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Original article

Effect of hydrophilic and hydrophobic emulsifiers on physio-chemical, structural and water distribution properties of meat-based cookies

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ABSTRACT

Meat-based products are highly susceptible to weak gel formation upon heating. In this study, the effect of different emulsifiers on the rheological, pasting and microstructural properties of meat-based sugar-snap cookies was investigated. Cookies were processed with the addition of freeze-dried fish meat (1% w/w) and emulsifiers (0.5% w/w of wheat flour). The addition of emulsifiers especially diacetyl tartaric acid ester of mono- and diglycerides (DATEM) increased storage modulus (G′), a viscous behavior, compared to control and other emulsifiers. Expansion and hardness were significantly ($P < 0.05$) improved in DATEM and sodium pyrophosphate (SPP) samples compared to the control indicating the compact structure of starch-protein and emulsifiers molecules. The gelatinization characteristics particularly peak and final viscosities were significantly ($P < 0.05$) increased in DATEM samples with high water holding capacity (WHC) suggesting the strong interaction among starch-protein and emulsifiers. Low-field magnetic resonance (LF-NMR) analysis exhibited that T_{21} relaxation time, an indication of tightly bound water, was also highest in DATEM sample while unbound water (T_{23}) was highest in control and sodium dodecyl sulphate (SDS) samples. Scanning electron microscopy (SEM) analysis revealed that DATEM containing samples were dense and uniform whereas wide holes were found in control and SDS samples. It was concluded that DATEM could be an effective emulsifier with improved physio-chemical, structural and pasting properties of meat-based cookies. The study will open a new window for the best use of suitable emulsifiers for meat-based cookies, however, future studies are needed to check their storage stability.

1. Introduction

The baking industry has received the immense attention of food technologists owing to its vast variety regarding taste, nutrition, aroma, cost competitiveness and long shelf life (Galić et al., 2009). This versatility depends on the starch-protein network, hydrophobic-hydrophilic interaction and micronutrients, which have a substantial influence on the qualitative features of bakery products (Nogueira and Steel, 2018). Thus, understandings of dough functioning and starch-protein network are critical points to understand the quality and functional features of bakery foods.

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Bakery foods have specific properties, such as expansion, texture, color, oil/water absorption index, range of gelatinization, and water movement inside the matrix (Zhang et al., 2020). These qualities are heavily influenced by the type of proteins, starch, micronutrients, and entrapped air in the final product (Zhang et al., 2021). Thus, studying these differential features is inevitable in the baking industry to develop new ingredients (Nawaz et al., 2021).

Wheat gluten protein and wheat starch, major ingredients of the baking industry, have been widely studied along with additional ingredients, such as buckwheat flour (Torbica et al., 2012), waxy rice starch (Giuberti et al., 2017), meat (Magalhães et al., 2020; Nawaz et al., 2019) and food additives (Nogueira and Steel, 2018). Among these ingredients, food additives are added to compensate the viscoelastic qualities of wheat gluten flour, which can cross-link when heated. However, the addition of non-glutinous proteins, such as meat proteins, may result in different viscoelastic and physiochemical properties including expansion and/or weak gel upon heating of myofibril proteins (Nawaz et al., 2021). Although, in bakery products, expansion is desirable but proper structure and shapes are important for a desirable sensory appearance. In addition, at high temperatures, proteins from meat may interact with other ingredients and produce some hazardous compounds, such as protein oxidation and heterocyclic amines (Nawaz et al., 2023). Moreover, due to the high fat and sugar used in the bakery, these proteins may also cause Maillard reaction, advanced glycation end-products and lipids oxidation. Thus, the addition of meat-based proteins is challenging for the bakery industry which needs the serious attention of scientists for the development of a proper network and structure.

Functional food hydrocolloids, for instance, antioxidants (Caleja et al., 2017), structurally different starches (Zhang et al., 2021), especially, emulsifiers have become a prominent practice in bakery and snack foods to improve the physicochemical and structural characteristics (Nawaz et al., 2019). Previous research have suggested that emulsifiers can be added during thermal processing to improve physicochemical characteristics and affect the interface of hydrophilic and hydrophobic phases of dough and meat-based products (Nawaz et al., 2023; Nawaz et al., 2019; Ding and Yang, 2013; Sheikholeslami et al., 2021). Furthermore, the rheological properties of dough that have been changed by the addition of emulsifiers have been thoroughly investigated (Nawaz et al., 2019). Low-field nuclear magnetic resonance (LF-NMR) is extensively used to compute the water mobility inside the matrix as well as to analyze the state and interaction of water with other macromolecules in meat-based products (Shao et al., 2016). The distribution and state of water in thermal processing have a considerable impact on the product's qualitative properties including water holding capacity (WHC), adhesiveness, hardness, tenderness, and appearance (Shao et al., 2016).

The addition of meat in the bakery industry is a novel and encouraging approach, however, proteins from meat exhibit weak gelling properties upon heating that affect the structure, rheology and physiochemical properties of the end product. Thus, the application of emulsifiers could be a promising approach, which may interact with different components in order to develop a compact structure. The capacity of emulsifiers to bind gluten protein, which promotes dough stability and builds a proper starch-protein network are the features of emulsifiers that improve the functional attributes of bakery products (López-Tenorio et al., 2015). Emulsifiers having different hydrophobic-lipophilic balances (HLB) attach to the hydrophobic end of proteins and promote gluten aggregation in the dough, which results in a superior product texture. In addition, the type of emulsifiers (ionic or covalent, low or high HLB) is of high importance for better compatibility and effectiveness (Hasenhuettl and Hartel, 2008). Furthermore, proper dose is also important for proper structure as well as health point of view. Therefore, studying the type and doses of emulsifiers is crucial when adding in a new product, such as meat-based cookies.

The present study aimed to investigate the effectiveness of different emulsifiers in meat-based cookies with respect to the physicochemical, rheological, and structural characteristics. Cookies were prepared by the addition of freeze-dried fish meat along with different emulsifiers. The degree of gelatinization was measured by determining the pasting properties of cookies. Low-field nuclear magnetic resonance analysis of meat-based cookies was studied to investigate the water distribution inside the matrix. The findings of this study will provide a platform for the appropriate type of emulsifiers to be used in meat-based cookies for a proper structure and physicochemical properties.

2. Experimental section

2.1. Materials and reagents

Three food-grade emulsifiers including sodium dodecyl sulphate (SDS), diacetyl tartaric acid ester of mono- and diglycerides (DATEM), and sodium pyrophosphate (SPP) were purchased from Sigma-Aldrich Inc, Natick, MA, USA. Wheat flour (12% protein) from Jinsha River Industry Ltd., Chengdu, Sichuan, China, sugar (Guangzhou Fugidoha Foods Co., Guangzhou, P.R. China), salt (Guangdong Multivarietal Salt Co., Guangzhou, P.R. China), and shortening (B & G Foods, Parsippany, NJ, USA) were obtained from a supermarket of Yangzhou, Jiangsu, P.R. China. Before using raw material, the quality and shelf life of all raw materials were assessed as per supplier's information. Fish minced meat from grass carp (*Ctenopharyngodon idella*) was obtained from a local supermarket in Yangzhou and freeze-dried at −70 ℃ for 72 h. The composition of whole fish powder was: protein 72%, fat 11%, and ash 14%. The supplementary reagents utilized in this investigation were of analytical grade and not purified further.

2.2. Preparation of cookies

Cookies were made following the AACC procedure (method No.10–52) after slight modifications adopting the method of the previous study (Zhang et al., 2021). For a control sample, 48 g, 2.4 g and 0.8 g of sugar, non-fat dry milk powder, and $NaHCO_3$, respectively, were combined in a micro-mixer (Bear Kitchen Aid, SJJ-B10T2) to form a creamy texture through the addition of 24 g Cirsco® vegetable shortening for 2.5 min at medium speed, followed by scrap blade cleaning after each stage. Then, with the appropriate amount of water, 8 mL solution A (79.8 g $NaHCO_3$ in 1 L water) and 4 mL solution B (101.6 g NH_4Cl and 88.8 g NaCl in 1 L of water) were mixed with 75.2 g of creamed mass. This creamed mass was mixed thoroughly at medium speed for 3.5 min while 80 g wheat flour (14% moisture content) and 1 g freeze-dried fish meat were added followed by blending for another 30 s. After scraping the dough from the blades and bowl, it was sliced into small pieces and placed on an oily baking piece. A stainless-steel shaper was used to cut the sheet to a thickness of 15 mm and a diameter of 55 mm. The dough for the molded cookies was baked for 12 min at 206 °C. The baked cookies were left to cool for 30 min before being stored in sealed bags of plastic for subsequent analysis. Four treatments including control and 3 emulsifiers (0.5% w/w of wheat flour) were prepared. The emulsifier dose (0.5% w/w of wheat flour) was adopted from our previous study (Nawaz et al., 2019) as excessive dose disrupts the structure. The experiment was repeated three times and all analyses were also done in triplicates.

2.3. Rheology of dough

A Hybrid Rheometer (HR1, TA Instruments, USA) was used to analyze the dynamic rheological properties of dough, which was fixed with parallel plate geometry of 40 mm diameter, as described previously (Nawaz et al., 2021). Between parallel plates, cookie dough was loaded and gap was minimized to 1.5 mm. To avoid moisture loss, the edges were covered with paraffin oil. After 55 min of loading, the measurements were taken to relax the usual tensions that occurred during loading. The linear viscoelastic area of the dough was measured using a strain sweep (0.01–100 percent) at a fixed frequency of 1 Hz. Rheological characteristics, such as storage modulus (G'), loss modulus (G''), and tan θ were determined in the linear viscoelastic zone at 25 °C with a frequency range of 0.1–10 Hz at 1.5% strain. For accuracy, measurements were taken in triplicates, and graphs were created by computing the average of triplicates.

2.4. Measurement of expansion and WHC

Linear expansion of cookies was calculated by adopting the methodology of previously published study (Nawaz et al., 2019). Briefly, the diameter and length of cookies were calculated before and after baking of cookies. For each treatment, at least six cookies were selected for measurements and mean values were calculated. WHC of baked cookies was also calculated by the previous study (Nawaz et al., 2019). The experiment was done in triplicates for the calculation of mean values.

2.5. Measurement of textural properties

The TA-XT2 plus texture analyzer (from Stable Micro System, London, Uk) was applied to calculate the texture of cookies. The methodology of Zhang et al. (2021) was used by means of HDP/3PB probe under the subsequent conditions: probe travel distance is 20 mm, trigger force is 5 g, and test speed is 1.0 mms^{-1}. Each treatment's hardness (N) was measured at the time of break-in six cookies for the calculation of mean values.

2.6. Pasting properties of baked cookies

Pasting properties, including peak and final viscosities, trough, breakdown, setback and pasting temperature were calculated using Rapid Visco Analyzer by adopting the previous methodology (Nawaz et al., 2019). For each sample, all calculations were performed in triplicate, and mean values were determined.

2.7. Low-field nuclear magnetic resonance (LF-NMR) analysis of cookies

LF-NMR analysis of cookies was performed following the previous methodology (Nawaz et al., 2019). Briefly, 1x1 cm cross-section pieces of baked cookies were placed in NMR tube and the following sets were used to measure the water distribution properties of cookies. The measurement of relaxation time T_2 was done at 20 MHz and 25 °C, which was done using sequence-based Carr–Pur cell–Meiboom–Gill (CPMG). The represented peaks were proportional to the amounts of protons in the population.

2.8. Microstructure of baked cookies

Microstructural characteristics of cookies were witnessed by the previous study (Nawaz et al., 2021). Concisely, freeze-dried cross-sectional pieces of baked cookies were used for SEM analysis. SEM analysis was done on SEM with a model of S-4800-II, which came from Hitachi, Tokyo, Japan. The freeze-dried pieces were coated using platinum before analysis. All micrographs were taken at an accelerated voltage of 10 kV.

2.9. Statistical analysis

The whole experiment was done in triplicates to reproduce the results. The statistically significant difference was determined by Duncan's Multiple Range test along with a one-way analysis of variance (ANOVA) at a significant difference of $P < 0.05$. SPSS 21 (SPSS statistics for Windows, Armonk, IBM, Carp., NY, USA) version was used to conduct statistical analysis.

3. Results and discussion

3.1. Rheological properties of dough

The rheological characteristics of dough in terms of frequency sweep test are displayed in Fig. 1. The findings disclose that all emulsifiers showed solid behavior as $G'>G''$, however, G' of DATEM was greater than all other samples indicating that DATEM increased the viscosity of dough. Similar results were conveyed by an earlier study when DATEM was added to dough (Ribotta et al., 2004). On the other hand, SPP showed the lowest G' even when compared with the control. A greater value of G' indicates the stronger gel network that was obvious in the case of DATEM, which suggests the improved tolerance to deformation. On the other hand, the decreased G' in the SPP sample might be due to the weak interaction of emulsifiers with starch and protein, which resulted in a loose structure and weak gel. An increasing trend was observed in the values of loss modulus (G''), which is an indication of the viscous properties of dough. The highest value of G'' was observed for DATEM followed by SDS and control. However, all values of G'' were less than G', which suggests the dominancy of viscous behavior over elastic behavior. The improved rheological properties with the addition of DATEM could be due to its interaction with starch, especially with the molecules of linear amylose, but also with amylopectin leading to the formation of complexes (Kohajdová et al., 2009).

The findings of rheological characteristics propose that samples prepared with DATEM and SPP promoted crosslinking whereas control and SDS might develop entangled networks rather than cross-linking. Thus, it can be inferred that DATEM might interact with the hydrophobic part of the protein, which resulted in the unfolding and crosslinking of the protein. Therefore, it is probable that the viscosity of the dough was increased because of cross-linking. However, further studies are needed to investigate the crosslinking of dough to understand the complete mechanism.

The results of tan δ are also shown in Fig. 1C. It has been established that higher values (close to 1) are believed to be solid behavior whereas lower values represent thinning or less viscous behavior. From the results, it is obvious that the value of tan δ of DATEM was highest followed by SPP and SDS, which suggests that these emulsifiers showed thickening behavior as compared to the control sample. Thus, it can be said that DATEM and SPP can be good choices regarding the best use of emulsifiers in meat-based sugar snap cookies.

3.2. Expansion and WHC of cookies

The results of expansion and diameter of cookies are disclosed in Fig. 2A, expansion was significantly ($P < 0.05$) increased in control followed by SDS and SPP. The lowest expansion was recorded in DATEM samples. Regarding diameter, it was significantly highest in control and lowest in DATEM. The increased expansion in the control sample could be due to the presence of fish meat that behaves as a weak gel upon heating (Tobin et al., 2013). On the

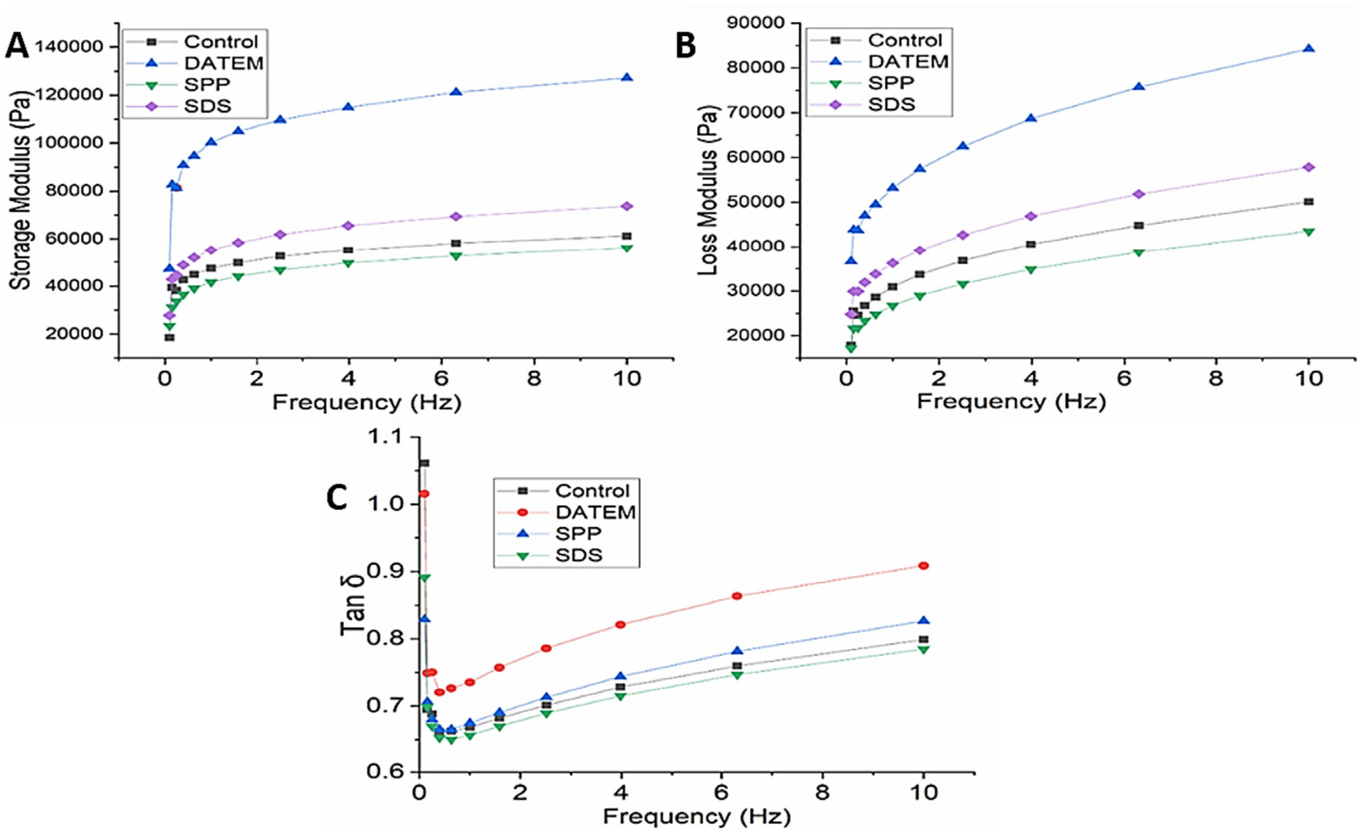

Fig. 1. Rheology of cookie dough prepared with various emulsifiers. A: storage modulus, B: Loss modulus: C: Tan δ.

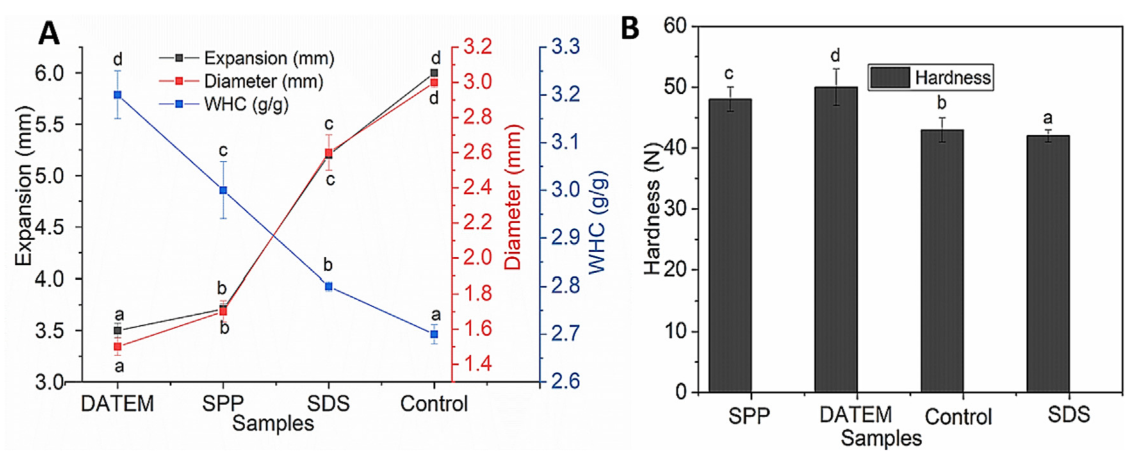

Fig. 2. **(A)** Expansion, diameter, and water holding capacity (WHC) of cookies prepared with various emulsifiers. SPP means sodium pyrophosphate, DATEM means diacetyl tartaric acid ester of mono- and diglycerides and SDS means sodium dodecyl sulphate. Reported values are the mean and standard deviation of 6 replicates for expansion and diameters while triplicates for WHC. Small letters (a, b, c, and d) over error bars represent the significant difference between control and other treatments using one-way ANOVA and Duncan Multiple range test at a level of $P < 0.05$. **B)** Texture profile analysis (hardness) of cookies prepared with various emulsifiers. Small letters (a, b, c, and d) over error bars represent the significant difference between control and other treatments using one-way ANOVA and Duncan Multiple range test at a level of $P < 0.05$.

other hand, the decreased or controlled expansion in DATEM samples could be due to the ability of DATEM to bind the fish meat and starch, which prevented them to associate with gel, thus, a compact structure or proper shape was obtained (Nawaz et al., 2019). It is worth describing that freeze-drying meat used in the present study is equivalent to five times fresh meat. Due to the viscoelastic qualities of fish meat, a higher proportion of fish meat could not be used, notably in cookies, because its shape and structure were disrupted when heated. Even with the addition of 1% dry fish meat, expansion increased, so we added emulsifiers to see how different emulsifiers affected the smooth structure and physio-chemical qualities. In cookies, higher expansion results in a loose structure shape, which affects the sensory appearance of cookies. Thus, the addition of emulsifiers especially DATEM and SPP can control the excessive expansion, which would improve the structure and shape of meat-based cookies. The use of emulsifiers controlled the linear expansion, which controlled not only the viscoelasticity of fish meat but also the extreme gluten protein unfolding when heated. The DATEM made the dough more resistant to collapse, resulting in less expansion. Furthermore, because DATEM may bind gluten protein, the controlled expansion in those samples could be owing to significant interactions between proteins, starch, water, and emulsifiers.

WHC of the end product is a key parameter, which predicts the gelling behavior of the product. It also determines the extent of starch-protein-water interaction in the matrix. Fig. 2A reveals that WHC of cookies increased with the addition of emulsifiers and it was significantly ($P < 0.05$) increased in DATEM tailed by SPP and SDS. On the other hand, WHC was found to be lowest in the control sample, which indicates less interaction of starch-protein and water. It also indicates insufficient gelatinization in the control sample. The reported outcomes are in line with the previous study, which also showed that WHC was increased with the addition of DATEM in fried meat snacks (Nawaz et al., 2019). The increased WHC of DATEM samples could be due to the different HLB of DATEM as compared to SPP and SDS. This could also be explained in such a way that DATEM consists of hydroxyl groups that interact with water, which promoted the increased WHC. In addition, DATEM is an ionic emulsifier that form hydrogen bridges with amide bonds proteins, including myofibrillar and gluten proteins (Gómez et al., 2013). On the other hand, SPP has also high ionic strength and it is probable that the addition of SPP also caused the increased WHC of protein as reported by previous studies (Carneiro et al., 2013; Shao et al., 2016).

3.3. Textural properties of cookies

Textural possessions are of high importance concerning gel properties, sensory and consumer acceptability. Generally, cookies with high hardness are preferred. As shown in Fig. 2B, the results showed that the hardness of cookies was significantly ($P < 0.05$) higher in DATEM followed by SPP compared to the control. The control sample was perceived as fragile indicating that the addition of meat resulted in a loose structure and weak interaction between water, starch, and proteins. This was also evidenced by a previous study when myofibrillar proteins were heated, which formed a gel, resulting in low hardness (Tobin et al., 2013). On the other hand, the increased hardness due to DATEM could be due to its ionic nature as DATEM consists of hydroxyl groups that react with water, resulting in the improved WHC, which was also evidenced in the form of increased WHC (Fig. 2A). Furthermore, DATEM can establish hydrogen bridges with amides bonds of proteins (Gómez et al., 2013). It has been found that DATEM can form hydrogen bonds with starch and glutamine, which is capable to promote gluten protein aggregation in dough by means of binding the hydrophobic surface (Kohajdová et al., 2009). The improved hardness also indicates the viscous behavior, which was also dominant in the results of rheological properties (Fig. 1) in which DATEM showed viscous behavior compared to all other samples. Referring to that, less expansion was also another evidence of high hardness as higher expansion causes more intercellular spaces, which lower the hardness. On the other hand, the improved hardness of SPP samples could also probably due to the ionic strength of SPP.

Conversely, the decreased hardness in SDS even lower than in control could be due to the low water-binding association of SDS with proteins. Although, less water is used in cookie making, its role is very crucial as it behaves as a plasticizer during thermal processing and promotes the viscosity of matrix (Nawaz et al., 2019; Nawaz et al., 2019). Thus, it can be said that the addition of DATEM increased the viscosity of the matrix, which resulted in less expansion and increased hardness as reported previously (Ding and Yang, 2013).

3.4. Effect of emulsifiers on pasting properties of cookies

The gelatinization properties, especially gelatinization behaviors of starch, are critical in the thermal processing of starch-protein-based matrix. These properties predict the processing and storage of end products. Generally, starch granules are present in form of microcrystals, connected by means of hydrogen bonds and insolvable in water at room temperature. Heat treatment disrupts the native structure of starch, leading to the gelatinization of starch, which can be better explained by studying the pasting properties of the end product.

The findings of pasting properties, displayed in Table 1 and Fig. 3, revealed that emulsifiers improved the pasting properties of cookies as compared to the control. The Fig. 3 shows a typically gelatinization curve indicating that at initial temperature, the viscosity of all samples did not change. With the increase in temperature, the viscosity increased gradually and attained the peak viscosity. Among these, peak viscosity, an indication of the strength of binding ability and viscosity, was perceived as significantly ($P < 0.05$) higher in DATEM. The increased peak viscosity in DATEM samples might be because of the ionic nature of DATEM and higher HLB values. Another possibility for the augmented peak viscosity could be attributed to the ester groups present in DATEM that promoted the leaching of amylose from the matrix, which resulted in the increased viscosity ultimately enhancing the swelling power of granules (Cozzolino et al., 2016). Moreover, there is the possibility that ester groups of DATEM promoted the formation of a complex between emulsifiers and amylose molecules as reported by a previous study (Kohajdová et al., 2009). While in the control sample, it is probable that protein molecules enclosed the starch grains resulting in insufficient gelatinization of starch granules and ultimately decreased pasting properties.

Breakdown viscosities represent the strength of starch molecules whereas final viscosity and setback viscosities indicate the extent of reorganization that includes the amylose and/or the alteration of viscous liquid to gel (López-Tenorio et al., 2015). Breakdown viscosity, an indication of the stability of the product during thermal processing, was considerably ($P < 0.05$) lower in control samples compared to all treatments that were made with the addition of emulsifiers. The sample having DATEM significantly ($P < 0.05$) enhanced the breakdown values, which suggests that DATEM increased interaction between protein peptide chains of meat protein and starch granules, which resulted in the increased binding strength of the dough. Our findings are in line with a previous study that also narrated the effectiveness of DATEM in terms of dough strength (Ding and Yang, 2013).

Final viscosity indicates the quality of the end product in which amylose contents play an important role and develop a gel structure after cooking. It also proposes the ability of ingredients to make a gel after heating. The values of final viscosity were significantly higher in DATEM whereas low values were observed for the control indicating that the addition of emulsifiers in meat-based cookies improved the gel strength after heating. Similar results for the improved final viscosity were also observed in a previous study in which DATEM increased the final viscosity of meat-based snacks (Nawaz et al., 2019). Pasting temperature indicates the cooking of food, which is important for the food industry in terms of cost and energy. The minimum pasting temperature was achieved in DATEM-containing samples while it was maximum in control, which suggests that the control sample requires high energy to completely cook the food. The increased pasting temperature in the control sample suggests the delayed swelling of starch granules. The increased pasting temperature in the control sample might be due to the hindering of water availability for granules, which delayed the swelling of starch (Magazù et al., 2008). Thus, it was revealed that DATEM improved the pasting properties com-

Table 1
Effect of emulsifiers on pasting properties of cookies.

Test	Peak Viscosity (RVU)	Trough (RVU)	Breakdown (RVU)	Final Viscosity (RVU)	Setback (RVU)	Pasting Temp (°C)
Control	322 ± 15.84^{a}	268 ± 13.91^{a}	54 ± 17.23^{a}	637 ± 45.79^{a}	369 ± 17.65^{a}	93.78 ± 0.13^{c}
SPP	302 ± 16.76^{a}	288 ± 19.89^{b}	14 ± 11.71^{a}	700 ± 33.86^{b}	412 ± 12.97^{a}	93.34 ± 0.18^{b}
DATEM	443 ± 35.81^{c}	394 ± 10.45^{c}	49 ± 25.99^{b}	929 ± 27.77^{c}	535 ± 18.81^{b}	92.50 ± 0.09^{a}
SDS	424 ± 31.66^{b}	345 ± 11.95^{c}	79 ± 14.66^{b}	910 ± 11.60^{c}	565 ± 15.23^{b}	92.61 ± 0.16^{a}

SPP means sodium pyrophosphate, DATEM means diacetyl tartaric acid ester of mono- and di glycerides and SDS means sodium dodecyl sulphate.
Small letters (a,b,c) represent the significant difference between treatments within rows using one-way ANOVA and Duncan Multiple range test at a significant level of $P < 0.05$.

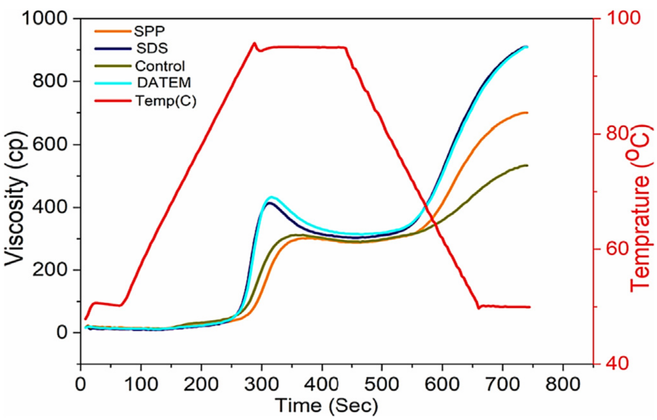

Fig. 3. Pasting properties of cookies prepared with different emulsifiers.

pared to all other studied emulsifiers and control samples. Therefore, it can be used as a dough-strengthening agent in partially meat-based sugar snap cookies.

3.5. LF-NMR analysis of cookies

Water mobility is crucial for meat-based products regarding quality, physio-chemical status, texture, and taste. These properties can be measured by measuring the water movement through the measurement of relaxation time (T_2) (Magazù et al., 2008). The relaxation time (T_2) is further divided into three sections: T_{21} (0.1–1 ms) designates the tightly bound water inside the macromolecules, T_{22} (1–100 ms) resembles the immobilized water and T_{23} (100–1000 ms) implies the presence of unbound water (Smarzyński et al., 2021). It has been established that lower amplitude within constant relaxation time represents a strong association whereas long amplitude shows the frail interaction between solids and water (Nawaz et al., 2021).

The findings of LF-NMR relaxometry are depicted in Fig. 4, which show the presence of three relaxation time (T_{21}, T_{22} and T_{23}). The findings show that the peak for T_{21} relaxation time was higher for SDS and control samples indicating that less water was bound in a matrix whereas relaxation time and amplitude for DATEM and SPP was less, which signifies the close association between solids and liquids. The increased amount of tightly bound water in DATEM and SPP is the hydrophilic and ionic nature of these two emulsifiers, which enhanced the WHC of the matrix as it was evidenced in the results of WHC (Fig. 2A). This suggests that water inside the matrix did not leach out of the matrix during thermal processing. The increased amount of tightly bound water in the presence of SPP has been reported previously in meat-based products (Nawaz et al., 2023; Shao et al., 2016). The less amount of tightly bound water in control could be due to the weak interaction between starch and protein molecules, which results in the seepage of free water from macromolecules.

Regarding T_{22} relaxation time, it was observed that SPP and DATEM had less amplitude and relaxation time, which suggests the increased amount of immobilized water. This further inferred that water inside the matrix was not seeped out and a strong association between starch and protein molecules was observed. Conversely, in the control sample, when the dried fish powder was hydrated during dough making and then in the thermal process,

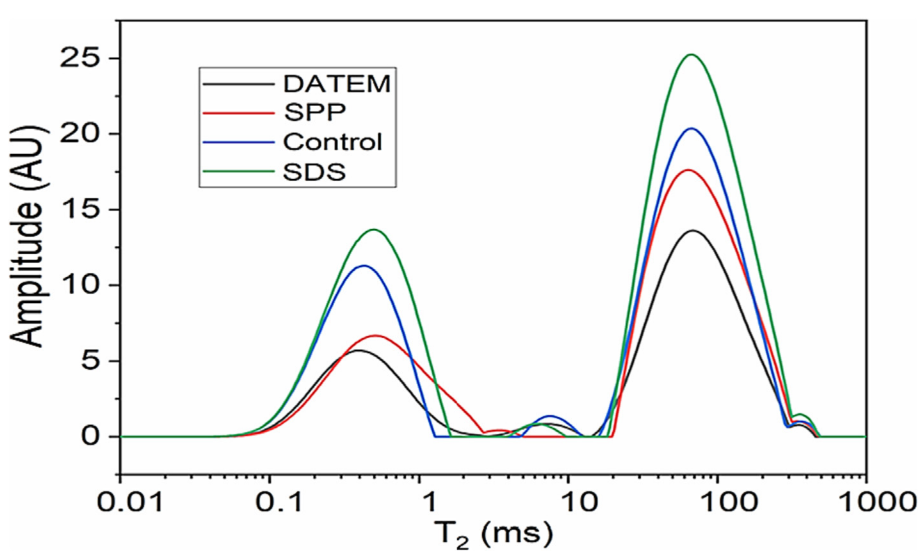

Fig. 4. Water distribution properties of cookies prepared with different emulsifiers.

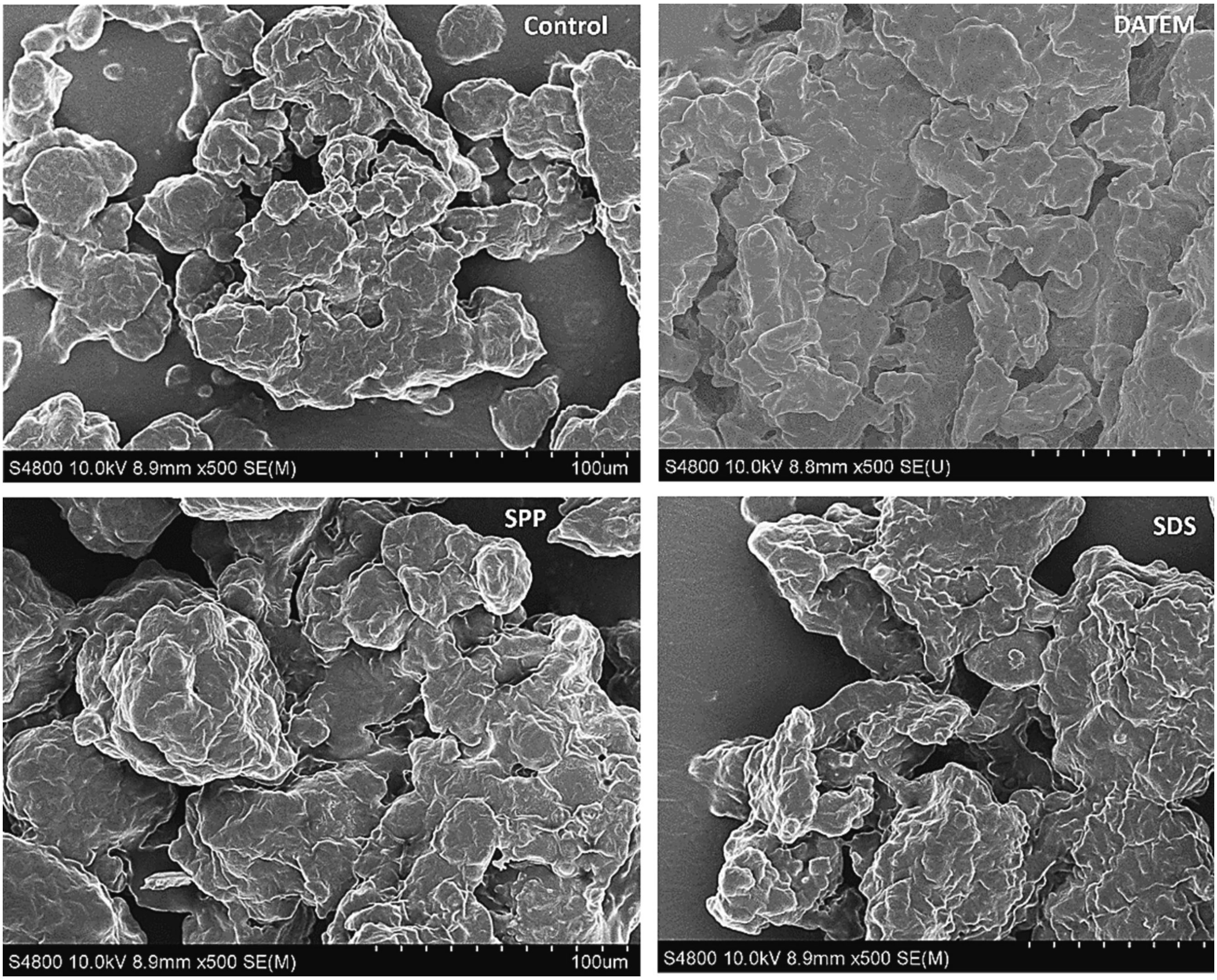

Fig. 5. Microstructure of meat-based cookies prepared with various emulsifiers.

the amount of free water was increased. However, with the addition of emulsifiers, this free water was bounded either absorbed by the emulsifier or by increasing the interaction between starch and protein molecules (Nawaz et al., 2023). T_{23} relaxation time, an indication of unbound water, which was higher in SDS and control samples whereas the addition of DATEM and SPP decreased the unbound water, which was evidenced in terms of less relaxation time for the corresponding peak (T_{23}). The less relaxation time in DATEM and SPP samples indicates the less availability of unbound water for these samples. The role of water addition (although a minute amount) in cookies is of high importance, it acts as a plasticizer and promotes the association between starch and protein. It is highly probable that once hydration was done in the control sample and then during heating, water absorbed by myofibrillar proteins was seeped out, which resulted in a high amount of unbound water. In contrast to DATEM and SPP samples, this water might be bounded caused by the intervention of emulsifiers. Thus, it can be said that the addition of hydrophilic emulsifiers increased the water binding inside the matrix. Similar findings were also reported in the previous study when phosphates were added to meat batters (Campbell et al., 2001). On the other hand, few studies are available on the investigation of water distribution of cookies more specifically prepared with partial substitution of meat and the role of emulsifiers. Our study is the first to report the addition of emulsifiers in meat-based sugar snap cookies.

3.6. Microstructure of cookies

Fig. 5 shows the micrographs of cookies made with various emulsifiers. Distinguished morphology was witnessed in all samples. As seen in Fig. 5, the control sample had wide holes that suggest the intercellular spaces, which was also evinced in terms of the high expansion of cookies (Fig. 2A). These intercellular spaces also caused the less hardness of cookies, which was also seen in terms of low hardness (Fig. 2B) in the control sample. Similar morphology was observed for SDS samples that also showed irregular structures with large holes. The increased intracellular spaces in those samples indicate the weak association between starch protein and water molecules. On the other hand, a uniform and dense matrix was observed in DATEM samples, which suggests a strong starch-protein interaction. The morphology of SPP samples was in between the control and DATEM, however, a relatively dense matrix was observed as compared to the control. DATEM is water soluble and ability to interact with proteins and starch, which makes the microstructure denser and thicker which was also evidenced in terms of increased pasting properties. Moreover, DATEM has the capability to dissolve the myofibrillar proteins, which may be accredited to the strong association between starch and proteins. The SEM images also signify the consequences of WHC and expansion: as the excessive expansion was noticed in SDS and control samples, the wider holes were perceived in those samples indicating the cutoff of the starch-protein network and low WHC, which is an indication of loose structure.

4. Conclusions

The addition of meat in cookies is an inspiring step in order to introduce new ingredients especially the substitution of gluten protein. However, a high amount of meat, and more specifically fresh meat is difficult to maintain the proper texture and structure of cookies. Thus, this study attempted to introduce the addition of emulsifiers in meat-based sugar snap cookies along with freeze-dried fish meat. The key results revealed that the addition of DATEM improved the physio-chemical properties, such as expansion, WHC, and hardness compared to the control. Furthermore, the rheological properties of dough were also improved with the addition of DATEM and SPP, which were further proved in pasting properties. The water distribution properties especially the amount of tightly bound water increased, which is a positive outcome and endorses the addition of ionic emulsifiers. The micrographs further proved the results of physiochemical properties. Thus, it can be said that the partial addition of meat in cookies along with emulsifiers has the potential to develop new cookies with improved physiochemical properties. However, there are some limitations of this study, e.g., the effect of emulsifiers on sensory evaluation, shelf-life, and storage stability of cookies should be investigated. The addition of meat to cookies may result in the development of hazardous compounds, such as protein oxidation, heterocyclic amines, and advanced glycation products. There is a need to conduct future studies to confirm the effectiveness of emulsifiers against the inhibition of hazardous compounds. With all these investigations, the addition of emulsifiers in meat-based cookies can be a meaningful step for the development of new products along with improved physiochemical and safety characteristics.

Declaration of Competing Interest

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

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