

Hot Water Dip: A Simple Pre-Storage Method to Improve Areca Nut Tolerance to Cold.

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ABSTRACT: In this work, the effects of the emulsifier concentration, sterilization process, and pH on the properties and stability of the model liquid creamer were evaluated. Applying diacetyl tartaric acid ester of mono- and diglycerides or DATEM at a concentration of 0.3% (w/w) in the presence of 2% (w/w) sodium caseinate produced stable model liquid creamers (10% (w/w) rambutan kernel olein) with a small particle size (Z-average $\approx$ 200 nm) and a narrow size distribution range (PDI $\approx$ 0.24). These creamers were stable regarding creaming and coalescence, having non-flocculated particles and a constant flow behavior index (n) after sterilization using autoclaving (121 °C, 1.1 bar for 15 min) and during storage for 150 days at 25 °C. The model liquid creamers were unstable at pH values near the isoelectric point of caseinate (pH 4–5). However, these were stable after mixing with hot coffee solutions based on no observed feathering or sedimentation. The whitening performance of the model liquid creamers compared well with commercial ones. Non-hydrogenated fat-based model non-dairy liquid creamer was successfully formulated using rambutan kernel olein as a fat component. The results obtained in this study are useful for the possible application of fractionated rambutan kernel fat in food products.

Keywords: Rambutan kernel olein · Non-dairy liquid creamer · Autoclave sterilization · Emulsion stability · Whitening performance

I. INTRODUCTION

Non-dairy creamers, also called coffee creamers, coffee whiteners or coffee lighteners, are formulated systems homologous in functionality and attributes to ordinary dairy products (Golde and Schmidt 2005; O'Brien 2009). These are widely used to whiten beverages such as coffee, cocoa and tea, soften the acidic taste and impart a desired flavor and texture (Golde and Schmidt 2005; Sher et al. 2013). Non-dairy creamers can be in powder, frozen and liquid forms. Dominant advantages of liquid creamers over other creamer forms include that these more closely simulate dairy creamers and provide a homogeneous beverage (Sher et al. 2013). Typically, non-dairy liquid creamers comprise vegetable fat, protein (sodium caseinate or soy protein isolate), carbohydrates (maltodextrin or corn syrup solid), emulsifiers or stabilizers, buffering salt and other food additives (sweetener, flavor and color) (O'Brien 2009; Sher et al. 2013).

The fat concentration in non-dairy liquid creamers can vary from 5 to 18% with melting temperatures of 35–42 °C (O'Brien 2009). The vegetable fats may comprise partially or fully hydrogenated oils with one or more of soybean oil, coconut oil, palm oil, palm kernel oil, corn oil, cottonseed oil, sunflower oil and blends thereof (Sher et al. 2013). However, using hydrogenated oils may have a negative impact on health, particularly partially hydrogenated oils (PHOs) as these oils are a source of trans-fat which has been reported as a factor that significantly increases the risk of cardiovascular disease (Dhaka et al. 2011). Moreover, the hydrogenation process of oils produces a number of spent metal catalyst wastes which could cause environment problems because of their hazardous nature (Hassan and Richter 2002). To overcome these concerns, non-hydrogenated vegetable fats contain natural saturated fatty acids could be an alternative ingredient for non-dairy creamer products.

To extract fat from plant materials, conventional solvent extraction and several emerging technologies, such as ultrasound, microwave, pulsed electric field, and high voltage electrical discharge either as a pretreatment method or assisted extraction are applied (Galanakis 2013). From the previous work, conventional solvent extraction was used to extract fat [approximately 33–37% (w/w)] from the rambutan (*Nephelium lappaceum*) seed kernel. The solvent-extracted fat is a white, soft solid at room temperature and contains oleic acid (C18:1; 36.8%) and arachidic acid (C20:0; 34.3%) as the main fatty acids (Sirisompong et al. 2011). The *in vivo* oral and skin irritation testing of rambutan fat has indicated that it is a non-toxic fat (Eiamwat et al. 2014). From our previous study (Mahisanunt 2016), the solvent fractionation of rambutan kernel fat successfully conducted and obtained the fractionated fats that had specific physical and chemical properties. Fractionation is a technique for changing the physical properties of fat by controlling the crystallization followed by physical separation of the low melting or liquid fraction (olein) from the high melting or solid crystalline fraction (stearin). Solvent (e.g. acetone, hexane, ethanol, and isopropanol) is often applied in fractionation to enhance the separation efficiency and purity of the fat fractions (Gibon 2006). The rambutan kernel olein fraction obtained from double acetone fractionation had a melting range of 39–43 °C and iodine values of 62.3. Its major fatty acids were oleic acid, arachidic acid, gondoic acid and stearic acid. This

olein was soft semi-solid at room temperature which indicated its potential for use as an alternative fat to replace hydrogenated oils (Mahisanunt 2016).

Acceptable non-dairy liquid creamer should be stable during storage without creaming, gelation, sedimentation or color change. In addition, it should retain a uniform emulsion with constant viscosity over time. Therefore, we investigated the effect of the emulsifier concentration and sterilization process on the properties and stability of a model non-dairy liquid creamer prepared with rambutan kernel olein. Besides the stability, the creamers should also disperse quickly with no feathering or oiling off, provide a good whitening capacity, and have a delicate, superior flavor and taste when added to a beverage, such as coffee or tea (Golde and Schmidt 2005; Sheret et al. 2013). Thus, the effects of pH and whitening performance of the model rambutan kernel olein-based liquid creamer were also evaluated by comparison with a commercial liquid creamer. From this study, model liquid creamer may suitably be used to whiten and soften the acidic taste of beverage products, especially coffee. Using rambutan kernel olein to replace hydrogenated fat may be a potential solution for reducing the processed trans-fat in foods.

II. MATERIALS AND METHODS

Rambutan seeds (*Nephelium lappaceum* L.) were kindly donated by Malee Sampran Public Co., Ltd. (Nakhon Pathom, Thailand). Diacetyl tartaric acid ester of mono- and diglycerides (DATEM) made from edible fully hydrogenated rapeseed oil was a gift from Berli Jucker Public Co., Ltd. (Bangkok, Thailand). Maltodextrin [dextrose equivalent (DE) 19] and sodium caseinate [93.7% (w/w) protein] were purchased from Vicchi Enterprise Co., Ltd. (Bangkok, Thailand). Commercial liquid creamer (* 11% (w/w) fat) and dark roast coffee powder were purchased from a local supermarket. Analytical grade dipotassium phosphate and sodium azide (NaN_3) were products of Ajax Finechem (New South Wales, Australia). Acetone and petroleum ether were products of Merck (Darmstadt, Germany) and Avantor Performance Materials (Pennsylvania, USA), respectively.

Extraction of rambutan kernel fat

The rambutan kernel fat was extracted according to the method modified from Sirisompong et al. (2011). After removing the seed shell, the kernels were dried in a hot-air tray dryer at 65 °C for 6 h [final moisture content * 7% (w/w)]. These dried kernels were then ground into fine particles using a blender (MX-T210GN, National Co., Thailand). The ground-dried kernels were put into an extraction bag and transferred into Soxhlet extraction equipment (Glassco Laboratory Equipments Pvt. Ltd., India). The fat was extracted using petroleum ether for 8 h with a kernel-to-solvent ratio of 1:8. After that, the petro-

leum ether in miscella (a solution of fat in solvent) was evaporated using a vacuum rotary evaporator (R-300, Buchi Ltd., Switzerland) at 60 °C and the residual solvent was further removed by nitrogen flushing. The extracted fat was kept at - 18 °C before using for the preparation of olein.

Preparation of rambutan kernel olein

The rambutan kernel olein was prepared using acetone fractionation according to the method of Mahisanunt et al. (2017) with some modifications. The fat was melted to destroy the crystalline form at 80 °C and then mixed with warm acetone (* 60 °C) at a ratio of fat-to-solvent of 1:6 (w/v). The fat-acetone mixture was kept at 25 °C for 24 h to enable high melting fat crystallization. After that, the fat crystals were separated using vacuum filtration through a Whatman No. 4 filter paper. The fat-acetone liquid fraction was kept at 20 °C for a further 10 h. Then, the liquid phase and solid fat crystals were separated. The liquid fraction obtained at this step was collected and the acetone was evaporated using a vacuum rotary evaporator. The remaining solvent was further removed from the residual fat (which was then called rambutan kernel olein (RKOLE) in the present work) using nitrogen flushing. The RKOLE fraction was kept at - 18 °C before being used to develop the model liquid coffee creamer.

Production of model liquid coffee creamer

The model liquid coffee creamers were produced by homogenizing 10% (w/w) RKOLE with 90% (w/w) aqueous phase. The aqueous phases containing 2.5% (w/w) maltodextrin, 2.5% (w/w) sucrose, 2% (w/w) sodium caseinate, 0.15% (w/w) di-potassium phosphate and various concentrations of DATEM [0.1, 0.3 and 0.5% (w/w)] were prepared by hydrating all components in hot deionized water (* 60 °C). Then, these aqueous phases were blended with RKOLE at the same temperature using a handheld homogenizer (Ultra-Turra® xT25 basic, Ika®-Werke GmbH&Co., Staufen im Breisgau, Germany) at 9500 rpm for 4 min. Subsequently, the coarse emulsions were passed through a high-pressure valve homogenizer (APV-2000, SPX Flow Technology Rosista GmbH, Holzwickede, Germany) three times at 300 and 30 bar for the first and second stages, respectively. The resulting hot fine emulsions were cooled to 25 °C in a cold-water bath (* 20 °C) and sodium azide [0.02% (w/w)] was added before keeping at 25 °C in a temperature control incubator for 24 h. For each creamer, the droplet characteristics (zeta-potential, droplets size and distribution) and stability were determined.

Sterilization by autoclaving

This experiment used the model liquid coffee creamers stabilized by 0.3% (w/w) DATEM which had been pre-

pared according to the procedure described above. The creamers were filled into 250 ml glass bottles (Duran, DWK Life Sciences GmbH, Wertheim, Germany) and sterilized using a steam autoclave (Model 25x-2, Wisconsin Aluminum Foundry Co., Inc., WI, USA) at 121 °C and 1.1 bar for 15 min. After cooling, the bottles were stored at 25 °C for 1 and 150 days, after which their droplets characteristics, color and rheology were analyzed.

Effect of pH

The initial pH of the sterilized model RKole-based liquid coffee creamers was 6.4. A sample of 200 ml of each creamer was transferred to a 250 ml glass bottle and then the pH of the liquid creamer was adjusted to the desired pH between 4 and 7 by adding HCl or NaOH solution (1 M). All creamer samples were stored at 25 °C for 24 h prior to analysis of zeta-potential, droplets size and distribution, color, creaming index and microstructure.

Preparation of coffee mix

The mixes between coffee solution and liquid creamer with and without adding sugar were prepared to evaluate the whitening capacity of the model RKole-based liquid creamer. A black coffee solution was prepared in hot deionized water (85 °C) at 1.1% (w/v) according to the manufacturer's recommendation. Then, 15 ml of commercial or one of the RKole-based liquid creamers was added to each 180 ml coffee solution in the absence and presence of 4 g sugar with stirring until homogeneous (* 5 s). The visual appearance, color, pH and droplets characteristics were measured.

Measurement of zeta-potential, droplet size and distribution

The zeta-potential, droplet size and distribution of each liquid creamer were measured using a dynamic light scattering instrument (Zetasizer Nano-S90; Malvern Instruments, Malvern, UK) at 25 °C. Before measurement, the liquid creamers were diluted with deionized water (refractive index, dielectric constant and viscosity were 1.331, 78.5 and 0.8872, respectively) to a droplet concentration of approximately * 0.1% (w/w) for eliminating the multiple scattering effects. The electrophoretic mobility of the oil droplets was measured and then the zeta-potential was calculated using the Henry equation. For the droplet size and distribution, the instrument analyzed the intensity of scattered light from droplets at an angle of 90 using a 4 mW He-Ne laser at 633 nm. The droplet size was therefore reported in terms of the volume intensity mean diameter or the Z-average. While the distribution of droplets was expressed using the polydispersity index (PDI).

Creaming index determination

In the present work, the creaming index (CI) was used to indicate the physical stability of model liquid creamers. Immediately after preparation, 10 ml of each creamer emulsion was poured into a glass test tube (internal diameter & 1.40 cm, height & 16 cm). The total height of the creamer emulsion (H_E) in each tube was measured and all samples were then stored at 25 °C in a temperature-controlled incubator (Sanyo Electric Co., Ltd, Osaka, Japan). After storage, the phase separation of creamers was observed and the height was measured for a bottom serum phase (H_S). The creaming index was calculated as:

$$CI(\%) = (H_S/H_E) \times 100 \quad (1)$$

III. RESULTS AND DISCUSSION

Influence of DATEM concentration on droplet characteristics and stability of model RKole-based liquid creamer

The model RKole-based liquid creamers (2% (w/w) sodium caseinate, 2.5% (w/w) maltodextrin, 2.5% (w/w) sucrose, 0.15% (w/w) di-potassium phosphate and 10% (w/w) RKole) were prepared using different DATEM concentrations [0.1, 0.3 and 0.5% (w/w)]. The droplet characteristics of the prepared liquid creamers are shown in Table 1. The particle size of samples decreased with increasing concentration of DATEM from 0.1 to 0.3% (w/w) (P B 0.05). This decrease was probably due to more emulsifier being adsorbed on the surface of newly developed particles during homogenization leading to their protection from coalescence and the production of small particles. Increasing the DATEM concentration to 0.5% (w/w) had no further effect on the particle size (P [0.05]). This could be attributed to all the lipid particle surfaces in the dispersion being saturated by the emulsifiers (Tcholakova et al. 2004; Yerramilli and Ghosh 2017). The PDI is an indicator used to determine the particle size distribution of the dispersion system. This value ranges from 0.0 (perfectly uniform system) to 1.0 (highly polydisperse system), with values less than 0.3 being considered as optimum values and indicating a monomodal distribution in the system (Danaei et al. 2018). The PDI values of all liquid RKole model creamers prepared in the present work were less than 0.24, indicating monodispersal with a narrow size distribution of samples. No significant difference was noted between the PDI values of the RKole creamers produced at different DATEM concentrations. Similarly, there was no significant effect of DATEM concentrations on zeta-potential of RKole creamers (Table 1). The zeta-potential provides information on the adsorption of ionic compounds on the droplet surface and can be used to predict the physical stability of an emulsion system. Normally, emulsions that have high absolute zetapotentials (C 30 mV) tend to have good physical

stability. The zeta-potential values of all the RKOLE creamers were negative and ranged from - 53 to - 54 mV, indicating acceptable physical stability. This negative charge was caused by the adsorbed DATEM and sodium caseinate at the interface (Perugini et al. 2018; Yesiltas et al. 2018). DATEM is anionic emulsifier that produces a negatively charged zeta-potential which is attributed to the presence of negative groups in the molecules (Sahafi et al. 2018). Typically, for the caseinate stabilized emulsion, when the pH of the emulsion is higher than the isoelectric

point or pI value (* pH 4.6), the oil droplets surface became negatively charged. Therefore, all the model RKOLE creamers at the preparing pH (* pH 6.3–6.4) had negative values of surface charge. The high negative zeta potential values in the present work was consistent with the values reported in previous works which ranged from - 40 to - 70 mV for caseinate alone and for mixed caseinate/small molecule surfactants (Perugini et al. 2018; Yesiltas et al. 2018).

Table 1 Particle size (Zaverage), polydispersity index (PDI), zeta-potential, and creaming stability of RKOLEbased liquid creamer prepared with different DATEM concentrations

Characteristics	DATEM concentration [% (w/w)]		
	0.1	0.3	0.5
Particle size (nm)	199.81 ± 2.91 ^a	189.28 ± 1.94 ^b	187.89 ± 3.01 ^b
PDI ^{ms}	0.238 ± 0.065	0.209 ± 0.039	0.187 ± 0.060
Zeta-potential (mV) ^{ms}	- 53.49 ± 0.82	- 53.58 ± 0.91	- 53.65 ± 0.29
Creaming stability ¹	+, -	+, +	+, +

^{a,b}Different letters in the same row represent significant difference ($P \leq 0.05$)

^{ms}Within the same row represent no significant difference ($P > 0.05$)

¹Creaming stability of RKOLE-based liquid creamer after storage at 25 °C for 24 h and 28 days, respectively. +, stable; -, unstable

The physical stability of the RKOLE liquid creamers was also observed visually during storage. All creamer samples appeared homogeneous with no phase separation after storage for 28 days. However, a small lipid layer on the surface was observed in the creamer prepared with 0.1% (w/w) DATEM (data not shown). This was consistent with the above results which showed that RKOLE creamer stabilized with 0.1% (w/w) DATEM had largest particle size and highest PDI (Table 1). All results in this experiment showed that using a DATEM concentration at 0.3% (w/w) could produce a stable model liquid creamer having both a small particle size and a narrow range of size distribution. Therefore, this emulsifier concentration was used in subsequent experiments.

IV. CONCLUSION

This study suggested that rambutan kernel olein has potential to be used as the fat component for fabricating non-hydrogenated fat-based non-dairy liquid creamer. The proper concentration of DATEM as emulsifier to produce a stable model creamer with desirable characteristics was 0.3% (w/w) in the presence of 2% (w/w) sodium caseinate. Sterilization could be applied to prolong the storage stability of liquid creamer without any effect on emulsion stability. The model liquid creamer had a more whitish appearance than the commercial one. Coffee solutions mixed with the

model liquid creamer did not have any feathering and were similar to that of the commercial creamer, though they did have a higher whiteness level. The study results indicated the efficient whitening power of the model liquid creamer. However, more information regarding the taste and flavor of coffee mixes containing rambutan kernel olein-based liquid creamer is necessary and future work is required. Besides, the effect of antioxidant especially natural extracts, such as plant polyphenols on the oxidative stability of this model liquid creamer may also need to study.

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