**Fabrication and Characterization of Oleofoams Composed of Tribehenoyl-glycerol: Toward a Stable and Higher Air-content Colloidal System**

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**Abstract:** Oleofoams have attracted great interest for many personal care applications due to their favorable physicochemical properties, including texture and detergency. To explore the potential use of mixtures of high-melting fat crystals [tribehenoyl-glycerol (BBB)] and edible oils as low-cost and stable aeration systems, we created oleofoams composed of olive oil and BBB. By whipping BBB/olive oil oleogels following rapid cooling and subsequent heating, we succeeded in preparing oleofoams without emulsifier additives. The mixtures of BBB/olive oil formed oleofoams at BBB concentrations of 4.0–20.0 wt.%. The resultant oleofoams maintained their overrun rates and did not coalescence, even with additional whipping after the overrun rate was maximized. More tightly arranged bubbles and concentrated bubble size distributions, as well as stronger interfacial elasticity, were attributed to increasing BBB concentrations, and thermal results revealed that more heating was required to damage the foam structure. The characteristics of these new oleofoams were closely related to their BBB concentrations, and observed effects were attributed to the network structure of the thickened crystal layer and enhanced gelling in the oil phase.

**Keywords**: oleofoam, fat crystal, aeration, crystal morphology, differential scanning calorimetry, rheology, tribehenoyl-glycerol

**Introduction**

In food and cosmetics industries, foam is preferred for its soft texture and has been applied for many years to whipped cream and facial cleansing foam. Recently, foam has attracted attention as a low-calorie functional food due to its favorable texture and growing public health consciousness, as well as for high-performance cosmetics for its high detergency and soft texture (Kusaka et al., 2018; Pugh, 2016). Both aqueous and non-aqueous foams may be formed depending on whether air bubbles are incorporated into the aqueous or non-aqueous (oil) phases, respectively. Aqueous foams have been extensively researched and widely applied in various fields, such as personal care products, aerated foods, and chemical porous materials (Fameau and Salonen, 2014; Rio et al., 2014). While non-aqueous foams have garnered substantial attention (Binks et al., 2014; Fameau and Saint-Jalmes, 2017; Shrestha et al., 2010), they are comparatively more difficult to stabilize because, the main difference between non-aqueous phase and aqueous phase foaming systems is the surface tension. In aqueous foams, the air–water surface energy can be reduced by the adsorption of amphiphilic substances at the air–water interface. But, the already low surface tension of the air–oil interface can not be reduced. A further difference is that there is no electrostatic force in non-aqueous foams. To prevent the Ostwald ripening and coalescence of air bubbles, foamed emulsions containing various compounds, including fats, emulsifiers, hydrocolloids, and particles, are aerated, and sometimes gelled (Bai et al., 2021).

The generative mechanisms of non-aqueous foams are quite different from those of aqueous foams in terms of the driving force of aeration, surface structures between the air and oil phases, and stabilization of bubbles dispersed in the oil phase during long-term storage. Recently, solid particles and crystals have been used to obtain highly stable non-aqueous foams and various studies have been performed on non-aqueous foams constructed using liquid oil and air-stabilizing materials with solid particles (Binks et al., 2011; Shrestha et al., 2006). The ﬁrst proof of the above concept was established by Mishima et al. (2016), who developed a protocol to produce oleofoams using excess fatty acid crystals as oleogels, enhancing the stability of the foam for drainage. However, the group found that crystal-stabilized oleofoams were temperature-sensitive and rendered completely unstable upon heating to the approximate melting temperature of the crystals. They were also able to track crystal morphologies and polymorphism and revealed that small, ﬁbrous, β-form crystals are necessary for whipping the oil in non-aqueous foams.Further study have confirmed the stabilization of non-aqueous foams by high-melting fat crystals using X-ray diﬀraction and polarized optical microscopy (Du et al., 2021). This study had been studied in more detail the formation of crystal networks during the whipping process and the microstructure of the foam. Therefore, oleofoams can be explored from a more multifaceted perspective, utilizing a variety of manufacturing methods and stabilizers. However, oleofoams still face technical issues in stabilizing.

Binks and Marinopoulos (2017) prepared foams based on triglycerides in the absence of emulsiﬁers. The system was then aerated to produce highly stable oleofoams. They demonstrated the existence of an optimum solid fat content (Ex. Tristearin) for foaming of approximately 20–30 wt.%. Nevertheless, oleofoams have not been shown to become more stable (i.e., no oil drainage, coarsening, or coalescence) for several weeks to months—much longer than for their edible aqueous counterparts (e.g., whipped cream), which generally cannot be stored above refrigerator temperatures. Thus, at present, it is desirable to ensure stability by adding high amounts of solid fats as stabilizers. However, in application, these approaches would be attributable to a determinable on higher costs and on deterioration of textural qualities (i.e., sticky texture or hard to spread) of the products.

In another research, the effects of headgroup size and hydrophobic chain length on oleofoam properties have been extensively studied by Shrestha and Aramaki (2012) using mono- and diglycerol fatty acid esters. They reported that foam stability increased as the hydrocarbon chain length of the fatty acid. Moreover, in a previous study (Matsuo and Ueno, 2021), we reported on the successful preparation of oleogels composed of edible oils (olive oil, vegetable squalene, and caprylic/capric triglyceride) and tribehenoyl-glycerol (BBB) at concentrations of 4, 6, 8, and 10 wt.% via rapid cooling and subsequent heating. The prepared oleogels demonstrated good stability (i.e., no oil drainage and constant rheological properties) over six months at 25 °C. Based on these results, to solve the stability issue of oleofoams, we identified BBB/olive oil mixtures as new stable gelators (i.e., TAGs with even longer hydrophobic chains than the fats that have been studied).

In this study, we explore the physicochemistry of oleofoams composed of BBB/olive oil mixtures for the first time. BBB is a high melting point fat widely used in food and cosmetics and has been shown to form oleogels after crystallization in oil. A series of BBB concentrations (4, 6, 8, 10, and 20 wt%) of oleoforms were successfully prepared and the effects of BBB concentration on interfacial structure behavior and physical properties were investigated by combining the results of microstructure, bubble size distribution, rheological properties, and thermal properties. Importantly, we observed changes in the microstructure of oleofoam at different whipping times and obtained a novel and interesting understanding of the analysis of the relative frequency changes in bubble diameter and overrun as whipping progresses. Our newly developed system exhibited 300% overrun values and retained its air content against agitation using oleogels that formed β-fat crystals without the use of emulsifier additives. This research will greatly enrich the information on oleofoam applications.

**Experimental Procedures Materials**

Cosmetic-grade BBB (Tmα = 53.2 °C, Tmβ′ = 57.7 °C, Tmβ = 60.6 °C, where Tmα is the melting temperature of the α-form BBB crystals, Tmβ′ is that of β′-form crystals, and Tmβ is that of the β-form crystals) and olive oil (Tm = 2.0−4.0 °C, where Tm is the melting temperature) were purchased from Croda Japan KK (Japan). BBB was composed approximately of tri-behenoyl (80%) and di-behenoyl (20%). The fatty acid composition of olive oil are shown in Table 1.

**Preparation of β-fat gel and whipped oil**

The β-fat gel was formed by adding 4, 6, 8, 10, and 20 wt.% BBB to liquid oils at 80 °C in a water bath. When the BBB was completely dissolved, the samples were maintained for 5 min at 80 °C; after heating, they were subjected to a specific tempering procedure to crystallize the BBB. Soon after the formation of the oleogels, aeration was performed using a rotating-type aeration machine (KENMIX KMM-770; Kenwood, Aicohsha Manufacturing Co., Ltd., Japan) attached to a whipper at a rotational speed of 200 rpm. The temperature of the aeration machine was held constant at 25 °C with circulating water connected to a water bath. During each study interval, the oleofoams were photographed with a digital camera to observe changes in appearance and sampled for subsequent measurements. All samples were prototyped in triplicate (N = 3) and stored in an incubator at 5 °C for a maximum of 24 h for subsequent analysis, unless otherwise specified. Overrun rates were defined as follows:

[(weight before aeration − weight after aeration) / weight after aeration]× 100, (1)

and averaged by the number of replicates (N = 3) to derive a mean overrun rate.

**Temperature protocols**

Based on our previous findings (Matsuo and Ueno, 2021), we adopted a specific temperature protocol that involved rapid cooling from 80 °C to 5 °C at a rate of 10 °C/min and subsequent heating to the final temperature (Tf, where Tf is Tm+5 °C) at a rate of 2 °C/min to produce a large number of β crystals and a strong network gel structure (Fig. 1).

**Differential scanning calorimetry**

Differential scanning calorimetry (DSC) experiments were conducted for the mixtures of liquid oils and oleofoams of BBB using a DSC 7000X (Hitachi High-Tech Science Corp., Japan). Each sample (5–15 mg) was sealed in an aluminum pan (50 mL), started at 25 °C, then go to 80 °C (5 °C/min), maintained for 10 min at 80 °C, cooled from 80 °C to 10 °C at a rate of 10 °C/min, maintained for 10 min at 10 °C, and finally heated to 80 °C at a rate of 2 °C/min (Fig. 1); all tests were performed in triplicate. A thermal analysis software (TA 7000, Hitachi High-Tech Science Corp., Japan) was employed to determine the melting (Tm) and crystallization (Tc) temperatures, as well as the melting (ΔHm) and crystallization (ΔHc) enthalpies by tangent method. Values are given as means ± SD from triplicate measurements.

**Microscopic analyses**

The microstructure of the oleofoams was visualized under polarized and non-polarized light at approximately 20–25 °C using a BX53 polarizing microscope (Olympus Co., Japan) with a DP-27 digital camera (Olympus Co., Japan). A small amount of freshly prepared oleofoam was placed on a glass slide with hole at room temperature, covered with a cover slip, and observed using a microscope. Non-polarized light was used to visualize the shape and arrangement of bubbles, while polarized light was used to view the distribution of BBB crystals. Crystal network formation and foam evolution were also observed using a polarized light microscope (PLM). All samples were observed at 20X, 40X and typical images of the same area under polarized and non-polarized light were captured.

**Bubble size distributions**

The distribution of bubble sizes in oleofoams prepared at various BBB concentrations may reflect differences in foam stability. To assess this difference, we produced photomicrographs under 20X immediately after foaming, and bubble sizes were calculated from the images using a digital microscope (VHX-6000; Keyence Corp., Japan). Bubble sizes were evaluated by comparing the images and represented graphically by plotting the size frequency distribution with software included in the VHX-6000. Values are given as means ± SD from triplicate measurements.

**Rheological properties**

The rheological properties of the oleofoams were measured at 25 °C using an AR-G2 rheometer (TA Instruments, USA) equipped with a Peltier system for temperature control. A parallel plate (40 mm) was applied with a measuring gap of 1000 μm. To obtain the variation in the elastic (G′) and viscous (G″) moduli with stress, an oscillatory sweep was conducted at a frequency of 1 Hz, and oscillatory stress from 0.1 to 500 Pa was applied. Variations in both moduli, with frequencies ranging from 0.1 to 100 Hz, were measured within the linear viscoelastic region. The viscosities of all samples were measured at shear rates of 0.1–1000 s−1 under steady shear conditions. For temperature sweep tests, an oscillatory strain of 1.0 Pa and a frequency of 1.0 Hz were applied to measure the variation in G′ and G″ with a temperature change between 0 °C and 80 °C at a rate of 5 °C/min.

**Stability evaluation**

After whipping, a homogeneous sample of each oleofoam was transferred into a 50-mL tube to monitor its stability under the 20 ºC and 40 °C. Samples were photographed at fixed times for at least 30 days to record the foam volume.

**Results and Discussion**

**DSC of BBB/olive oil mixtures**

The melting temperatures (Tm) and the crystallization temperatures (Tc) of the β- and α-form BBB crystals, respectively, are shown in Figure 2. We obtained Tmβ and Tcα by observing the endothermic DSC peaks during heating (2 °C/min) and exothermic DSC peaks during cooling (10 °C/min), respectively (Fig. S1). Since BBB is a mixture of different triacylglycerols (TAGs) with different fatty acid moieties, the β-form DSC melting peaks were broad or split because of the eutectic mixing of multicomponent TAGs. Therefore, we represent the peak temperatures for the broad melting peaks and the mean temperatures for the split melting peaks in Figure S1. We previously determined that different tempering procedures result in very different oleogel properties, mainly owing to differences in BBB crystallization characteristics, specifically crystal size and morphology (Matsuo and Ueno, 2021. Thus, the oleofoams in this study were fabricated using a specific tempering procedure (Fig. 1), which produced a large number of β-form crystals and a strong network structure. The alteration of crystallization temperature and enthalpy of BBB was partly higher depending on the BBB concentration as shown in Figs. 2 and S1. This would be due to the condensing effect. BBB interacts intermolecularly and with olive oil together by hydrophobic interaction of alkyl groups, which subsequently impacted the crystal growth, polymorphic transition and led to the modifications on the physical properties of BBB such as crystal sizes and microstructures.

**Microscopic analyses of oleofoams**

Figure 3 shows the optical and polarized microscopic images and histograms of oleofoams obtained by whipping oleogels prepared at different BBB concentrations (4, 6, 8, 10, and 20 wt. %) for 60 min. Bubbles dispersed in oleofoams at a 4 wt.% BBB concentration mainly appeared as non-spherical (irregular) and often large. However, the oil–air interface was found to be covered with few crystals that were incapable of stabilizing the bubbles, and some crystals were observed in the continuous oil phase. At concentrations of 6 and 8 wt.%, the samples were filled with more tightly arranged bubbles with relatively regular spheres, though they varied in size. A complete bubble structure was formed as sufficient crystals were adsorbed onto the oil–air interface, which was distinctly bright under PLM. The results indicated the presence of Pickering stabilization-based oleofoams, in which crystals act as solid particles (Binks and Marinopoulos, 2017; Gunes et al., 2017). Because of van der Waals forces and intermolecular hydrogen bonding, the irreversible adsorption of crystal particles on the surface prevents the coalescence of bubbles in such oleofoams (Binks et al., 2016). High-magnification images (Fig. 3) illustrate that the presence of non-adsorbed crystals in the continuous oil phase, which increased the bulk phase viscosity and restrained the collapse of the foam structure.

Compared to the relatively low BBB concentration (i.e., <8 wt.%) mixtures, the performance of 10 and 20 wt.% BBB oleofoams was improved; they also contained smaller bubbles, having a more uniform spherical shape, which were surrounded by thicker crystal layers that hindered shape relaxation. Additionally, the gel network was strengthened because of the presence of a sufficient number of crystals in the bulk phase. Distinct changes in the microstructure of 10 wt.% BBB oleofoams, such as the shape change from polyhedral to spherical, were related to the increase in BBB concentration, which enhanced crystal adsorption at the oil–air interface and gelling in the oil phase. Bubble size distributions quantitatively reflected the changes observed in the microscopic images.

At lower BBB concentrations (e.g., 4 and 6 wt.%), most of the bubbles in the oleofoams were approximately 40–120 μm in diameter, and there were many large bubbles, even up to 100–200 μm, indicating that the size distribution spanned a wide range. As the BBB concentration increased, narrower size distributions were obtained, and the most frequently encountered bubble sizes became smaller. In the 10 wt.% BBB foaming system, almost all of the measured bubbles were concentrated between 20 μm and 100 μm in size. A comparison of the average bubble diameter also showed a clear declining trend (i.e., from 79.2 μm at 4 wt.% to 34.4 μm at 20 wt.%) and we found that the bubble diameter of oleofoams decreased markedly with increase in BBB concentration.

The bubble sizes of oleofoams became smaller and more uniform as the gelator concentration increased, because more BBB crystals were adsorbed at the oil–air interface. Jammed crystal layers maintained the structural integrity of the spherical bubbles, and according to the Laplace pressure theory, larger air bubbles are more deformable than small bubbles, such that the decrease in bubble sizes enhanced their resistance to deformation (Petkova et al., 2012). In terms of foam performance, BBB concentrations determined the packed layer of crystals around the bubbles and the viscosity of the continuous phase, which further manifested in the foam structure. BBB is a TAG with a large hydrophobic portion, so the high hydrophobicity of the BBB crystals would adsorb at the air–oil interface and stabilized bubbles. In previous work (Matsuo and Ueno, 2021), the G′of the oleogel increased with BBB concentration (data not shown.), with a value of ~103 Pa (at 6wt.% BBB). As reported by Liu, *et al*. (2021), the air fraction increased with increasing G′ in the range of 103–105 Pa. Based on these studies, it would be considered that the oleogel was able to be aerated at various BBB concentrations (4, 6, 8, 10, and 20 wt. %).

**Whipping ability**

Overrun represents the aeration capacity of oleogels. Figure 4(a) shows the overrun of all oleofoams as a function of whipping time. The overrun of oleofoams throughout the whipping process could be roughly divided into three phases, particularly at BBB concentrations of 6, 8, and 10 wt.%. The first phase involved approximately 0–20 min of whipping, during which the foaming rate increased rapidly and exceeded 100%; however, the stability of the foam structure was poor and most bubbles would rupture after standing for a while. In the second phase, approximately 20−40 min of whipping, the overrun continued to increase but at a lower rate. Nevertheless, the foam stability greatly improved and the foam exhibited an excellent structure due to the adsorption of a large number of BBB crystals to the oil–air interface. At this point, most filled bubbles in the oleofoams were effectively stabilized under the effect of partial crystal coalescence at the interface. In the third phase, over 40−120 min, the overruns of oleofoams with 4 and 20 wt.% BBB still increased, but those with 6, 8, and 10 wt.% BBB stabilized. At these lower concentrations, overrun rates were maintained at nearly 300% from 60–120 min, meaning that stable foam structures of were also maintained. The higher overrun rates of these foams indicated greater hardness and stability, and the combined greater bulk viscosity and thickness of the crystal adsorption layer at the interface may explain the increased foaming ability (Jakubczyk and Niranjan, 2006).

In the early phase of whipping, the bubbles filling the system gradually increased and crystals were continuously adsorbed to the interface. Partial coalescence initially occurred between the crystals at the bubble interfaces, which ensured the foam stability. Excessive whipping did not result in further crystal bridging between adjacent bubbles; instead, small and evenly distributed bubbles were maintained. Previously, Lupi et al. (2016) reported that policosanol oleogels exhibit a gel-like behavior, which the group attributed to the many interactions between the long alkyl chains of the fatty alcohols, while Shrestha et al. (2006) reported that needle-shaped β-form crystals could stabilize foams by covering their surfaces. Based on these studies, we assumed that BBB formed a closely packed intermolecular system in the oils owing to the presence of numerous interactions between the long alkyl chains of behenic acid. From our microscopy (Fig. 4b) and DSC results (Fig. S2, Table S1), it was apparent that BBB crystals were adsorbed at the oil–air interface and created a firm gel network through strong van der Waals interactions from the alkyl groups between the BBB and olive oil. Therefore, stable foam structures were maintained even after excessive whipping without crystal bridging between adjacent bubbles, such that the bubbles began to merge within the oleofoams. It is known that the main component of olive oil is oleic acid. Oleic acid is slightly more hydrophilic than other fatty acids because it has double bonds, and therefore has a higher oil–air interfacial tension, which would be able to stabilize the bubbles generated by whipping. This would cause the particularly high overrun rates at 6–10 wt.% BBB. Oleofoams prepared from 20wt.% oleogels, on the other hand, incorporated less air (overrun ~200%) due to air incorporation being limited by the relatively high elastic modulus of the parent oleogels. On the optical and polarized microscopic images as shown in Fig.3, it would be assumed that oleofoams at 4wt.% BBB incorporated less air due to the crystals in the bulk were too small to trap bubbles, or the viscosity of the continuous phase was too low to hold bubbles.

**Oleofoam rheology**

To explore the effects of different BBB concentrations on the physical properties of oleofoams, a dynamic rheological analysis of the strain, angular frequency, and shear sweeps was performed. The results are shown in Figures 5 and S3. In terms of strain sweeps at a fixed frequency of 1.0 Hz (Fig. 5a), the elastic modulus (G′) was higher than the viscous modulus (G″), confirming the formation of an elastic network. Consequently, increasing the amount of BBB effectively enhanced the strength and interfacial force of the oleofoams, which resulted in a stronger resistance to external stress. As an example, at BBB concentrations of 6 wt.%, oleofoam exhibited higher the G′ of ~3x103 Pa than that of oleogel (~103 Pa, Matsuo and Ueno, 2021). It is revealed that a large increase in G′ after the incorporation of bubbles. This attributed to the air bubbles entrapped by BBB crystals acted as active fillers (Gunes et al., 2017), which can enhance the network rigidity as observed in the oleogels with BBB that was able to capture more air bubbles during whipping process (Figs 3 and 4).

The frequency sweep shown in Figure 5(b) demonstrates that the prepared oleofoams were viscoelastic and exhibited a solid-like behavior. For most samples, G′ > G″, and the crossover where the rheological behavior shifted from the elastic to the viscous state occurred at particularly low frequencies (i.e., 0.01 Hz).

According to the theory underlying whipped creams, the dependence of G′ on the frequency indicates that the structure of the oleofoams was more viscoelastic. The higher the BBB concentration, the higher the G′ obtained, suggesting that less elastic deformation occurred at a given stress. As shown in Figure 5(c), all samples exhibited shear-thinning behavior and a positive correlation was observed between the BBB concentration and the apparent viscosity. The viscosity of oleofoam increased with increasing BBB concentrations, indicating the increase in the strength of foam structure via interactions between the BBB crystals. To further investigate the stability of oleofoams prepared by oleogels at different BBB concentrations, temperature sweeps were performed (Figs. 6 and S4). In the temperature sweep curves, G′ and G′′ decreased gradually as the temperature increased (with heating) and a sharp decrease occurred when temperature rose to a certain point (approximately Tm). In all oleofoams, G′ > G′′ and solid-like behavior was exhibited when T < Tm. Conversely, G′′ > G′ when T > Tm, indicating that the oleofoams began exhibiting liquid-like phase behaviors due to the melting of BBB crystals. The collapse of the oleofoam structure reflects the thermal sensitivity of the system (Fameau et al., 2015). In the cooling curves, abrupt changes appeared in G′ and G′′ within a few degrees, corresponding to the phase transition between the liquid and oleogel states. Additionally, the G′ and G’’ values of oleofoams gradually increased depending on the BBB concentration. Thus, increased BBB concentrations had a greater influence on the rheological properties of oleofoams. The degree of structural recovery also varied depending on the BBB concentration. At >10 wt.% BBB, oleofoams displayed a certain mechanical strength, a good degree of structural recovery, and thermal stability; however, at 4, 6, and 8 wt.% BBB, they did not. We previously explored the crystallization and gelling activity of BBB through the preparation of oleogels and found that rapid cooling led to the crystallization of BBB in the α-form, such that oleogels did not form (Matsuo and Ueno, 2021). In this study, notable interactions occurred between the crystals and influenced the network rigidity. The observed differences were attributed to the rheological properties and degree of structural recovery of the oleogels formed by the BBB crystals.

**Oleofoam stability**

Oleofoams stored at selected preparation temperatures over time are shown in Figure 7. At 20 °C (Fig. 7a), Their volumes remained constant until day 3 but rose from the drainage of clear liquid oil at the bottom of the tube by day 14, the oleofoams primarily contained no crystals. These trends continued until approximately day 21, after which no more oil was released. In contrast, at 40 °C (Fig. 7b), the less viscous foam was stable by at least day 3, but reduced in volume by day 5, mainly due to a collapse of bubbles without the draining of clear liquid oil and contained no crystals. This phenomenon continued up to approximately day 14, after which evaporation no longer occurred. Photomicrographs of the foams stored at 20 ºC and 40 °C are shown in Figure S5. At both temperatures, the oleofoams became marginally rougher, bubbles sizes increased, and their shapes became non-spherical. As As reported by Saha *et. al.* (2020), bulk rheological properties of the oleogel surrounding the bubbles therefore play the dominant role in preventing bubble dissolution. In this work, oleofoam with BBB concentrations of 8 and 10 wt.% exhibited the G′ of the bulk itself remains high at 40°C as shown in Figs 6 and S4. However, oleofoams cannot be stabilized at 20 ºC and 40 °C. We could assume that BBB crystals adsorb at the interface of bubbles, but stabilization by the Pickering mechanism not be sufficient to stop bubble dissolution in this system. Subramaniam et al. (2006) reported non-spherical bubbles are a signature of the jamming of absorbed particles on the oil–air interface. This jamming causes the loss of isotropic surface tension and changes the bubbles from spheres to irregular shapes. Du et al. (2021) also reported that, during whipping, adjacent bubbles in oleofoams merge to form large bubbles, which clogs the interface with a layer of monoacylglycerol crystals. As a result, crystal bridging occurs, leading to a rougher appearance and lower overrun rate. Based on these studies, we assumed that the structure, which supported and merged the bubbles, was damaged by the Brownian motion of bubbles and the weakening of the gel network under higher temperature conditions. Jamming of the absorbed particles at the oil–air interface was promoted, and bubble shape became non-spherical with the merging of adjacent bubbles. As a result, bubble size increased, the oleofoams became rougher, and the air content declined (Figs. 7 and S5). Crucially, no change was observed in the average bubble size with time, demonstrating the superior stability of the coalescence and disproportionation of gas between the bubbles.

**Conclusions**

In this study, we aimed to explore the application of oleofoams, composed of olive oil and BBB, by observing their microstructures during the whipping process and measuring their rheological properties. The BBB concentration significantly affected the microstructure, bubble size distribution, rheological behavior, and thermal properties of the prepared oleofoams, mainly due to the differences in BBB crystal adsorption, which caused them to wrap around the bubbles, and the distribution of crystals in the continuous phase. We also demonstrated that oleofoams with a high overrun rate (~ 300%), mechanical strength, and thermal stability can be formed at BBB concentrations greater than 8 wt.%.

By observing photomicrographs continuously during whipping, we found that excessive whipping did not cause crystal bridging between adjacent bubbles in the prepared oleofoams as the BBB crystals were adsorbed at the oil–air interface and formed a firm gel network via the strong van der Waals interactions of alkyl groups between the BBB and olive oil. Our results can be used to improve the properties of oleofoams by varying their whipping conditions, such as the gelator and whipping time. Here, we provide the first evidence that BBB crystals may cause aeration to produce a stable whipped system in liquid oils without the assistance of other substances, such as solid or liquid crystalline emulsifiers. This finding is of particular importance in the development of commercial applications for oleofoams in low-calorie and soft functional foods and cosmetic products. We believe that this work will facilitate novel technological developments, especially in the food and cosmetic industries.

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**Authorship:** K. Matsuo conceived, designed the study, and wrote the first draft of the manuscript, Y. Fujii. carried out the research and analyzed the data. All authors contributed to and approved the final draft of the manuscript.

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**Figure Legends**

**Fig. 1** Outline of the temperature protocols

**Fig. 2** Melting temperatures (Tm) of β-form and crystallization temperatures (Tc) of α-form BBB crystals in the BBB/olive oil mixtures

**Fig. 3** Photomicrographs of oleofoams with BBB concentrations of (**a**) 4 wt.%, (**b**) 6 wt.%, (**c**) 8 wt.%, (**d**) 10 wt.%, and (**e**) 20 wt.%, under bright field (**left**, 20X) and polarized light (**middle**, 20X; **right**, 40X) microscope and their corresponding bubble size distributions shown as frequency histograms

**Fig. 4** (**a**) Overrun of oleofoams at different BBB concentrations as a function of whipping time. Values are given as means ± SD from triplicate measurements. (**b**) optical microscope images of oleofoams with 8 wt.% BBB

**Fig. 5** (**a**) Strain, (**b**) frequency, and (**c**) flow sweeps for oleofoams containing 4 wt.% and 8 wt.% BBB

**Fig. 6** Temperature sweep curves (a cycle between 0 °C and 80 °C) for oleofoams at BBB concentrations of (**a**) 4 wt.% and (**b**) 8 wt.%

**Fig. 7** Appearance of 10 wt.% BBB oleofoams stored over time at (**a**) 20 °C and (**b**) 40 °C