 200 data points were recorded for each sample. All experiments were performed in duplicate at 24.0 ± 0.5 °C. The viscosity measured at the shear rate 100/s was used for the statistical analysis.

2.4. Density analysis

The density of the 18 commercial samples was determined using a gravimetric method. A measuring cylinder (25.0 mL) was weighed (m_1) using an electronic balance (± 0.001 g), then 25.0 ± 0.1 mL of sample (v)

was quantified using the measuring cylinder. Subsequently, the measuring cylinder and the sample were weighed together (m_2). The density of the sample was calculated by Eq. (2):

$$\rho = \frac{m_2 - m_1}{v} \times 1000 \quad \text{Eq. (2)}$$

where ρ is the density of the sample (g/L), m_1 is the weight of empty measuring cylinder (g), m_2 is the total weight of the measuring cylinder and the cream sample (g), and v is the accurate volume of the sample (mL). The sample was equilibrated at 24.0 ± 0.1 °C in a water bath before the measurement, and the measurements were conducted in duplicate at room temperature (24.0 ± 0.5 °C).

2.5. Compositional analysis

The composition of the 18 commercial samples was measured using a MilkoScan FT120 instrument (Foss Electric, Hilleroed, Denmark). The compositional features obtained for the milk samples included fat content, protein content, total solids content (TS), solids non-fat content (SNF), and lactose content, and for the cream samples included the fat content, protein content, TS content, and SNF content. Lactose could not be measured in the creams because the cream module of the instrument was not calibrated for this parameter. Samples were equilibrated to 24.0 ± 0.1 °C in a water bath prior to analysis. All analyses were conducted in duplicate, resulting in 36 measurements. The average value of the duplicate was used for analysis.

2.6. Calculations and statistical analysis

2.6.1. Density and compositional calculations

The density and composition of the diluted samples were calculated based on their non-diluted counterparts. The density and compositional feature of the $a\%$ sample was calculated using Eq. (3) and Eq. (4), respectively:

$$\rho_{a\%} = \frac{100 \text{ g}}{[a\% \times 100 \text{ g} / \rho_{\text{dairy}}] + [(1 - a\%) \times 100 \text{ g} / \rho_{\text{water}}]} \quad \text{Eq. (3)}$$

$$X_{a\%} = a\% \times X_{\text{dairy}} + (1 - a\%) \times X_{\text{water}} \quad \text{Eq. (4)}$$

where $\rho_{a\%}$ is the density of the diluted sample (g/L), $a\%$ is equal to the weight percent of product in the diluted sample, thus being 80%, 60%, 40%, and 20%, respectively. ρ_{dairy} is the density of the sample (g/L), ρ_{water} is the density of water (g/L). $X_{a\%}$ is the compositional feature of the diluted sample (g/100 g), X_{dairy} is the compositional feature of the commercial dairy product (g/100 g), i.e. protein, fat, lactose, TS, SNF for the milk samples, and protein, fat, TS, SNF for the cream samples. X_{water} is the corresponding feature of water (g/100 g).

2.6.2. Calculation of compressibility

The compressibility of the samples is calculated based on the means of velocity and density. The velocity in a fluid is described by the Wood equation (Wood, 1946), which is also called Laplace equation:

$$v = \sqrt{\frac{1}{k \times \rho}} \quad \text{Eq. (5)}$$

Consequently the compressibility was obtained using Eq. (6):

$$k = \frac{1}{v^2 \times \rho} \quad \text{Eq. (6)}$$

where k is the compressibility (Pa^{-1}), v is the velocity of ultrasound (m/s), and ρ is the density (g/L).

2.6.3. Statistical analysis

Means and standard deviations of the measured physical properties

were calculated for each sample, and for the six dairy product groups. Because not all the data was normally distributed, non-parametric Kruskal-Wallis tests were applied for comparisons between groups, followed by the pairwise Mann-Whitney U tests. Correlation between physical properties were determined by computing Pearson correlation coefficients (r). The non-parametric tests were performed using SPSS statistic 25 (IBM, Chicago, IL, USA), whereas the other analyses were performed using R 3.6.1 (R Foundation for Statistical Computing, Vienna, Austria).

3. Results and discussion

3.1. Ultrasonic velocity in the dairy products

3.1.1. Ultrasonic velocity in the original dairy samples

The ultrasonic velocities in the 18 dairy samples are presented in Table 1. In general, the ultrasonic velocity in the milk samples (ranging from 1520 to 1525 m/s) is significantly higher than that in the cream samples (ranging from 1489 to 1506 m/s). More specifically, the ultrasonic velocity in the light cream samples was significantly higher than those in the double cream and normal cream samples, and the ultrasonic velocity in the milk samples appears to have an upward trend for the milk products from whole milk samples to skimmed milk samples (Table 1). Based on this data, it may be hypothesised that the ultrasonic velocity in dairy products depends on its fat content.

Milk and cream can both be considered as an emulsion of continuous water phase with a dispersed lipid phase (Huppertz & Kelly, 2006). The main content of the continuous phase, also called milk plasma, is water containing protein, lactose, minerals, and other dissolved solids. The dispersed phase is composed of milk fat globules. Ultrasonic velocity in a pure liquid was first described by the Wood equation (Wood, 1946). Based on the equation, scholars developed the theory that the ultrasonic velocity in an emulsion depends on the volume ratio of the two phases of

Table 1

Means and standard deviations of the ultrasonic velocity, viscosity^a and density for the six dairy groups and 18 commercial dairy samples. Different letters indicate significant differences between the dairy groups (Kruskal-Wallis test followed by pairwise Mann-Whitney U test, $p < 0.05$).

Sample	Velocity (m/s)	Viscosity (mPa.s)	Density (g/L)
Double cream 1	1491.4 ± 6.1	30.0 ± 0.5	974 ± 2
Double cream 2	1493.7 ± 7.3	17.0 ± 0.7	993 ± 2
Double cream 3	1483.7 ± 3.0	36.6 ± 1.7	981 ± 1
Mean double cream group	1489.6 ± 5.2	27.9 ± 10.0 ab	983 ± 9 a
	a		
Normal cream 1	1499.3 ± 4.7	11.5 ± 0.3	995 ± 2
Normal cream 2	1497.0 ± 2.5	11.8 ± 0.2	998 ± 2
Normal cream 3	1492.5 ± 0.0	15.1 ± 0.7	980 ± 3
Mean normal cream group	1496.3 ± 3.4	12.8 ± 2.0 c	993 ± 12 a
	a		
Light cream 1	1507.2 ± 3.1	86.4 ± 10.5	1013 ± 1
Light cream 2	1503.8 ± 0.0	6.1 ± 0.0	1007 ± 1
Light cream 3	1508.3 ± 2.5	28.6 ± 0.3	1013 ± 0
Mean light cream group	1506.4 ± 2.4	40.4 ± 41.4 a	1011 ± 3 b
	b		
Whole milk 1	1520.9 ± 4.1	2.2 ± 0.0	1036 ± 0
Whole milk 2	1519.8 ± 4.8	2.0 ± 0.0	1035 ± 1
Whole milk 3	1523.2 ± 3.2	2.2 ± 0.0	1037 ± 0
Mean whole milk group	1521.3 ± 1.8 c	2.1 ± 0.1 bc	1036 ± 1 c
Semi skimmed milk 1	1522.1 ± 2.6	1.8 ± 0.0	1037 ± 1
Semi skimmed milk 2	1522.1 ± 4.8	1.9 ± 0.0	1037 ± 0
Semi skimmed milk 3	1525.6 ± 2.6	1.9 ± 0.0	1038 ± 1
Mean semi skimmed milk group	1523.2 ± 2.0 c	1.8 ± 0.1 bc	1037 ± 1 cd
Skimmed milk 1	1525.6 ± 4.9	2.3 ± 0.1	1040 ± 1
Skimmed milk 2	1523.2 ± 3.2	1.7 ± 0.0	1042 ± 1
Skimmed milk 3	1526.7 ± 4.1	1.7 ± 0.0	1038 ± 1
Mean skimmed milk group	1525.2 ± 1.8 c	1.9 ± 0.4 c	1040 ± 2 d

^a Viscosity at shear rate 100/s was used.

emulsion, the dissolved solid content in the continuous phase, and the physical properties, for instance the droplet size, of the fat fraction of the emulsion (McClements & Povey, 1987; Povey, 1989). The dissolved solids in the continuous phase of an emulsion correlated positively to the ultrasonic velocity, while the volume fraction of milk fat was shown to have a negative effect on the ultrasonic velocity. Considering the composition of the milk plasma is similar for milk and cream products, therefore the large difference in the ratio of the continuous phase and dispersed phase is probably a key factor affecting the ultrasonic velocity. This is consistent with the results that ultrasonic velocity is decreasing with the increasing fat content of samples, as shown by the result of the ultrasonic velocities in this study (Table 1).

To sum up, the ultrasonic velocity increased with decreasing fat content of the samples. Using the ultrasonic velocity, cream samples can be differentiated from milk samples, and light creams can be differentiated from double and normal creams. Considering that the composition of continuous milk plasma phase for the six dairy products is almost identical, the proportion of fat content appears to be the main factor determining the difference in ultrasonic velocity between the six product groups.

3.1.2. Ultrasonic velocity in the diluted dairy samples

The ultrasonic velocity in the diluted liquid dairy samples revealed a variable response that differed between the six dairy products (Fig. 2). For the three liquid milk samples (whole milk, semi-skimmed milk, and skimmed milk), the ultrasonic velocity decreased and got closer to the velocity of water (1492.5 m/s) as the sample was more diluted. The velocity in the diluted light cream samples showed a similar trend with those in the diluted milk samples, but with a less steep slope. The change of velocity in the diluted normal and double cream samples was much smaller.

It is important to point out that the dilution of liquid dairy samples with water could cause two opposite effects on the ultrasonic velocity: on the one hand, lowering the fat fraction by dilution could cause an increase in velocity (McClements, 1988). On the other hand, lowering the dissolved solids content in the continuous phase could cause a decrease of velocity (McClements & Gunasekaran, 1997). Since the milk plasma is the principal phase of the milk products (>95% weight fraction), the declining dissolved solids concentration of these samples after dilution is considered as the principal factor for the velocity decrease.

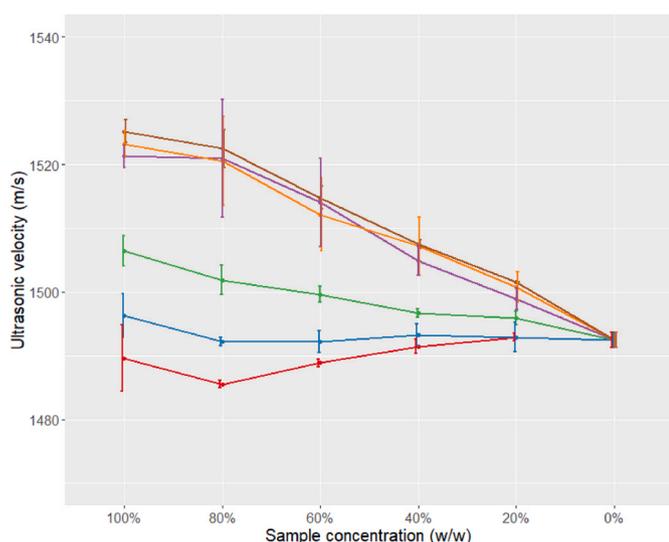


Fig. 2. Ultrasonic velocities in the double cream (red), normal cream (blue), light cream (green), whole milk (purple), semi skimmed milk (orange), and skimmed milk (brown) of various concentrations (100%–0%). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

The velocity in the light cream generally declined less with dilution, compared with the milk samples (Fig. 2), which is probably caused by the smaller difference between the milk plasma and fat content in the light cream than in the milk samples. Consequently, the velocity decline caused by dilution of the dissolved milk plasma was partly counteracted by the simultaneously decreasing fat content. This effect became more extensive in the normal and double cream samples, which means that the impact of dilution of the continuous milk plasma phase was almost completely balanced out by the decreasing fat content, resulting in almost no change of the velocity in these diluted cream samples (Fig. 2).

In summary, the ultrasonic velocity in the diluted milk samples decreased with dilution level, caused by a reduced total dissolved content in the samples. The ultrasonic velocity in the diluted cream samples showed smaller impact by the increased level of dilution, due to the counterbalancing effect of dilution on the dissolved milk plasma phase and the fat content.

3.2. Viscosity of the dairy products

The shear rheological behaviour of the undiluted samples is shown in Fig. 3. The apparent viscosities of the cream samples were generally larger than those of the milk samples. For emulsion systems, the viscosity increases linearly with the fraction of the dispersed phase (Bakshi & Smith, 1984; Huppertz & Kelly, 2006; Kyazze & Starov, 2004; Phillips, McGiff, Barbano, & Lawless, 1995), explaining why the larger fat fraction of the cream samples correlated with the higher viscosity (Fig. 3). This is also in line with the result that higher viscosity was observed for the whole milk samples compared to the semi-skimmed and skimmed milk.

In contrast to the general relation between fat content and viscosity, it is noted that the viscosity of the light cream samples was higher, and its decreasing rate with the shear rate was lower, compared with the other two types of cream products (Fig. 3). This may be due to the fact that, for all the 18 commercial products used in this study, only the three light cream samples contained ingredients other than milk, including carrageenan, skimmed milk powder and glucose (according to their labels). These components are added as thickeners to obtain the desired texture of the product, which is more similar to higher fat cream products. Previous studies showed that carrageenan can greatly increase the viscosity of a cream product, by means of bridging the casein micelles

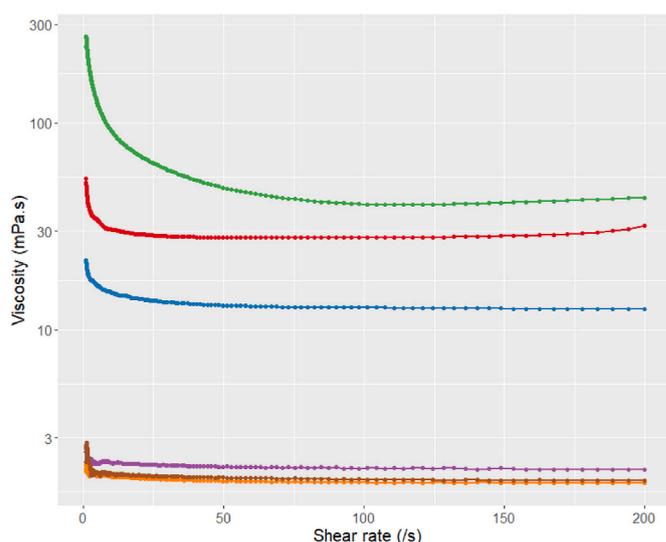


Fig. 3. The result of the viscosity of the double cream (red), normal cream (blue), light cream (green), whole milk (purple), semi skimmed milk (orange), and skimmed milk (brown) samples. The average value of the three samples in each group was used. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

(Camacho, Martínez-Navarrete, & Chiralt, 2005; Langendorff et al., 2000). As a result, the viscosity of the light cream samples was higher than was expected based on its fat content.

To summarize, the viscosity of the cream samples was higher than the milk samples. Due to the addition of polymers, the viscosity of light creams was higher than the value expected from its fat content only.

3.3. Density of the dairy products

The density of the six groups of dairy products showed an upward trend with decreasing fat contents (Tables 1 and 2). The density of the cream products was significantly lower than that of the milk products. As discussed in 3.1.1, the dairy products could be considered as an emulsion of two phases: milk plasma and milk fat globules. The density of the milk plasma changes very slightly depending on the components dissolved in it, which results in a density of around 1.030 g/L at 20 °C. The density of the milk fat depends on variations in the fat composition, resulting in a density around 0.92 g/L at 20 °C (Walstra, Wouters, & Geurts, 2005). The lower density of the milk fat therefore causes a decreasing density of dairy products with increasing fat content, leading to the highest density for the skim milk and the lowest density for the double cream.

3.4. Relations between the ultrasonic velocity, viscosity, and density

3.4.1. Relation between the ultrasonic velocity, viscosity, and density for the original samples

To explore the relation of the ultrasonic velocity, viscosity, and density for the original and diluted dairy samples, scatter plots between the velocity and viscosity and density for both original and diluted dairy samples are presented (Fig. 4). As shown in Fig. 4a, no clear relationship between the velocity and viscosity was observed for the original dairy samples, which was supported by the Pearson correlation analysis, which resulted in no significant correlations between these two features for the original dairy samples ($r = -0.45, p = 0.063$). On the contrary, it can clearly be observed that the velocity in the dairy samples increased with their density (Fig. 4b). Correspondingly, a significant positive correlation between velocity and density ($r = 0.98, p < 0.05$) for the original dairy samples was obtained.

3.4.2. Relation between the ultrasonic velocity, viscosity, and density for the diluted dairy samples

It is shown in Fig. 4a that no clear pattern was observed between the velocity and viscosity for the diluted dairy samples. The velocity in the diluted samples increased with their density, as shown in Fig. 4b, resulting in a significant positive correlation between the velocity and density ($r = 0.94, p < 0.05$). Considering that density depends largely on

Table 2

Means and standard deviations of the milk composition for the six dairy groups ($n = 3$).

Features	Double cream	Normal cream	Light cream	Whole milk	Semi skimmed milk	Skimmed milk
Fat (g/100 g)	58.0 ± 3.1	50.3 ± 0.9	35.7 ± 2.4	3.5 ± 0.2	1.6 ± 0.6	0.2 ± 0.1
Protein (g/100 g)	3.3 ± 0.1	3.8 ± 0.1	4.5 ± 0.5	3.9 ± 0.1	3.9 ± 0.1	4.0 ± 0.12
TS (g/100 g)	63.3 ± 2.5	56.3 ± 0.7	42.8 ± 2.3	13.6 ± 0.3	11.8 ± 0.5	10.6 ± 0.6
SNF (g/100 g)	0.6 ± 0.3	1.7 ± 0.2	2.7 ± 0.5	10.0 ± 0.1	10.1 ± 0.2	10.4 ± 0.5
Lactose (g/100 g)	N.A.	N.A.	N.A.	4.5 ± 0.0	4.5 ± 0.1	4.7 ± 0.3

N.A., non-available because the feature was not calibrated for the instrument.

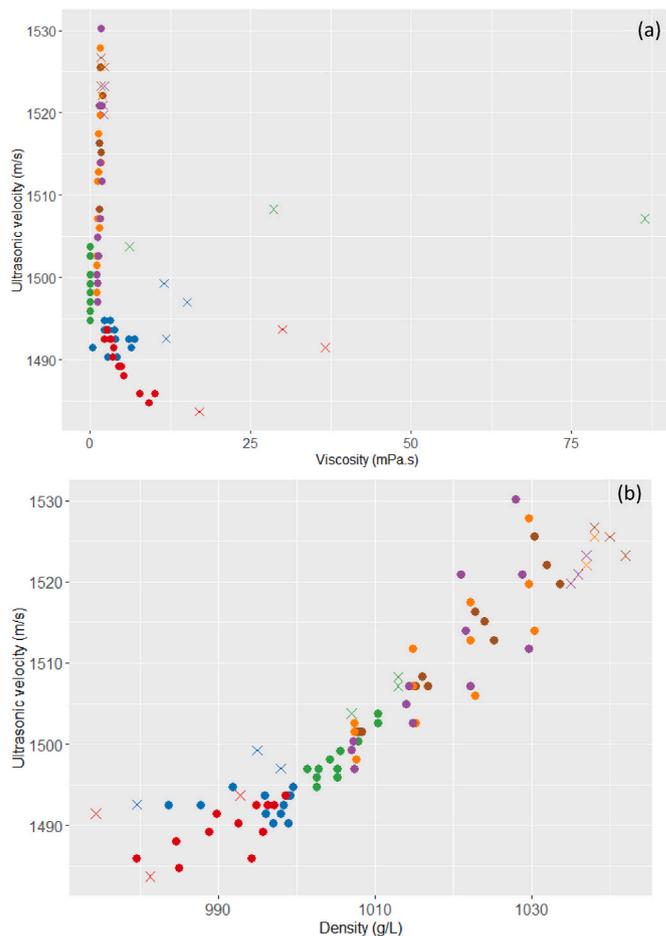


Fig. 4. Scatter plots of (a) ultrasonic velocity and viscosity for the double cream (red), normal cream (blue), light cream (green), whole milk (purple), semi skimmed milk (orange), and skimmed milk (brown), for the original samples (x) and their diluted counterparts (circle); and (b) ultrasonic velocity and density for the double cream (red), normal cream (blue), light cream (green), whole milk (purple), semi skimmed milk (orange), and skimmed milk (brown), for the original samples (x) and their diluted counterparts (circle). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

the composition, the correlation between velocity and density may be related to the composition of the samples. The relations between the velocity, density, and fat content were therefore analysed.

3.5. Relation between the ultrasonic velocity, density, compressibility, and fat content

To explore the correlation of the velocity, density, and the compositional features for the original dairy samples, the Pearson correlation

Table 3

Pearson correlation coefficients for the correlation between the ultrasonic velocity, density, and the compositional features^a for the original dairy group ($n = 18$).

Compositional Features	Velocity	Density
Fat	-0.98*	-0.98*
Protein	0.43	0.45
Total solids	-0.98*	-0.98*
Solids non-fat	0.96*	0.95*

* Significant correlations ($p < 0.05$) are marked with *.

^a Only the available compositional features for both milk and cream samples are used for the calculations.

coefficients (r) between the physical and compositional properties are presented in Table 3. The velocity and density were significantly negatively correlated with the fat content and TS content, and positively correlated with the SNF content. For the original dairy samples, the fat to milk plasma ratio of the samples decreases, starting from the samples of highest fat content (double cream milk) to the ones containing no fat (skimmed milk). Therefore, the fat content is considered the main factor explaining the differences of the velocity in the samples. This is in line with the hypothesis that the ultrasonic velocity in the dairy products depends on the fat content of the samples.

The changes of the velocity caused by dilution of the six types of dairy products showed different trends, which were further related to the changes of the compressibility. Relations between the velocity, compressibility, and fat content in this study are presented in Fig. 5. According to the Wood equation (Eq. (5)), velocity in a fluid is influenced by both density and compressibility. The dilution of the milk samples impacts differently on the density and compressibility, which resulted in opposite influences on velocity: on the one hand, the increasing compressibility led to a decrease in the velocity; on the other hand, the decreasing density led to an increase in the velocity. The results show that the velocity in the milk samples decreased with the dilution (Fig. 5b), indicating that the increase in compressibility has dominant effects on velocity, which resulted from the dilution of milk plasma. While for the diluted double cream and normal cream samples, the increase of velocity indicates that the decrease in compressibility has a larger impact, which resulted from the considerable reduction in fat content (Fig. 5). This result is in line with the discussion in section 3.1.2, that the different ratio of fat to milk plasma can affect the velocity in diluted samples: the larger the difference between the proportion of the two phases, the greater the change in velocity caused by dilution. The counteracting effects between the fat content and continuous milk plasma phase on the velocity provide a potential for determination of the ratios of the two phases in dairy products using ultrasonic velocity, as a larger effect of dilution indicates a larger fraction of milk plasma. In addition, for the milk samples with a relatively low-fat content, the velocity might be used for detection of adulteration with water.

In summary, the difference of fat content is the principal factor resulting in the difference of ultrasonic velocity in the undiluted dairy samples. While for the diluted samples, the changes in ultrasonic velocity caused by dilution are influenced by the ratio of fat and continuous phases in the original samples.

4. Conclusions and outlook

The study showed a potential to apply ultrasonic velocity measurement for characterization of liquid dairy products. The ultrasonic velocity in liquid dairy products decreases with increasing fat content. The fat content appears the determining factor for ultrasonic velocity in undiluted dairy products, whereas both fat content and the fraction of the continuous phase play a role in the changes in ultrasonic velocity in diluted dairy products. Consequently, the higher the fat content of a sample, the smaller the change in ultrasonic velocity caused by dilution.

The strong correlation between the velocity and fat content could be used for characterization of the dairy products. The opposite effects on velocity caused by dilution, namely increase of velocity with decreasing fat content and decrease of velocity with dilution of the continuous phase, provides a potential for determination of the ratio of two phases in the dairy products using ultrasonic velocity. As a rapid and non-destructive technique, ultrasonic velocity is promising to be applied for detection of milk adulteration with water. In addition, it would be also interesting to explore its capability with regard to the detection of other common types of milk adulterations, especially those that are aimed at masking the dilution with water.

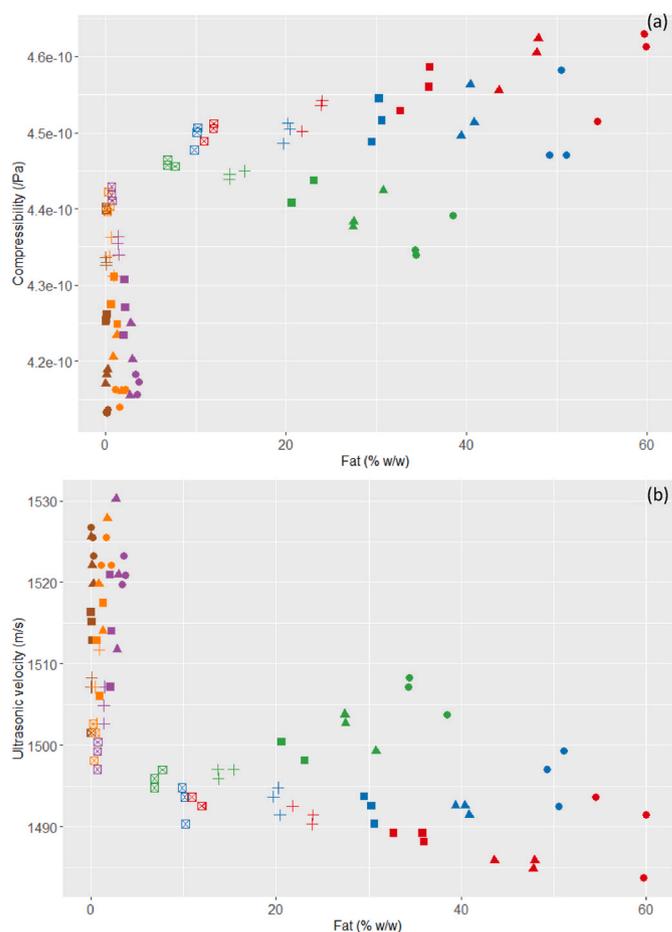


Fig. 5. Relation between (a) compressibility and fat content, and (b) ultrasonic velocity and fat content, for the double cream (red), normal cream (blue), light cream (green), whole milk (purple), semi skimmed milk (orange), and skimmed milk (brown) samples of concentration of 100 g/100 g (circle), 80 g/100 g (triangle), 60 g/100 g (square), 40 g/100 g (+), and 20 g/100 g (x in square). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

CRediT authorship contribution statement

Yuzheng Yang: Conceptualization, Investigation, Formal analysis, Writing - original draft, Writing - review & editing. **William M.D. Wright:** Methodology, Software, Formal analysis, Writing - review & editing. **Kasper A. Hettinga:** Formal analysis, Writing - review & editing. **Saskia M. van Ruth:** Conceptualization, Methodology, Writing - review & editing.

Declaration of competing interest

None.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.lwt.2021.110616>.

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