Impact of thermo-sonication on quality indices of starch-based sauces

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ARTICLE INFO

Keywords:
Ultrasound
Rice starch
Rheology
Texture
Freeze/thaw cycle
White sauce

ABSTRACT

In this study, ultrasoundation, a physical, relatively cheap, and environmentally benign technology, was investigated to characterize its effect on functional properties of rice starch and rice starch-based sauces. Temperature-assisted ultrasound treatment improved the granular swelling power, fat and water absorption capacities, and thermal properties of rice starch, signifying its suitability in the formulation of starch-based sauces. Rheological characterization of the formulated sauces revealed a shear-thinning flow behavior, well described by the Ostwald-de Waele model, while viscoelastic properties showed the existence of a weak gel. Results indicated that ultrasoundation significantly enhanced the pseudoplastic behavior of starch-based sauces. Additionally, textural analysis showed that textural attributes (stickiness, stringiness, and work of adhesion) were also improved with ultrasoundation. Moreover, enhanced freeze/thaw stability was also achieved with ultrasound-treated starch-based sauces. Overall, the results from this study show that ultrasound-treated starches can be used in the formulation of sauces and potentially other food products, which meets the requirements for clean label and minimally processed foods.

1. Introduction

Nowadays, starch is increasingly finding numerous applications in the food processing, cosmetic, and pharmaceutical industries. This is because it is ubiquitous, inexpensive, biodegradable, biocompatible, non-toxic, and possesses peculiar physicochemical properties [1]. Starch is contained in the amyloplast as discrete granules with varying morphology depending on plant type. In its polymeric form, it is essentially composed of straight-chain amylose (≈30%), connected by α-1,4-glycosidic bond, and branched-chain amylopectin (≈70%), which is connected by α-1,6-glycosidic bond [2].

Native starch is extracted from naturally existing roots and cereals, such as cassava, wheat, and rice. However, the industrial application of native starch in food formulations is limited mainly due to syneresis, retrogradation, poor thermal, and shear stability [3]. To overcome these impediments, several starch modification technologies have been developed. These methods are designed to improve the functional properties of starch and include chemical, enzymatic, genetic, and physical manipulations. Detailed reviews on these modification techniques have been discussed by several authors [4,5]. Chemical modification techniques, such as acetylation, and cross-linking, subsume new functional groups without affecting the morphological properties of the starch granules [6]. Though desired functional properties are achieved, chemical modification utilizes chemical reagents that are non-eco-friendly. Genetic and enzymatic modification techniques are usually associated with high cost [7] and the need to prove their safety, especially when it is intended to be used as a food ingredient [8]. Moreover, chemical modification techniques conflict with the increasing consumer demand for clean label and minimally processed foods. Clean label foods are foods that are organic, natural, and free from artificial additives. With this definition, food products formulated with starches modified through chemical, enzymatic, and genetic means do not meet the description of clean label foods and are usually identified by an E number on the label of the product [9]. Against this background, the food industry generally favors the use of physical modification techniques, such as high-pressure processing, and ultrasonication. Over the past decade, ultrasonication has gained increased usage owing to its shorter processing time, lower operation and maintenance cost, increased product yield, and the ability to inactivate pathogens [10].

Ultrasonication is designed as a non-thermal environmentally friendly modification technique that disrupts the arrangement of the polymers within the starch granule [11]. Ultrasound refers to sound waves possessing a frequency beyond the threshold of human hearing (16 – 20 kHz). High-intensity ultrasound is usually characterized by a frequency of 20 – 500 kHz and intensity greater than 1 W/cm² [12]. The severity of ultrasonication effect depends on the origin and moisture

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https://doi.org/10.1016/j.ultsonch.2021.105473
Received 8 December 2020; Received in revised form 24 December 2020; Accepted 16 January 2021
Available online 2 February 2021
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content of the starch sample, intensity, frequency, temperature, and duration of application [6]. Amini et al. [13] reported that temperature and time of exposure to ultrasonication strongly influenced functional and rheological properties of corn starch, while concentration and amplitude of ultrasonication had little effect. Indeed, ultrasound waves create cavities or bubbles in the medium through which it is passed [14]. When these cavities implode, they lead to the formation of fissures or holes on the granular surface; thereby weakening its structural integrity and eliciting gelatinization and enhanced functional properties [15].

Many studies have scrutinized the effect of ultrasound treatment on the functional and physicochemical properties of starch paste [16–18]. Ultrasoundation was reported to reduce retrogradation and improve intrinsic viscosity and textural properties of potato starch paste [19]; though Fourier transform infrared spectroscopy did not demonstrate any noticeable molecular degradation. Zhu and Li [15] showed that ultrasonication increased water solubility and in vitro starch digestibility of quinoa flour, while decreasing gelatinization temperature, in vitro antioxidant activity, and total phenolic content. Low-intensity ultrasonication increased water solubility and

2.2. Ultrasound treatment

The method previously described by Amini et al. [13] was adopted with slight modification. Native rice starch (40 g) in water (100 mL) was treated for 20 min at different temperature levels (15, 30, 45, 60, and 75 °C, coded as S1, S2, S3, S4, and S5, respectively) by a high-intensity ultrasonic processor (Sonics VibraCell VCX500, Sonics & Materials Inc., Newton CT, USA) equipped with a 3 mm tapered probe. The processor was operated at a frequency of 20 kHz, power of 500 W, and amplitude of 40%. The probe was immersed in the sample at an approximate depth of 2 cm. The equipment was operated under an 80% pulse mode to ensure inhibition of heat build-up and enhance particle aggregation under the probe. During sonication, appropriate temperature levels were maintained using an unstripped water bath (JB Nova, Grant Instrument Ltd, Royston, United Kingdom). The sonicated samples were transferred into Ziploc bags and lyophilized for 70 h (Thermo Savant Modulyo Benchtop Freeze Dryer, Thermo Electron Corporation, Beverly, MA, USA). Holes were made on the top of the Ziploc bags to enhance moisture loss during drying. The control/non-sonicated sample (S0) was prepared by dispersing the appropriate weight of native starch in water. The dry samples were ground to powder and placed in a desiccator before further use. Temperature was chosen as the operating variable because it is one of the most influential factors affecting the severity of ultrasound treatment [13].

2.3. Starch characterization

2.3.1. Granular swelling power

Granular swelling power was determined using a method described by Liu et al. [28]. 50 mg of the native and modified starches were transferred into dry 15 mL centrifuge tubes, weighed (M1), and mixed with 5 mL of water. The tubes were heated in a water bath at different temperature levels (55, 65, 75, 85, and 95 °C) for 20 min. The samples were homogenized at intervals of 5 min using a vortex mixer at 1000 rpm (Corning Inc., New York, USA). After heating, the samples were cooled to room temperature and centrifuged at 657 × g (Sorvall Legend XTR, Thermo Electron LED GmbH) for 20 min. The supernatant was decanted and the tube with its residue was weighed (M2). The granular swelling power was computed using eqn. (1).

\[
\text{Granular swelling power} = \frac{(M_2 - M_1)}{\text{weight of starch}} \times 100\% \tag{1}
\]

2.3.2. Fat and water absorption capacities

Fat and water absorption capacities were determined separately. Starch (0.5 g) was transferred into dry centrifuge tubes with predetermined weight (W1). For determination of water absorption capacity (WAC), 5 mL of water was added using a bottle top dispenser (Thermo Fisher Scientific, Ontario, Canada), while for fat absorption capacity (FAC), 5 mL of sunflower oil was added to the centrifuge tubes using a syringe. The resultant sample was homogenized for 1 min at 1000 rpm using a vortex mixer. The mixture was allowed to rest for 5 min, homogenized again, and centrifuged at 1500 rpm for 15 min at an operating temperature of 23 °C. The supernatant was carefully decanted and the tube with its residue was weighed (W2). FAC or WAC was determined using eqn. (2).

\[
\text{FAC or WAC} = \frac{(W_2 - W_1)}{\text{weight of starch}} \times 100\% \tag{2}
\]

2.3.3. Thermal analysis

Starch gelatinization behavior was evaluated using a differential scanning calorimeter (DSC250, TA Instruments, New Castle, Delaware, USA). The instrument was calibrated using an indium standard before sample measurements. Starch samples (14 ± 2 mg) of 1:1.3 (starch–water ratio) were transferred into aluminum pans and hermetically sealed. The samples were then heated from 25 to 80 °C at a heating rate of 2 °C/min. An empty aluminum pan was used as a reference and a flow rate of 50 mL/min for dry nitrogen was maintained throughout the experiment. Onset (T onset), peak (T peak), conclusion (T end) temperatures, and the enthalpy of gelatinization (ΔH) were evaluated.

2.4. White sauce preparation

White sauce was prepared using the method described by Arocas et al. [29]. 9.3 g powdered skimmed milk (Selection, Metro Brands, Qu´ebec, Canada), 2.55 g sunflower oil (Selection, Metro Brands, Qu´ebec, Canada), 0.23 g salt (Sifto Table salt, Compass Minerals Canada Corp., Ontario, Canada), 6 g starch, and 82 mL of water were placed in a cooking device (Thermomix TM31, Vorwerk Electrowerke GmbH & Co. KG, Wuppertal, Germany) and heated to 90 °C at a heating speed of 1100 rpm in 5 min. The prepared sauces were transferred to glass containers, covered with aluminum foil, and allowed to cool to room temperature.
2.5. Rheological measurements

White sauce rheological properties were measured using a controlled stress rheometer (AR2000, TA Instruments, New Castle, Delaware, USA). The instrument was equipped with a 40 mm parallel plate geometry using a gap of 1000 µm. Laboratory grade compressed air (30 psi) was maintained throughout the experiment. Rheological properties were measured at 23 °C, regulated using a Peltier plate with an accuracy of ± 0.1 °C. Measurements for freshly prepared sauces were conducted four hours after preparation, while the other portion was frozen at −18 °C for three days to study the effect of a freeze/thaw cycle. At the end of the third day, the samples were allowed to thaw in a water bath (at room temperature) before rheological measurements were taken. For each procedure, samples without mechanical history, i.e. fresh samples, were utilized. Before measurements were taken, five minutes was allowed for sample equilibration or structure recovery. Excess samples were trimmed using a spatula after the head of the rheometer was lowered. Silicon oil (Fisher Scientific, Fair Lawn, New Jersey, USA) was placed around the edges of the samples to prevent drying during testing. Data analysis was performed using TRIOS software v5.1.1 provided by the instrument’s supplier.

2.5.1. Flow behavior

Flow properties were measured by recording the shear stress obtained when the shear rate was linearly increased from 1 to 200 s⁻¹ in 2 min (upward curve), then 200 – 1 s⁻¹ (downward curve) at the same time. This measurement is important because common food processes, such as mastication, stirring, and flow in pipes occur within the evaluated shear range [30]. The data obtained from the upward curve was adjusted to the power law model (eqns. (3)), previously used to model the flow behavior of white sauce [22].

\[
\sigma = k\dot{\gamma}^n
\]  
(3)

Where \(\sigma\) is shear stress (Pa), \(k\) is consistency index (Pa.s\(^n\)), \(\dot{\gamma}\) is shear rate (s\(^{-1}\)), and \(n\) is flow behavior index (dimensionless). Additionally, the percentage relative hysteresis (PRH) was calculated (eqn. (4)) as a means of comparing the structural integrity of the sauces.

\[
PRH = \left(\frac{A_{uc} - A_{dc}}{A_{uc}}\right) \times 100
\]  
(4)

Where \(A_{uc}\) and \(A_{dc}\) are areas of the upward and downward curves, respectively.

2.5.2. Viscoelastic properties

Oscillatory strain sweep was first conducted to determine the linear viscoelastic (LVE) region of each sauce. This was achieved by ramping strain from 0.5 to 100% at a frequency of 1 Hz. The samples were next subjected to dynamic frequency sweep experiments between 0.1 and 10 Hz, using a controlled variable of 2% strain within the LVE region. Values of viscoelastic parameters, such as storage modulus (\(G'\)), loss modulus (\(G''\)), and loss tangent (\(\tan \delta = G''/G'\)) were recorded as a function of frequency.

2.6. Textural analysis

Sauce texture analysis was performed using a texture analyzer (TA. HDplus, Stable Micro Systems, Surrey, United Kingdom) fitted with a 25 mm Perspex cylindrical probe (P/25P). The compression was carried out in a Perspex extrusion rig using a 50 kg load cell. The pre-test speed, test speed, and post-test speed were 4.0, 2.0, and 1.0 mm.s\(^{-1}\), respectively. Once a trigger force of 5 g has been detected on the surface of the sauce, the probe proceeds to compress it by traveling 5 mm. The maximum force required to separate the probe from the sample was termed stickiness. During the retraction of the probe, the sample was noticed to form a string. The distance (which quantifies the measure by which the sample stretches) the sample remains connected to the probe upon withdrawal was termed stringiness. The total amount of force (area under the force–deformation curve) required to withdraw the probe was termed work of adhesion. Textural analysis of the fresh sauces was conducted four hours after preparation. The same test was repeated after the samples were frozen for three days and allowed to thaw in a water bath at room temperature.

2.7. Freeze/thaw stability of white sauce

The freeze/thaw stability of white sauce was determined as described by Román et al. [23]. 15 g (\(W_c\)) of thawed white sauce after three days of frozen storage was weighed into 50 mL falcon tubes and centrifuged at 3800 \(\times\) g for 15 min. The supernatant was decanted and weighed (\(W_s\)). Syneresis was evaluated using eqn. (5). For freshly prepared sauces, syneresis was evaluated four hours after it was formulated.

\[\text{Syneresis(\%)} = \frac{W_c - W_s}{W_c} \times 100
\]  
(5)

2.8. Statistical analysis

All experiments were conducted in three replicates. The data obtained were subjected to analysis of variance (ANOVA) using the generalized linear model procedure (PROC GLM) in SAS software v9.4 (SAS Institute Inc., Cary, NC, USA). Treatment means were separated using Tukey’s adjustment for multiple comparisons, while statistical significance was determined at a 5% probability level.

3. Results and discussion

3.1. Granular swelling power

The swelling power of both native and ultrasonicated starch is presented in Fig. 1. Here, ultrasonication is seen to have a significant effect (\(p < 0.05\)) on the swelling power of rice starch. Native starch initially had a higher swelling power compared to S1 and S2 samples but at higher ultrasonication temperature (≥ 45 °C), the swelling power significantly increased. Similar results were reported for sonicated corn starch [13]. Studies have shown that high-intensity ultrasonication causes severe physical damage on the granular surface, by creating fissures or cracks, thereby increasing the ability of the granules to retain more water [31]. The increase in swelling power as a consequence of ultrasonication may be explained by disruption of the crystalline molecular structure of starch and bonding of water molecules to free covalently bonded hydroxyl groups of amylose and amyllopectin [32], and morphological changes as a result of increased cavitation leading to
permeation of water into the granules [33]. Additionally, temperature remarkably increased the swelling power of both native and modified starches. This is also in agreement with studies reported for potato, wheat, corn, and rice starches [18].

3.2. Fat and water absorption capacities

Fat and water absorption capacities of native and modified corn starches are shown in Table 1. Water absorption capacity quantifies the ability of starch to hold water owing to the hydrophilic sites of its polymer chain. It is also important because it is an indication of the starch viscosity. Starch modification through ultrasonic treatment is known to create pores or visible fissures on the surface of the granules [34]. Sonication was seen to have a statistically significant effect (p < 0.05) on water absorption capacity. This effect became more apparent with increasing sonication temperature; the least effect observed in native starch. The significant increase in water absorption capacity can be associated with the movement of water through the pores and a subsequent increase in granular surface area. A similar effect was reported in lentil starch [35], corn, wheat, and rice starches [18]. Ultrasound may also induce a reduction in the amylopectin fraction of starch, which has less affinity for water [36].

Likewise, ultrasonication significantly affected (p < 0.05) the fat absorption capacity of native and modified starches. Fat absorption is the entrapment of oil within the starch granules, following porosity and cavitation created by sonication. These morphological changes may have increased the immensity of granular surface area available for fat absorption [18]. Additionally, the loose helical and hydrophobic inner structure of amylose, forming a crystalline structure may have influenced the increased fat absorption capacity [37]. The results obtained in this study are in agreement with the work reported for ultrasound-modified oat starch [31] and modified wheat starch [38]. On the contrary, the fat absorption capacity of ultrasound-treated starch nanoparticles decreased significantly after size reduction [36]. The authors attributed the decreasing fat content to the reduced particle size and change in the intermolecular structure affecting the hydrophobicity of starch nanoparticles.

3.3. Thermal properties

The results of thermal properties of rice starch as affected by ultrasonication are reported in Table 2. Native starch dispersion witnessed a thermal transition from 59.04 to 70.46 °C, very similar to studies reported in the literature [31]. Ultrasonication temperature was seen to significantly (p < 0.05) reduce enthalpy, onset, and conclusion gelatinization temperatures (Table 2). Within the gelatinization range (60 – 70 °C), our results for starch dispersion treated with the most severe

### Table 1

| Sample code | FAC (±) | S0 | S1 | S2 | S3 | S4 | S5 |
|-------------|---------|----|----|----|----|----|----|
|             | 2.43 ± 0.74 | 3.24 ± 0.31 | 3.64 ± 0.72 | 4.62 ± 0.43 | 6.73 ± 0.38 | 8.36 ± 0.02 |
|             | 2.01 ± 0.08 | 2.05 ± 0.02 | 2.14 ± 0.02 | 2.39 ± 0.15 | 6.74 ± 1.53 | 7.51 ± 0.65 |

Results are mean of three replicates ± standard deviation; across each row, values followed by different letters are statistically different (p < 0.05)

### Table 2

| Sample code | T_0 (°C) | T_p (°C) | T_c (°C) | ΔH(J/g) |
|-------------|----------|----------|----------|---------|
| S0          | 59.04 ± 0.81 | 65.63 ± 0.17 | 70.46 ± 0.01 | 3.85 ± 0.05 |
| S1          | 57.99 ± 0.02 | 64.41 ± 0.31 | 69.38 ± 0.15 | 1.91 ± 0.01 |
| S2          | 57.56 ± 1.55 | 64.49 ± 0.40 | 64.36 ± 0.26 | 1.61 ± 0.01 |
| S3          | 53.70 ± 1.55 | 65.94 ± 0.09 | 71.54 ± 1.04 | 1.04 ± 0.05 |
| S4          | nd       | nd       | nd       | nd      |
| S5          | nd       | nd       | nd       | nd      |

Results are mean of three replicates ± standard deviation; within each column, values followed by different letters are statistically different (p < 0.05); nd: not determined
ultrasonication conditions (S4 and S5), did not show any thermal transition or endothermic peak (Fig. 2), indicating complete gelatinization was achieved [23]. Studies have suggested that the reduction in enthalpy and gelatinization temperature may be attributed to the disruption of the crystalline region of the granules [39]. Moreover, the destruction of the double-helical structure following ultrasonication may also have contributed to reduced gelatinization temperatures [40]. Though our data shows a downward trend in \( \Delta H \) values with increasing ultrasound treatment per the literature, we, however, note that values reported here are smaller in magnitude, in comparison to values ranging between 6.3 and 9 J/g reported for ultrasound-treated corn starch [13] and oat starch [31]. There is strong evidence in the literature to support the assertion that increased heating rate [41] and moisture content [42] lead to elevated enthalpy values. Though many studies reported a heating rate of 10 °C/min, Saeed et al. [43] opined that a lower heating rate, like 2 °C/min used in the current study, can help reduce thermal gradient effect and distinguish transition peaks.

3.4. Flow properties

The upward curve demonstrated a non-ideal or pseudoplastic behavior in all the sauces, characterized by decreasing viscosity in response to rising shear rate or shear stress (Fig. 3). A reverse phenomenon is observed with the downward curves; viscosity was seen to increase when the shear rate was ramped down, signifying thixotropy in the sauces. It is important to note that thixotropy is a time-dependent pseudoplastic behavior of fluids, because it explains why food products, like ketchup, have the behavior of a solid when at rest, but behaves like a liquid (flows) when the container is squeezed. In time-independent pseudoplastic behavior, the fluid recovers along the same path when the shear is removed. In time-dependent fluids, however, when the shear rate is ramped down, the downward curve lags behind the upward curve, leaving an area between both curves called the hysteresis loop.

At rest, starch polymer chains are entangled, forming a stabilized molecular structure. Upon application of shear, the polymer chains begin to disentangle and re-align themselves in the direction of shear. This leads to a reduction in the internal resistance of the sauces and consequently causes the pseudoplastic behavior, also called shear-thinning, observed [44]. Shear-thinning behavior has also been reported for other starch-based sauces [22,23]. The data from the upward curve was adjusted to the Ostwald de Waele model; the \( k, n, \) and PRH values are shown in Table 3. The results obtained show that ultrasonication had a significant effect (\( p < 0.05 \)) on the model parameters, for both freshly prepared and freeze/thawed sauces. The consistency coefficient is a measure of the thickening capacity or viscosity of a material. Increased ultrasonication temperature significantly reduced the consistency coefficient (viscosity) of the modified starch sauces. Studies on the loss of viscosity as an effect of ultrasonication are well documented in the literature [11,19,45]. Reduction in viscosity is an indication of structural disintegration. This can be explained by the collapse of cavitation bubbles formed during ultrasonication, leading to

### Table 3

| Sample code | Freshly prepared | Freeze/thawed |
|-------------|------------------|---------------|
| k           | n                | \( R^2 \) | RMSE | PRH | k           | n    | \( R^2 \) | RMSE | PRH |
| S0          | 31.94 ± 0.10a    | 0.13 ± 0.00a | 0.94 | 6.57 | 28187.90 ± 963.57a | 45.37 ± 1.31a | 0.15 ± 0.01a | 0.97 | 3.44 | 16787.15 ± 118.55a |
| S1          | 40.30 ± 1.12b    | 0.25 ± 0.11b | 0.92 | 6.93 | 23667.95 ± 437.55b | 32.03 ± 3.23b | 0.25 ± 0.00b | 0.95 | 5.03 | 19034.05 ± 596.45b |
| S2          | 35.34 ± 0.81c    | 0.24 ± 0.01bc | 0.92 | 8.47 | 27765.65 ± 874.45a | 26.26 ± 0.00c | 0.19 ± 0.00c | 0.95 | 6.32 | 14427.58 ± 364.08c |
| S3          | 28.01 ± 0.88d    | 0.21 ± 0.01cd | 0.87 | 11.10 | 25026.45 ± 6.15bc | 21.20 ± 0.57d | 0.21 ± 0.01d | 0.90 | 8.88 | 15426.15 ± 216.05cd |
| S4          | 20.92 ± 0.39e    | 0.22 ± 0.02de | 0.91 | 7.59 | 25026.30 ± 217.50bd | 17.82 ± 3.8de | 0.27 ± 0.00e | 0.94 | 8.29 | 17682.93 ± 372.03c |
| S5          | 17.67 ± 1.05f    | 0.22 ± 0.01bdf | 0.92 | 7.71 | 27127.68 ± 646.89a | 10.22 ± 0.27bf | 0.24 ± 0.00bdf | 0.97 | 3.17 | 16030.45 ± 620.65af |

Results are mean of three replicates ± standard deviation; within each column, values followed by different letters are statistically different (\( p < 0.05 \)).
the emergence of high shear forces that can dissociate the covalent bonds between amylose and amylopectin, culminating in structural disintegration and loss of viscosity [46]. Moreover, the high shear produced during ultrasonication depolymerizes the starch molecules, resulting in reduced viscosity [47]. A freeze/thaw cycle further reduced the viscosity of modified starch sauces (Fig. 3) and increased the viscosity of the native starch sauce. The values of the flow behavior index were always less than unity, confirming the pseudoplastic behavior earlier claimed [48]. Similarly, ultrasonication significantly affected (p < 0.05) the percent relative hysteresis. The values show a reduced degree of hysteresis, indicating a more stable structure [49]. Overall, results from flow behavior tests are well in agreement with studies reported for starch [50] and starch-based sauces [22, 23, 51].

### 3.5. Viscoelastic behavior

Strain sweep experiments, used to access the extent of linear viscoelasticity, showed a boundary where the properties of viscoelasticity (G′ and G″) were unaffected by the applied strain (Fig. 4). Such boundary is called the LVE region, and it provides valuable information regarding the structural integrity of the sauces in response to shear; however, more attention is paid to the G′′ curve since it is more vulnerable to structural deformation [52]. Within the strain range evaluated, the values of G′ were always greater than G″, signifying dominant solid-like properties. The results obtained show that a value of 2% strain is well within the LVE region of the sauces, hence this value was used as a controlled variable for subsequent frequency sweep experiments.

The oscillatory spectra (Fig. 5) for both native starch and ultrasonicated starch sauces were very similar to those reported for white starch and ultrasonic-treated starch sauces, a freeze/thaw cycle led to an increase in viscoelastic moduli (Table 4). Syneresis and rearrangement of starch ghost within the continuous matrix may have caused the increased viscoelastic moduli [55]; thereby leading to a more closely packed or denser structure. The loss tangent (tan δ), which is a ratio of loss modulus to storage modulus, is also an indication of structural

### Table 4

Estimates of the Power law parameters and percentage relative hysteresis for the sauces.

| Sample code | G′ (Pa) | G″ (Pa) | tan δ |
|-------------|--------|--------|-------|
| Fresh       | Freeze/thawed | Fresh | Freeze/thawed | Fresh | Freeze/thawed |
| S0          | 43.12 ± 5.15 | 46.87 ± 89.61 | 19.14 ± 45.29 | 17.14 ± 24.85 | 0.44 ± 0.24 | 0.37 ± 0.28 |
| S1          | 31.35 ± 3.09 | 91.22 ± 2.45 | 14.46 ± 2.67 | 23.63 ± 1.25 | 0.46 ± 0.23 | 0.26 ± 0.16 |
| S2          | 39.35 ± 2.67 | 141.65 ± 9.15 | 17.36 ± 1.04 | 32.26 ± 1.04 | 0.44 ± 0.23 | 0.23 ± 0.13 |
| S3          | 28.47 ± 1.76 | 105.3 ± 0.08 | 13.11 ± 0.06 | 25.03 ± 0.10 | 0.46 ± 0.24 | 0.24 ± 0.14 |
| S4          | 32.49 ± 2.68 | 120.75 ± 0.28 | 14.73 ± 0.10 | 29.49 ± 0.10 | 0.45 ± 0.24 | 0.24 ± 0.16 |
| S5          | 45.29 ± 7.56 | 89.61 ± 2.68 | 16.94 ± 0.71 | 24.85 ± 0.71 | 0.37 ± 0.28 | 0.28 ± 0.15 |

Results are mean of three replicates ± standard deviation; within each column, values followed by different letters are statistically different (p < 0.05).
Integrity. For ideal solids, $\tan \delta = 0$, since there is no liquid/viscous portion, $G'$ completely dominates; whereas $\tan \delta = \infty$ for an ideal liquid. A perfect balance between elastic and viscous behavior has a $\tan \delta$ value of unity [56]. Over the frequency range evaluated, $\tan \delta$ values increased, depicting variability in interactive forces holding the gel structure [57]. The values obtained for $\tan \delta$ is $0.1 < \tan \delta < 1$, indicating a viscoelastic behavior and the existence of a weak gel. Unlike viscoelastic moduli, a freeze/thaw cycle decreased $\tan \delta$ values (Table 4). In terms of the magnitude of the viscoelastic property, there was no clear trend with increasing ultrasonication temperature.

### 3.6. Textural analysis

Texture is a critical quality attribute of foods that influences consumer acceptance. In white sauce, starch provides a stabilizing effect (gelation) when amylose leaches out from the starch granule due to increased temperature. Results from textural analysis are shown in Table 5. Textural properties evaluated (stickiness, stringiness, and work of adhesion) were seen to decrease significantly ($p < 0.05$) with increasing ultrasonication temperature. The reduced textural properties observed may be explained by the weakened structural integrity of the starch granules owing to mechanical vibration and cavitation effect [58]. Stickiness and work of adhesion values of mild ultrasound-treated white sauces (S1 and S2) were generally higher compared to their native counterpart. After a freeze/thaw cycle, the values of textural properties (due to increased ultrasound temperature) reported here are useful in the formulation of white sauces, and indeed other food applications, because adherence of foods to the teeth and palate is unappealing [19]. Overall, the data obtained from textural analysis further confirms the results reported in flow behavior measurement and are well in agreement with related studies [59].

### 3.7. Syneresis

Frozen starchy foods experience textural changes that may be explained by amylose and amylopectin retrogradation and undergo syneresis after a freeze/thaw cycle [60]. The phenomenon of syneresis happens when water leaches out from the food matrix after thawing and portends negative quality. In both native and ultrasound-treated freshly prepared sauces, syneresis was not observed, which signifies an initial stable structure. This was reported in other studies conducted on white sauce and has been related to high water absorption capacity [23,29]. Conversely, syneresis was observed after a freeze/thaw cycle. Additionally, the percentage syneresis significantly reduced with increasing ultrasound treatment (Table 5), indicating ultrasonication can be used to enhance the freeze/thaw stability of starch-based sauces. Luo et al. [61] explained reduced syneresis in ultrasound-treated starch to be due to cleavage of starch chains in the amorphous region which caused considerable re-ordering of chain segments. Hence, after a freeze/thaw cycle, more water leached out from native starch sauce in comparison to ultrasound-treated starch sauces. Román et al. [23] reported greater freeze/thaw stability in coarse-grained wheat and rice starch sauces compared to their fine-grained counterparts, indicating that coarse grain starches provided a larger surface area needed to absorb more water.

### 4. Conclusion

The study described in this paper adopted ultrasonication, a physical, inexpensive, and ‘green’ starch modification technology, and investigated its suitability in the formulation of starch-based sauces. Ultrasonication was seen to improve the functional properties of native starch, indicating its suitability in the formulation of sauces. Rheological measurements showed shear-thinning flow behavior, which was well described by the Ostwald de Waele model, while the viscoelastic behavior showed the existence of a weak gel. Ultrasound treatment improved rheological properties and textural characteristics (stickiness, stringiness, and work of adhesion) of starch-based sauces compared to their native counterparts. Moreover, enhanced freeze/thaw stability was also achieved in ultrasonicated starch sauces. Overall, the results of this study show that ultrasound-treated starches can be used in the formulation of sauces and potentially other food products. This is in line with current consumer interest in clean label and minimally processed foods.

### CRediT authorship contribution statement

**Valentine C. Okonkwo**: Conceptualization, Methodology, Data curation, Writing - original draft. **Ebenizer M. Kwofie**: Methodology, Writing - review & editing. **Ogan I. Mba**: Writing - review & editing. **Michael O. Ngadi**: Conceptualization, Funding acquisition, Supervision.

### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

### Acknowledgment

The authors gratefully acknowledge support from the Natural Science and Engineering Research Council (NSERC), Canada.

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