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ABSTRACT
Rheological properties of gelled foods that may relate to the physics of the fluids in the swallowing process of complex food components are determined by ultrasonic spinning rheometry (USR) [T. Yoshida et al., “Efficacy assessments in ultrasonic spinning rheometry: Linear viscoelastic analysis on non-Newtonian fluids,” J. Rheol. 63, 503–517 (2019)]. Through rheological evaluations of thixotropic gelled food, the inaccuracies in standard rheometer data to capture the true-rheological property are discussed first with steady rotational and oscillatory tests; the inaccuracies arise from commonly existing problems that cannot be directly observed in standard rheometers (wall-slip, shear banding, shear localization, elastic instability, etc.). The results evaluated by standard rheometers would be related to the measurements being specific response, depending on the geometry of the measurement device. The USR test discussed here shows the potential to overcome these problems in the rheological evaluation of gelled foods and reflects the advantages offered by USR such as spatial, local, and oscillation cycle measurements; the results with the transient flow curve that has not previously been discussed can be usefully interpreted, and the stability of the food materials in the unsteady shear displayed is of great importance in understanding which rheology indicates the better texture.

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I. INTRODUCTION
Rheology of foods related to the chewing and swallowing processes has been extensively investigated as a safety issue and recently also to improve the quality of life of consumers; for example, usual oatmeal is good for people who have weak chewing and swallowing abilities, especially among older people and infants, but due to very soft texture, chewing is not enjoyable when good taste and good texture are valued. Differently, gelled foods have problems with aspiration even with the better impression arising from eating. Both questionnaires and rheological tests have been used in rheological research, but questionnaire research is affected by personal preferences and rheological tests are not always suitable despite attempts to provide quantitative evaluations; this is because of both the complexity of the chewing and swallowing processes and the rheological properties of foods. Rheological evaluations, therefore, require an understanding of complex properties together with the development of a rheometry that captures both the complex properties and individual and preferential processes of swallowing involved.

Developments in research have tried to evaluate complex-rheological properties, such as shear thinning, yield stress, viscoelasticity, effective viscosity with multiphase dispersion, and other qualities, by creating novel rheometers with different geometries. The precision expressed in the results with recent rheometers (different from measurement accuracy) is ensured with sensitive torque sensors and with the improvements in the geometry of the rotational part, but little attention has been paid to the “physics of fluids” in the measurements by rheometers. This is the reason why there are “common problems of the rheometers” arising from, for example, the
include shear history effects, shear banding, shear localization, wall-slip, elastic instability, and other phenomena. When rheological evaluations are attempted without considering these influences, the obtained results will reflect the specific response as it is dealt with in the measurement device, not exposing or clarifying the true-rheological properties. These problems are caused by the fluid characteristics, and efforts to solve the problems have to be approached from the perspective of fluid mechanics.

To improve the quality of rheometry, utilizing spatiotemporal velocity distributions of the fluid motion is required because the velocity information reflects all the rheological information of a complex fluid. Approaches to achieve this from fluid mechanics have mainly focused on standard rheometers coupled with inner visualization techniques (e.g., Refs. 12–14) and have made it possible to directly identify the common problems (usually invisible). Then, the influence exerted by those problems has been investigated by integrating standard rheometer readings with ultrasonic imaging. Gallot et al. proposed a technique that explains the unstable shear-banding flow of non-Newtonian fluids, and on this basis, Fardin et al. found that the potential impact of inner flow patterns could be observed for both complex as well as Newtonian fluids in the large amplitude oscillatory shear (LAOS).

Research into velocity-profiling rheometry has dealt with time-averaged velocity profiles limited to steady flow states, as one of the examples of velocity-profiling rheometry, Derakhshandeh et al. performed measurements of transient behaviors of thixotropic fluids using a Couette rheometer with a wide gap and ultrasonic velocity profiling (UVP). This was able to detect yielding regions of the fluid from quasisteady velocity profiles, also when the torque measurements may be influenced by existence of wall-slip that could lead to critical errors in evaluations of the rheological characteristics. This leaves problems, as one of the non-Newtonian characteristics involves both shear-rate-dependence in the rheological properties and time-dependence (structural recovery, relaxation, and other aspects), and techniques and measurements must be able to detail time-dependent properties for evaluations of transient rheological properties.

We have developed a novel velocity-profiling rheometry that supplements the rheological evaluations beyond the capabilities of standard rheometers and have termed it ultrasonic spinning rheometry (USR). Based on the ultrasonic measurement of instantaneous velocity profiles, the technique has advantages, such as ease of handling and offers the option of employing it with opaque fluids, and it has been applied to the measurement of foods in general. The basic concept of this rheometry is that velocity profiles are substituted into the equation of motion to estimate the rheological properties, and the potential value of USR with various complex fluids has been established in previous reports.

In this paper, the rheological properties of gelled foods that may relate to flows in the swallowing process of complex food materials are investigated by means of USR. We examine both test materials prepared with a recipe suggested by a food company developed to enable better eating and also intentionally modified recipes. A comparison of results of rheological evaluations of both materials could lead to the establishment of a methodology necessary to understand what is involved in optimum swallowing sensations. Here, as a test material, a milk dessert gelled by the chemical attraction between low-methoxyl (LM) pectin (mainly used as a thickener for foods, such as fruit jam and pastelike sweets) and including calcium ions was chosen, and three test fluids prepared with different recipes were examined by comparative experiments involving the standard rheometer and the USR. In Sec. III, the inaccuracies in the capture of the true-rheological property with standard rheometers are discussed by examining the rheological evaluations of steady rotational and oscillatory tests. After that, a rheology for better texture of complex food materials is discussed by considering the findings obtained by USR (Sec. IV).

II. MATERIALS AND METHODOLOGY

A. Recipe for test materials

The test material, “Fruiche” is a popular dessert in Japan and is available as a basic source material from House Foods Group, Inc., Japan. The Fruiche source includes much fruit pulp, O(1–10 mm) in mean diameter, irregularly shaped and deforming easily under shear stress. Mixed with whole milk, the completed Fruiche dessert changes drastically into a gel due to the aggregating reaction between low-methoxyl (LM) pectin (see the detailed chemical features in Ref. 26) dispersed in the Fruiche source and calcium ions dissolved in the milk as depicted in Fig. 1. Here, the whole milk is provided by Yotsuba Milk Products Co., Ltd., Japan; the nutrient composition is protein 34 g/l, lipid 40.5 g/l, carbohydrate 48.5 g/l, Na 0.39 g/l, and Ca 1.14 g/l. The chained molecules of the LM pectin structures the network linked by the calcium ions, and the structure is commonly called an “egg carton model.” From the perspective of rheology, the complete Fruiche displays highly complex-rheological properties (shear-thinning viscosity, viscoelasticity, yield stress, thixotropy, and modifications of the effective viscosity by multiphase dispersion). These would give rise to the common problems with the standard rheometer mentioned in Sec. I.

The basic source of Fruiche is designed by the company for good texture in chewing and the following swallowing, and to do that, the best weight ratio of the milk to the source is set as 1:1. Here, to evaluate how differences from the recipe affects the rheology, we...
intentionally prepared the Fruiche in different ratios (milk:source = 2:1 and 1:2) from the recipe. After the mixed materials were placed on a 30°-inclined glass plate for 10 min, different behaviors were observed, as shown in Fig. 2. In Fig. 2(a) (milk:source = 2:1), the mix has fluidity and adheres on the surface of the plate. In Fig. 2(b) (1:1), the material had firmly adhered to the glass surface and its shape deformed only a little toward the lower part of the slope within 10 min. In Fig. 2(c) (1:2), the material had slipped on the surface while keeping its shape the same with the feature in Fig. 2(b). These observations represent the simplest rheological test and reflect important rheological characteristics of the test materials qualitatively, such as adhesion, deformability, fluidity, and others qualities, and these features can be used to verify the evaluated properties.

B. Steady rotational and oscillatory shear tests with the standard rheometer

A rheometer with geometry of Taylor-Couette system (Anton Paar MCR-502, parallel-plate geometry PP25) was used in the rheological tests of the completed Fruiche prepared by the different recipes (Sec. II A). As is obvious from Fig. 2, all the three different test materials show large difference in the rheological characteristics. Although a variety of geometries depending on the characteristics of the test materials should be chosen, the same parallel plate was used for all the test materials to evaluate the test materials under the same condition. Otherwise, the geometric difference will affect the rheological evaluations. The test materials are maintained at a constant temperature ($T_0 = 15 \degree C$).

For the steady rotational and oscillatory tests, the following conditions and considerations are maintained to ensure accuracy: (a) one-directional flow (strain), (b) the shear rate is a linear function in the axial direction, (c) the walls are subject to nonslip conditions, (d) the fluid is homogeneous, (e) inertial effects from fluid motion are disregarded, and (f) there are no secondary flows. A thin layer $O(0.1–1 \text{ mm})$ of the test material is required to satisfy the assumptions; however, this thickness is inadequate to prevent the appearance of non-Newtonian behaviors, and the shear rate $\dot{\gamma}$ and shear stress $\tau$ are regarded as apparent shear rate $\dot{\gamma}_{app}$ and an apparent shear stress $\tau_{app}$.

In the ideal condition of oscillatory tests, the fluid response should be determined depending on its rheological characteristics (e.g., viscous, viscoelastic, and elastic) as shown in Fig. 3(a), and Lissajous curves can be derived by considering the shear strain/shear rate and shear stress as horizontal and vertical axes [Fig. 3(b)]. Clearly seen, the rheological characteristics can be distinguished by the feature of the Lissajous curves. Lissajous curves consisting of shear strain and shear stress draw circles for viscous, ellipse for viscoelastic, and diagonal lines for elastic materials. In the nonlinear viscoelastic regime, the fluid response may not show clear sinusoidal characteristics due to occurrences of unexpected phenomena. To understand the invisible phenomena of the fluid with the nonlinearity in the gap of the rheometer, large amplitude oscillatory shear (LAOS) measurements (e.g., Refs. 27 and 28) are performed for the rheological evaluations of complex fluids. The basic concept behind the LAOS measurement is that the shear stress in response to sinusoidal shear deformations are evaluated with the approximations to ensure the accuracy mentioned above. Nonsinusoidal responses depending on the rheological characteristics with complexities (e.g., viscoelasticity, yield stress, and multiphase dispersions) will be obtained using the LAOS. So, it is possible to qualitatively understand the nonlinear rheological response caused by the complex fluid characteristics. The purpose of LAOS measurement in
this paper was to clarify the rheological complexities in the test material, and it will also help to reveal the vagueness in the rheological evaluations of the standard rheometer.

C. Ultrasonic spinning rheometry (USR)

The experimental apparatus is an open cylindrical container made of acrylic resin; the container has 2-mm-thick side walls, 145-mm inner diameter, and is 60-mm high. The container was mounted at the center of a water bath to control the temperature, $T_w$, of the test fluids and to avoid any influence from coreflected ultrasonic waves. Oscillations of the cylinder were controlled by a stepping motor to a set the oscillation angle $\Theta$. Ultrasonic waves were generated by an ultrasonic transducer (resonance frequency 4 MHz and 5-mm active element diameter) mounted with a gap offset 4 MHz and 5-mm active element diameter) was mounted with a gap offset $R\Theta$ (see Ref. 23 for details). UVP-Model Duo (Met-Flow S.A., Switzerland) was used to measure instantaneous velocity distributions. To obtain the azimuthal velocity component, an ultrasonic transducer (resonance frequency 4 MHz and 5-mm active element diameter) was mounted with a gap offset $\Theta$ from the center coordinates of the cylindrical container. With axisymmetric flow and the radial velocity component negligible, the azimuthal velocity $u_\theta$ is calculated from the geometric relation $u_\theta = v_t / \Delta y$. Empirically, $\Delta y = 15$ mm was selected, and further details of the setup for the transducer were detailed in Ref. 19.

Important theoretical considerations for the linear viscoelastic analysis in USR are as follows: assuming that the fluid flows are one-directional and axisymmetric, Cauchy’s equation of motion is given as $\rho \partial u_\Omega/\partial t = \partial \tau / \partial r + 2 \tau / r$, where $\rho$ is the density of the test fluids and $\tau$ is the shear stress. To describe the relation of $u_\theta$ and $\tau$, the Maxwell model, $\tau + (\mu E) \partial \tau / \partial t = \mu (\partial u_\theta / \partial r - u_\theta / r)$, is selected as the simplest expression to represent the linear viscoelastic characteristics, where $\mu$ and $E$ indicate the viscosity and elasticity of the fluid. From the Fourier transform with respect to $t$, Cauchy’s equation and the Maxwell model can be modified as

$$i\omega \hat{u}_\theta = \left( \frac{\partial}{\partial r} + \frac{2}{r} \right) \hat{r}, \quad (1)$$

$$\hat{r} + i\omega \frac{\mu}{E} \hat{r} = \hat{\mu} \left( \frac{\partial \hat{u}_\theta}{\partial r} - \frac{\hat{u}_\theta}{r} \right), \quad (2)$$

where the Fourier-transformed velocity and shear stress are denoted as $\hat{u}_\theta(r, \omega) = \mathcal{F}[u_\theta(r, t)], \hat{r}(r, \omega) = \mathcal{F}[r(r, t)]$, with the angular frequency $\omega$. Considering the cost function

$$F(\mu, E) = \left[ i\omega \hat{u}_\theta - \left( \frac{\partial}{\partial r} + \frac{2}{r} \right) \hat{r} \right]_{\tau = \hat{r}}^2 \quad \text{s.t.} \quad \hat{r} + i\omega \frac{\mu}{E} \hat{r} = \hat{\mu} \left( \frac{\partial \hat{u}_\theta}{\partial r} - \frac{\hat{u}_\theta}{r} \right), \quad (3)$$

$\mu$ and $E$ can be determined by satisfying the optimization problem, determining the $\mu$ and $E$ to minimize the cost function $F$. From Eqs. (1) and (2), the shear stress, the shear rate, and the flow curve can be obtained from the Fourier components obtained from the velocity (see Refs. 21 and 23 for detailed calculations).

According to Yoshida et al., the radial profile of the phase lags calculated from the velocity distributions,

$$\varphi(r) = \tan^{-1} \left[ \frac{2 \left\{ \mathcal{F}[u_\theta(r, \omega)] \right\}}{\mathcal{F}[r(r, \omega)]} \right] - \varphi_{\text{wall}}, \quad (4)$$

can be used to distinguish the rheological characteristics of test fluids: Constant phase lag regimes correspond to rigid rotation, and such regimes may be regarded to occur with fluids having elastic properties or very high viscosity values. Regimes with a changing phase lag indicate fluidization areas and thereby can be regarded as fluids with viscous or viscoelastic properties. For example, radial profiles showing discontinuous variations indicate the existence of boundaries between different regimes of a rheological property.

Simultaneous consideration of both linear viscoelastic analysis and phase lag in the USR offers the possibility to elucidate the rheological properties with the transient behaviors. Such transient-rheological properties cannot be measured by standard rheometers because of the limitations of methodology, and several valuable findings by the USR will be discussed next.

III. RHEOLOGICAL EVALUATIONS BY THE STANDARD RHEOMETER

In this section, rheological evaluations of the test materials prepared with different recipes are examined by the standard rheometer with the parallel plate geometry described in Sec. II B. The steady rotational and oscillatory tests using the standard rheometer measured the rheological properties of the test materials (Secs. III A and III B), and then, inaccuracies with the standard rheometer in measuring the true-rheological properties, arising from the common problems that may occur between the plates, are discussed in Sec. III C.

A. Results of the steady rotational tests

The original source of Fruchi includes numerous strawberry pulps as mentioned in Sec. II A [see Fig. 4(a)]. The sizes of the pulp component vary up to approximately 30 mm. The completed mixture with the dispersed ingredients critically influences the rheological evaluations using the standard rheometer [Fig. 4(b)] as exemplified by jamming of the gap between the parallel plates. Because the rheological evaluations in the standard rheometer should be done without very large ingredients in the test fluids, the pulp components were removed for all of the experiments in Sec. III.

The steady rotational tests were conducted by both shear rate and shear stress sweeps. In the shear rate sweep, the angular speed representing the shear strain rate was logarithmically ramped up from $10^{-1}$ s$^{-1}$ to $5.0 \times 10^1$ s$^{-1}$ [Figs. 5(a)–5(c)]. In the shear stress sweep, the angular speed representing the shear stress was ramped up linearly from 0 to 100 Pa [Figs. 5(d)–5(f)]. The vertical and horizontal axes indicate the apparent shear strain rate and shear stress defined in Sec. II B. The gray and black solid curves represent the results obtained with or without settling for 1000 s after preshearing, respectively.

In the shear rate sweep results in Figs. 5(a)–5(c), the apparent shear stresses, $\tau_{\text{app}}$, at low shear rates showed large differences in the condition of time left at rest: for the 2:1 mix [Fig. 5(a)] in $\tau_{\text{app}} < 4$ s$^{-1}$, the $\tau_{\text{app}}$ immediately after the preshearing is much lower than that after being left at rest, while the $\tau_{\text{app}}$ agrees between the conditions in the 4 s$^{-1} < \tau_{\text{app}}$; for the 1:1 mix [Fig. 5(b)], at $\tau_{\text{app}} < 1$ s$^{-1}$, the $\tau_{\text{app}}$ immediately after the preshearing is much lower than that after being left at rest similar to 2:1 mix, however, the $\tau_{\text{app}}$ immediately after the preshearing around $\tau_{\text{app}} = 1$ s$^{-1}$ showed an
abrupt but slight drop. Then, the curves obtained in the different conditions behave with similar manner at the $1 \text{s}^{-1} < \dot{\gamma}_{\text{app}}$ value. For the 1:2 mix [Fig. 5(c)] and for all of the $\dot{\gamma}_{\text{app}}$ range, there are clearly differences between the conditions in the leaving time; after leaving at rest, the $\tau_{\text{app}}$ shows a plateau at $0.2 \text{s}^{-1} < \dot{\gamma}_{\text{app}} < 1 \text{s}^{-1}$. These results show significant differences that have to be understood for the different rheological characteristic, for example, at the 1:2 mix [Fig. 5(c)], as a typical flow curve, the measured plateau $\tau_{\text{app}}$ indicates the occurrence of wall-slip (or shear banding) that would be caused by a weak thixotropic behavior.

In the measurements with a shear stress sweep conducted to evaluate the critical yield stresses at 2:1 mix [Fig. 5(d)], $\tau_{\text{app}}$ after being left at rest for 1000 s is larger than that immediately after the preshearing. This feature is also observed for 1:2 mix [Fig. 5(f)], while the $\tau_{\text{app}}$ for 1:1 mix [Fig. 5(e)] after being left at rest shows more drastic changes than the others, suggesting that the rheological response for 1:1 mix [Fig. 5(e)] has a stronger slippage on the wall than the others. This would be caused by the adhesion differences of the tested mixture detailed in Fig. 2. It is important to bear in mind that in sweeping the shear stress, these trends represent the wall-slip and are generally considered as showing a "yielding behavior"; this strongly affects the accuracy of the rheological evaluation, as mentioned in Sec. I. Therefore, usually, these critical responses are regarded as the "apparent yield stress" to estimate the specific rheological response when depending on standard rheometer readings.

All together, the rheological characteristics obtained from Figs. 5(a)–5(f), representing the rheological properties of the Fruiche, show large difference depending on the length of time it was left undisturbed, indicating a structure that recovers with time, i.e., thixotropy. Generally, rheological properties of thixotropic fluids would be evaluated as the equilibrium state, or as an equilibrium flow curve. Yoshida et al. reported, however, that especially in such thixotropic fluids, the flow curves obtained from rotating rheometers do not always provide correct evaluations without the influence of shear localization, wall-slip, and other factors, even when the shear stress apparently assumes equilibrium states. This clearly suggests that alternative tests have to be used to evaluate these test materials to capture the true-rheological properties. The results evaluated by steady rotational tests provided some hints to
understand the rheological features. For instance, it is speculated that the strength of the gelled structure would be in the order of the mixtures, 1:1, 1:2, and 2:1, here.

B. Results of oscillatory tests

From the measurements of steady rotational tests (Sec. III A), it is found that the rheological characteristics of the completed Fruiche are time dependent, i.e., thixotropy. To evaluate the equilibrium state in oscillatory tests, long duration measurements at different shear strain amplitudes (1–300%) with the same frequency (1.0 Hz) for the 1:1 mix were conducted (Fig. 6). The \( G' \) and \( G'' \) amplitudes here were measured twice, immediately after preshearing and at 5000 s elapsed after the start of oscillation. Immediately after the preshearing [solid line in Fig. 6(a)], the \( G' \) amplitudes decrease monotonically as the shear amplitude increases, while \( G'' \) amplitudes remain constant. After 5000 s, the two amplitudes are very different to those immediately after the preshearing; the \( G' \) amplitude displays a local maximum at around \( \gamma_0 = 100\% \).

It is hard to guarantee exact linearity of the test material deformation in the oscillatory test shown in Fig. 6(a), because there may be shear banding effects in the gap of the rheometer; the photograph in Fig. 6(b) shows the condition of Fruiche when removing the shaft of the standard rheometer immediately after the long duration measurements at \( \gamma_0 = 50\% \). As shown in Fig. 6(b), a thin layer with \( O(0.1 \text{ mm}) \) thickness was formed, and the diameter was almost the same as that of the parallel plate. Here, the material sticks to the bottom plate forming a thin layer, whereas the material at the top can be deformed because of shearing by the upper plate. This is clear evidence that the Fruiche was influenced by shear banding, resulting in thixotropic characteristics here with the time scale longer than \( O(10^3) \) s. In such cases, the obtained \( G' \) and \( G'' \) should be considered as the “apparent” value.

To estimate the relaxation characteristic of the test materials with different frequencies (0.1, 0.3, 1, and 3 Hz), oscillatory tests in a shear amplitude sweep from \( \gamma_0 = 1\% \) to 300% were conducted after preshearing and leaving at rest for 5000 s. The storage and loss modulus \( (G' \text{ and } G'') \) obtained are shown in Fig. 7(a). For the 2:1 mix, the \( G' \) and \( G'' \) gradually change with respect to the strain amplitude, while more abrupt variations are obtained in the other mixtures (1:1 and 1:2). For the 1:1 mix, an abrupt variation is observed at around \( \gamma_0 = 50\% \) in all conditions at 0.1–3 Hz. For the 2:1 mix, the abrupt variation appears at very similar \( \gamma_0 \) as for the 1:1 mix, there with a frequency lower than 0.3 Hz. Although the amplitude sweep measurements would be influenced by shear history effects such as the oscillations, the abrupt variation would suggest that phenomena invisible here possibly arise from the wall-slip or some other effects, suggesting that there may be significant differences in the relaxation behaviors of the test materials at \( O(0.1–1) \) s.

To understand details of the rheological response during oscillations, LAOS measurements were conducted simultaneously with the \( G' \) and \( G'' \) evaluations in Fig. 7(a); Figure 7(b) shows the changes in Lissajous curves (shear strain vs shear stress) with respect to the oscillation frequency \( f \) (0.1, 0.3, 1, and 3 Hz) and the maximum shear strain amplitude \( \gamma_0 \) (10%, 30%, 100%, and 300%). As shown in Sec. II B, the Lissajous curves have been used to distinguish the rheological properties from the curve shape as three kinds of features representing viscous, viscoelastic, and elastic.

At the high frequency (3 Hz), in Fig. 7(b), the Lissajous curves in each test material suggest an elastic response irrespective of the shear amplitude, and at \( f = 1 \) Hz and \( \gamma_0 = 100\% \), the shapes of Lissajous curves represent more viscoelastic characteristics compared to the others of shear amplitude \( \gamma_0 \). At \( f = 0.3 \) Hz, all the curves for all test materials are less regular with kinks than at higher frequencies; these results can be explained by the qualitative deformation features described in Fig. 2 for the behavior on the inclined glass plates.

For the 2:1 mix, the Lissajous curve at \( f = 0.3 \) Hz and \( \gamma_0 = 30\% \) has a nearly circular shape representing a viscous reaction coexisting with irregular features [Fig. 7(b)]. Here, Fig. 2 shows easily deformable characteristics of the material. For the 1:1 mix, the Lissajous curve at \( f = 0.3 \) Hz and \( \gamma_0 = 30\% \) shows a close to elastic response with some kinks [Fig. 7(c)], similar to the more viscoelastic 1:2 mix [Fig. 7(d)]. The factors causing this difference can be understood by the qualitative differences shown in Fig. 2. The 1:2 mix [the right-hand column in Fig. 7(a)] has a more viscoelastic 1:2 mix [Fig. 7(d)]. The factors causing this difference can be understood by the qualitative differences shown in Fig. 2. The 1:2 mix suggests results influenced by the wall-slip. This suggestion is supported by the results in the steady rotation tests under the shear stress sweep (Sec. III A); the critical yielding behavior of the 1:1 mix was larger than that of the 1:2 mix, as shown in Figs. 5(e) and 5(f). It may be concluded that the gelled appearance in Fig. 2 is accompanied by the wall-slip.

**FIG. 6.** (a) Storage and loss modulus \( (G', G'') \) vs the maximum shear strain amplitude, where the black and gray symbols represent the elapsed time and circle and triangle symbol representing storage and loss modulus \( (G', G'') \), and (b) the photography of the condition of Fruiche after removing the shaft of the standard rheometer immediately after the long duration measurements at \( \gamma_0 = 50\% \).
The wavy features observed on the curves at frequencies lower than \( f = 1 \) Hz at low shear strain amplitudes, and the wavy features of the 2:1 and 1:2 mixtures are larger than those of the 1:1 mix. These features are observed at lower shear strain amplitudes than those resulting in abrupt changes in the Lissajous curves, for example, at the 2:1 mix at \( f = 0.3 \) Hz from \( \gamma_0 = 10\%–30\% \). According to Fardin et al., unstable shear banding would arise from elastic instabilities, when shear stress in flow curves exhibits a plateau with respect to the shear rate under unsteady shear conditions. The geometry used in this study is different from the Taylor-Couette system used in the previous study, and it is of importance to be aware that elastic instability would be triggered by a transient state in the rheological characteristics of complex materials.

C. Summary: The sources of physical inaccuracies in a standard rheometer

The standard rheometer has great advantages in the measurement of viscous or elastic materials without abrupt phase transitions such as yielding behavior. As shown by the experimental findings in Secs. III A and III B, the torque sensitivity in measuring the rheological response will augment inaccuracies arising from bias errors in the understanding of the rheology of test materials in the standard rheometer; the influence from the presence of a depleted layer arising from abrupt phase transitions is very significant here; the data output is a "specific-mechanical response" influenced by the geometry of the rheometer. Many reports have attempted to explain such shear banding problems as mentioned in Sec. I, with most focusing on measuring the shear rate profiles in the gap of the rheometer. However, the problems arising from the standard rheometer cannot be solved by looking at only the influence of elastic instability, even under steady state variations, as there are 3-dimensional unsteady flows, as reported elsewhere. Furthermore, it may not be possible to correct the data obtained from oscillatory tests because the unsteady changes depend on the rheological characteristics of complex fluids. In oscillatory tests, inaccuracies will be significantly magnified as the basic principle of oscillatory tests does not consider the "fluid inertia." Experimental proof of such inaccuracies will be discussed in Sec. IV by comparing the rheological evaluations of standard rheometers with those by USR.

IV. USR TESTS OF THE RHEOLOGY OF GELLED FOOD

In this section, inaccuracies in the rheological properties evaluated by standard rheometers (Sec. III) will be explained using USR.
The USR has the great advantage that it can evaluate materials with dispersed ingredients contained in the original source. Furthermore, based on the equation of motion and rheological model detailed in Sec. II C, rheological evaluations can be realized in unsteady shear conditions by frequency domain analysis (detailed explanations of the algorithm can be found in Refs. 21 and 23).

A. Rheological evaluation on phase lag in velocity distributions

Spatiotemporal velocity distributions measured for the 1:1 mix using UVP are shown in Fig. 8. The vertical and horizontal axes indicate the radial positions normalized by the radius of the container, \( R \), and the spin-cycle period \( t_f \). The contours represent the azimuthal velocity normalized by its maximum value at the cylinder wall (\( r/R = 1 \)), \( U_{\text{wall}} = 2\pi f_0 R \Theta \). The distributions here were obtained at different elapsed oscillation times with \( f_0 = 1.0 \) Hz in the oscillation frequency and \( \Theta = 60^\circ \) in amplitude. The resolutions of the UVP are 25 ms in time, 0.7–0.8 mm in space, and 1.35–1.4 mm/s in velocity. To eliminate the shear history effects, UVP measurements were conducted leaving the test materials at rest for longer than 15 min after stirring the fluid medium.

Immediately after the start of oscillation (\( t_f = 0–1 \)), the test material oscillates in the azimuthal direction almost in phase from the wall to the center of the cylinder as in a rigid body. As the gel structures of the test material are gradually broken and become fluidized due to the shear stress from the oscillating wall, the oscillations of the test material assume small phase lags in the radial direction, for example, at \( t_f = 60 \) (center column), and this is significant after \( t_f = 1800 \). This phase lag development can be used to distinguish the rheological characteristics of test fluids.

From Eq. (4), and after calculating the Fourier components from the velocity distribution, radial profiles of the phase lag for the test materials can be quantified as shown in Figs. 9(a)–9(c). The vertical axis represents the radial position normalized by the cylindrical wall, \( R \), where the horizontal axis shows the phase lag. Here, the range of phase lags are different for the three materials in Figs. 9(a)–9(c) to show the differences in the phase lags. The profile shape does not drastically change at least after \( t_f = 1800 \), and the profiles are assumed to reach the terminal state at \( t_f = 1800 \). The figures show some similarities in the changes accompanying the development of the phase lag with the oscillation period \( t_f \). These suggest shear thinning of the test material with the
shear deformations taking place during oscillations. For the 2:1 mix in Fig. 9(a), the development is more rapid than for the other mixtures in Figs. 9(b) and 9(c). The terminal phase lag profile \(t_f = 1800–1810\) for the 1:2 mix [Fig. 9(c)] shows a knee around \(r/R = 0.7–0.75\), while the profile for the 1:1 mix [Fig. 9(b)] has smoother variation around the radial position. It suggests that the 1:1 mix has a viscoelastic layer functioning as a buffer for the momentum propagation between the liquid and gelled regions. There are also large difference in the terminal phase lag profile between the 1:1 and 1:2 mixtures, that is, the gradient of the terminal phase lag for the 1:2 mix [Fig. 9(c)] near the wall \(0.8 < r/R < 1.0\) shows as much larger than that at the 1:1 mix [Fig. 9(b)]. This can be interpreted as showing a lower viscosity of the 1:2 mix [Fig. 9(c)] than that of the 1:1 mix [Fig. 9(b)].

The rheological characteristics of the different test materials can be summarized from the above experimental results as follows: the rheological characteristic of the 2:1 mix is the one that is most like a liquid and where it is easiest for the gelled structure to break down; in the 1:1 mix, the rheological characteristics show a strong viscoelastic response at the interface between the liquid and gelled regions; in the 1:2 mix, the rheological characteristic has a more abrupt change at the interface between the regions than is the case for the 1:1 mix. These results may then be interpreted to be in close agreement with both the qualitative features of each mixture (Fig. 2), as well as that they display the differences suggested by the standard rheometer measurements (Sec. II), and overall, the rheological characteristics may be described by the terminal shape of the phase-lag and the spatial gradient of the phase-lag.

From the gradient of the phase differences calculated by numerical differential of the phase lag in the radial direction, the fluid motions in the oscillations suggests a division into the liquid and gelled regions. The borderline of the division is now defined as the gradient of the phase-lag and the spatial gradient of the liquid and gelled regions estimated from \(t_f = 1800–1900\) obtained from each material together. Such “transient flow curve” is typical in the linear viscoelastic analysis by USR and is not obtained by standard rheometers. Here, the ranges of \(r/R\) used for the flow curves are from \(r/R = 0.95\) to the radial position corresponding to the physically determined borderline between liquid and gel, \(r/R > 0.6\).

For the 2:1 mix shown in Fig. 10(a), the phase lag and the yielding border described with a dotted curve vary more rapidly, immediately after the oscillation starts than in the case for the other mixtures shown in Figs. 10(b) and 10(c). Here, the gray scale in Fig. 10(a) showing the phase lag is different from the others [Figs. 10(b) and 10(c)] because of the drastic changes in the rheological characteristics. In the 1:1 [Fig. 10(b)] and 1:2 [Fig. 10(c)] mixtures, the variations in the positions where yielding starts show similar changes and vary more gradually, even immediately after the oscillation starts. The phase lags with the 1:2 mix [Fig. 10(c)], however, display larger values than that for the 1:1 mix [Fig. 10(b)].

### B. Results of the linear viscoelastic analysis

In the linear viscoelastic analysis by USR, the values of the rheological properties, such as viscosity, elasticity, shear strain rate, shear strain, and shear stress, are obtained from the measured spatiotemporal velocity distributions (e.g., Fig. 8). Furthermore, by utilizing the gradient of phase-lag as described in Sec. IV A, the physical borderline between liquid and gelled states can be simultaneously estimated for every oscillation cycle. To quantify the rheology of the test materials, the rheological evaluations were performed by calculating flow curves from the viscosity and elasticity obtained via the linear viscoelastic analysis.

Figure 11 shows the flow curves obtained from averaged Fourier components for each of the test materials, with the vertical and horizontal axes representing the shear rate and shear stress, respectively. The gradation of the plots indicates the corresponding radial position in the cylindrical container normalized by \(R\). Each symbol in the plots shows the period of the oscillation cycle, \(t_f = 1.0\) Hz, \(\Theta = 60\), \(T_\text{p} = 15\) °C, and \(U_{\text{rad}} = 477\) mm/s. respectively.

![Fig. 10](image-url)
For the 2:1 mix [Fig. 11(a)], the flow curves have almost monotonically increasing trends with respect to the shear rate, but the shear stress shows larger values locally at the lowest shear rates in the periods from $t_f = 0$ to $t_f = 64–68$. The tendencies of these changes are quite similar to the typical flow curves observed in shear banding fluids, where most of the results were evaluated by steady rotational tests with standard rheometers in combination with a technique to visualize the inside of the gap between the plates. In the terminal flow curve for the 2:1 mix, the gradients of the flow curve are almost constant in the double logarithmic expression in the present range of shear rates. Comparing the initial and terminal flow curves, it is noteworthy that the flow curves overlap over a wide range of shear rates. For the 1:1 mix [Fig. 11(b)], the initial flow curve ($t_f = 0–4$) has a negative gradient at smaller shear stresses, and after some periods of oscillations ($t_f = 4–16$), the flow curve appears similar to that of the 2:1 mix. At the terminal state ($t_f = 1800–1900$), however, the flow curve bends around $\dot{\gamma} = 4 \text{ s}^{-1}$, and this may be physically understood to show that the viscous resistance of the test material changes greatly at a specific critical shear rate. For the 1:2 mix [Fig. 11(c)], the stress stresses in the flow curves from $t_f = 0–4$ to $t_f = 64–68$ are much larger than those of the other materials, and after reaching the terminal state ($t_f = 1800–1900$), the flow curve becomes linear except around the lowest shear rates.

In comparison of terminal flow curves [Fig. 11(d)], the gradient of the flow curve of the 1:2 mix is the smallest of the three mixtures, and the gradient of the 1:1 mix in $4 < \dot{\gamma} < 10 \text{ s}^{-1}$ is in good agreement with that of the 2:1 mix. Furthermore, at shear rates lower than $4 \text{ s}^{-1}$, the flow curve of the 1:1 mix shows the steepest gradient. Relating these differences in the flow curve to the qualitative observations for each mixture (Fig. 2) suggests rheological explanations of behaviors for each of the mixtures. For the 2:1 mix, the mixture behaves as a liquid due to the lower yield stresses; however, the mixture sticks to the glass surface after placing the glass plate on the flat surface. For the 1:1 mix, the main feature is a large viscous resistance in the range of low shear rates, which contributes to sticking to the inclined glass plate (Fig. 2). For the 1:2 mix, the agglutinating property is much smaller than in the other mixtures because of the decrease in viscous resistance after yielding.

Figure 12(a) shows both flow curves for the 1:1 mix in the rheological evaluations of USR and with the standard rheometer, where the flow curves (stars) were calculated from the experiment in Fig. 6. The vertical and horizontal axes represent the shear rate and shear stress, respectively, but the values for the standard rheometer are apparent values, as mentioned in Sec. III. The shear stresses at $t = 5000 \text{ s}$ by the standard rheometer are much larger than those by USR, but at $t = 0 \text{ s}$, the standard rheometer values show good agreement with the $5 < \dot{\gamma}$ values by USR at the terminal state.
Figure 12(b) shows the Lissajous curves, obtained by LAOS at the experiments in Fig. 12(a), and help to elucidate the factors causing the differences in the flow curves. The amplitude of the apparent shear stresses increases with time because of the restoration of the structures present in the mixtures, i.e., thixotropy, however, an increased influence of the viscous characteristics can be observed from the shape of the Lissajous curves at \( \gamma_0 = 50\), 80\%, and 100\%. If the test materials display thixotropy, the rheological properties would shift from somewhat viscous to elastic, but the results of the Lissajous curves show an opposite change. It may be suggested that the rheological evaluations of the standard rheometer are influenced by shear banding causing the occurrence of a depleted layer at the oscillating wall. The apparent shear rate would increase if there is a thin layer causing the shear banding, and the presence of this can be assumed from the changes in the rheological properties displayed in the flow curves obtained by USR.

The depleted layer as the result of shear banding can be seen on the photograph [Fig. 12(a)] taken after removing the shaft of the standard rheometer following the long duration measurements at \( \gamma_0 = 50\% \). As also described in Sec. III B, the material was attached to the bottom plate in the form of a solid structure of a thin layer of \( O(0.1\) mm) thickness, while the material at the top was softer and could be deformed because of shearing by the upper plate. The speculations of a shear banding effect in the gap are strongly supported by this observation.

C. Discussion: The rheology of a better-prepared dessert

From the rheological evaluations by USR based on fluid mechanics, the relations of the shear strain rate and shear stress around the critical shear rate for yielding were quantified. These findings are significant as it is difficult to evaluate the true-rheological properties mentioned in Sec. III with the standard rheometer. The schematic illustrations in Fig. 13 summarize the findings: The figure represents the gelling behavior schematically with the ratio of included Ca\(^{2+}\) and pectin.

When the ratio is low [Fig. 13(a)], for the whole milk:Fruiche source = 2:1, there is a low concentration of structured pectin, and it does not contribute to gelling, but it increases the viscosity. With moderate ratios [Fig. 13(b)] for whole milk:Fruiche source
can evaluate rheological properties from the measured velocity pro-
Figure 13(c), for whole milk:Fruiuche source = 1:2, the yield stress estimated by USR shows larger values, and the viscosity is lower than the mixture with intermediate ratios; this may be interpreted to suggest that the gelled structures were fully developed and further that the material has only a small influence on the viscosity increase arising from the electrical attraction after yielding.

If the recipe suggested by the food company is assumed to pro-
vide the best eating quality (or texture), evaluations of these are given as the rheological property of moderate viscoelasticity and yield stress as described above. Furthermore, the stability of gelled structures with moderate yield stress is also required to be evaluated, and the stability might be supported by the thixotropic behavior as the structures need to be maintained after stirring.

V. CONCLUDING REMARKS

Rheological evaluations on three test mixtures, prepared by adding whole milk to Fruiuche (a commercially available dessert) source in different ratios, were conducted by both a standard rheometer and USR for better understanding of qualities and physical features evaluated as better quality for eating. Using the standard rheometer with parallel plate geometry, the evaluations showed inaccuracies in measuring accurate rheological properties caused by commonly existing problems in the rheometers (wall-slip, shear banding, shear localization, elastic instability, etc.); for steady rotational tests, there are the problems around the critical shear causing yielding or slipping of the test material on walls, and the rheological evaluations showed significant differences between the shear rate and stress sweep data. In oscillatory tests, drastic changes in the storage and loss moduli in shear amplitude sweeps were found, especially at low oscillation frequencies, and the Lissajous curves suggested unexplained problems by LAOS measurements.

To better understand the inaccuracies in measuring the true-rheological properties by the standard rheometer, the rheology representing the textures of complex food materials was suggested using USR based on the equation of motion and spatiotemporal velocity information. In conclusion, the flow curves for all the test materials evaluated by USR showed more reliable rheological characteristics than the data obtained by the standard rheometer. The word “reliable” here means guaranteed precision with physical meaningfulness. Some unclarities in the rheological evaluations using the standard rheometer were found from the experimental results (Sec. II). Not surprisingly, such complex test fluid showing rheological changes drastically against shear deformations must affect the rheological tests. Based on the equation of motion, the USR can evaluate rheological properties from the measured velocity profiles and, in this paper, present true-rheological properties from the perspective of physics of fluids compared to standard rheometers.

The significant findings were represented as follows: transient flow curves were acquired by linear viscoelastic analysis in USR at each period of oscillations and that could not be obtained using the standard rheometer; comparing the rheological evaluations by USR and LAOS measurements, the efficacy of USR was ensured by the obtained Lissajous curves; the qualitative-rheological characteristics of the test materials prepared by different recipes support the validity regarding the evaluated flow curves.

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