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Physical Properties of Modified Table Margarine from Buffalo Fat Fractions with Oil Oloegels

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ABSTRACT

This study aimed to produce a modified table margarine (TM) using oleogels (OGS) of, olive sesame and flaxseed oils with beeswax at 6:3:1:1.78 ratio. Substitute the OGs with buffalo milk fat fractions (Mffr) at two levels 60:40 and 50:50 ratio to produce TM and they were compared with breakfast butter (BM). The resulting butters were characterized on physical, chemical, and nutritional properties. Slip melting oils (SMP), solid fat content (SFC), and thermal analysis were investigated. The SFC in all samples was lower either using 40:60 or 50:50 (w/w). There was a significant increase ($p<0.05$) in PV of all samples during storage at 5◦C. Thermal analysis of incorporated OGs with Mffs may be affected by the concentration of OGs and type of Mffr. It can be concluded that OGs can be used successfully to produce of healthy table margarine

Keywords: Healthy Table margarine, oleogels, Beeswax.

INTRODUCTION

Recently a new a approach of beeswax (BW) oleogel has been successfully produce with different types of oils and applied for production of breakfast margarine (BM) (Öğütcü et al. 2015). Oleogels have some advantages such as not changing the fatty acid composition and trans and saturated fatty acids (SFA) are decreased (Toro-Vazquez et al. 2007; Marangoni, and Garti, 2018; Marangoni, 2011). Beeswax consists of long chain monohydric alcohol compounds and a mixture of esters of fatty acids and fatty alcohols. It's able to act an edible gelator with different oils (Öğütcü et al. 2015; Marangoni, 2011; Marangoni et al., 2012).

Thus, its used in many foods as a new structuring called as organ gelation to prepare the healthier products for lowering the saturated and trans-fat content in food (Marangoni et al., 2012; Yılmaz, and Öğütcü, 2014; Marangoni, 2011).

Organogel define as production of thermo-reversible ,three dimensional gel network and self-standing from oil which has gel called an oleogel instead of organogel without an changes in fatty acid composition of the oil such as beeswax and monoglyceride in hazelnut oil (Toro-Vazquez, et al. 2007; Martins et al. 2016; Marangoni 2011; Marangoni, et al. 2012).

During last few decades, the dietary intake of trans-and saturated fat has been with an increase in serum of low- density lipoprotein (LDL) cholesterol also called "bad cholesterol". At the same time, unlike the effect caused by the consumption of saturated fat, trans –fat has been associated with a decrease of blood levels of high –density lipoprotein (HDL), known as : good cholesterol". Both effects are responsible for increasing the risk of coronary heart disease (Rogers 2009).

In previous works various have been focused on formation of organogold, including beeswax and sunflower was in olive oil (Marangoni, et al. 2012) carnauba wax in canola oil or beeswax in grapseed oil (Mozaffarian et al. 2007).

Thus, in our study, the blending of different oils at ratio (olive oil: sesame oil: flaxseed oil –BW at ratio 6:3:1:1:1.76) replacement of the three mixture of buffalo milk fat fractions (Mffs) from pervious study (Bora, et al. 2017) at 60:40 and 50:50 (w/w) to improvement in nutritional profile of modified butter.

MATERIALS AND METHODS

Materials

1. Fresh Butter and vegetable oil

Raw buffalo butter (~82 % fat) was obtained from Faculty of Agriculture, Cairo University then stored at -18OCuntil used for 3 month

Olive, sesame and flaxseed oils got from the local oil extraction place at Fath oil press machine Company Cairo.

Beewax obtained from El Gomhouria Co. for trading in Drugs, Chemicals and Medical requirements El Sawash Cairo

2. Butter Milk Powder

Butter milk powder, was purchased from Bob's Red Mill Natural Foods, Milwaouke, Oregon 97222, USA. The analytical data was taken from printed packaging material of buttermilk material of buttermilk powder were (Total fat 1g , saturated fat 0.5g ,Trans fat 0g Cholesterol 10 mg , sodium 85 mg ,Total carbohydrate 7g,Protein 5 g)

3. Skim Milk Powder

Low heat spray dried skim milk powder was obtained from Dairy America, California-USA. The analytical data of skim milk powder were (Protein –SNFBasis 34%, Moisture 4%, Fat 1.25% Titratable acidity 0.15%and Solubility Index 1.2ml)

4. Emulsion mixture

Soy lecithin Adlec for Food Use (E322) was purchased from ADM, Hamburg – Germany. Mono-diglycerides, Grindsted®, for Food Use was purchased from DuPont, NHIB, ApS Langebrogade 1-Denmark.

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5. Preparation of butter oil:

Preparing butter oil (BO) by the method of Amer *et al.*, (1985). BO was prepared from fresh butter by melting the butter at 60^{O}C for 2h without agitation, then removing the top oil layer by filtering the oil by (Whatman No.1 paper) and drying over anhydrous sodium sulfate under vacuum in a Buchner funnel to remove ingredient of aqueous phase material. The collected oil was then stored at -18^{O}C until dry fractionation .BO crystallization was carried out by stepwise and successive cooling procedure described by van Aken *et al.*, (1999) to obtained liquid fraction (L15, L25and L35) and solid fraction (S15,S25and S35).

6. Preparation of mix vegetable oil

Mix vegetable oil containing 60% olive oil, 30% sesame oil and 10% Flaxseed oil were heated at 60 C until melted, then taken 85part from vegetable oil

7. Preparation of oil oleogels

Blended vegetable 85 part were melted with 15 part of beeswax at 70◦C

8. Preparation of modified healthy butter (table margarine)

The formulation and the production procedure of OGs and margarine are showed in Figure 1. The water content in watery formulations was 16%, Different milk fat fractions, three vegetable oil and bee wax used to prepare samples.

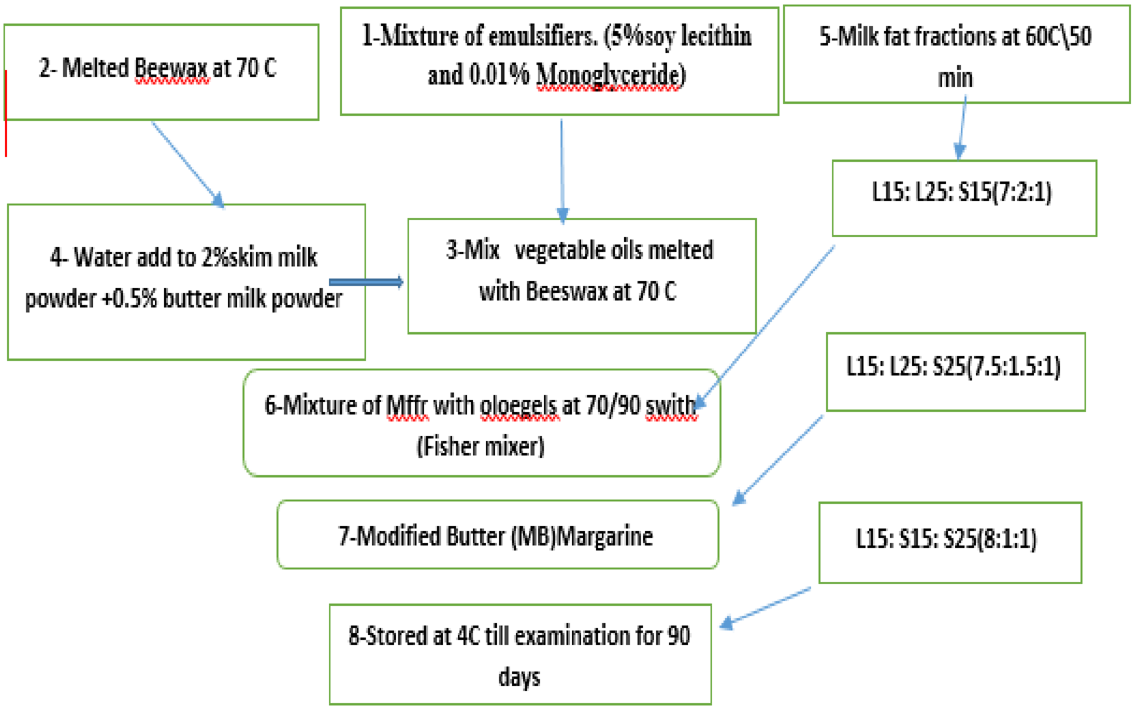

Fig. 1. Technological flow chart for modified butter production

Methods

1. Moisture and fat content

Determination of moisture, solids not fat and fat by difference in butter using routine methods according Evrs et al (2003)

2. Determination of Iodine value

Iodine Value was obtained from fatty acid profile by using reacting ratios (calculation factors) between I (iodine) and either the fatty acids bound to a triglyceride or the free fatty acids according to the methods described by Ham *et al.*, (1998) using following equation

Calculated IV = (% Tetradecenoic acid * 1.1212) + (% Hexadecenoicacid * 0.9976) + (% Octadecenoic acid * 0.8985) + (%Octadecadienoic acid * 1.8099) + (% Eicosenoic acid * 0.8173) + (% Docosenoic acid * 0.7496) + (% Tetracosenoic acid * 0.6923).

3. Determination of Cholesterol

HPLC system was used to determine cholesterol according to (Daneshfar et al.2009)

4. Determination of SMP

The SMP table margarine was determined according to AOCS (2006)

1.Oxidation Stability index (OSI)

An oxidative stability of butter oil and its fractions was determined by using rancimat equipment model 689 (metrohm Ltd, CH-9100 Herisau,Switzeland) as described by Mendez, *et al.*, (1996).

6. The radical scavenging activity (RSA)

The free radical scavenging activity (Antioxidant activity) of the samples was examined by 1,1-diphenyl-2-picrylhydrazyl (DPPH) using the method of Espin, et. al (2000). Characterization of the total free radical scavenger capacity of vegetable oils and oil fractions using 2, 2diphenyl1picrylhydrazyl radical was determined by the following equation:

$$\textbf{Inhibition} = [(Ac - As) / Ac] \times 100$$

Where the Ac is the absorbance of the control (DPPH solution without sample), the As is the absorbance of the test sample (DPPH solution plus test sample

7. Statistical analysis

The level of statistical significance taken as $P < 0.05$, using SAS (version 9.4 TS Level 1M3, SAS Institute Inc., Cary, NC, USA). Least squares mean was calculated and presented throughout (Snedecore and Cochran 1994)

RESULT AND DISSECTION

1. Chemical composition and physical properties of modified butter (table margarine)

The physical and chemical composition of BM (Breakfast Margarine), BR (Butter Recombined) and Table Margarine (TM) are shown in Table (1). There were no significant differences in fat, moisture and solid not fat (SNF) of MB which produce Mffr: oil oloegel blend either 60:40 or 50:50 (w/w). The lowest fat content was recorded. BM (control) to a diverse a higher moisture content which was 40%. Thus. The BM had much lower energy value than those of TM oloegels.

With respect to Iodine Value (IV) was the higher of BM than those all MB oloegel. Whereas its value was lowest in RB being 32.5 while, reached 60.55 to 68.97 for MB oloegels.

The iodine value (IV)is important parameter for measuring degree of unsaturation fatty acids (UNSFA), softness in butter products and tits nutritional quality (Bobe et al. 2003 and Stortz et al. 2012).

Concerning. to Slip Melting Point (SMP) of all MB oloegel either 60:40 blend or 50:50 blend (w/w) had higher than those RB and BM (control) which it values ranged from 44.26 to 46.03 C. These results were attributed to formation of stronger crystal lattic with oil-BW oloegel and three dimensional gel network from oil (Marangoni 2011; Marangoni et al. 2012).

Although, the Table Margarine (TM) contained more USFA, their melting point was higher than RB. These results can be an important view attribute in the application of oloegels in many foods.

As for, cholesterol content ranged from 121.45 to 103.97 (mg/100g) for 60:40 blend and from 103.05 to 109.82 (mg /100g) for 50:50 blend while BM control 105.35 (mg/ 100g) was observed.

From the same Table, it can be clear that the oxidative stability index (OSI) was significantly the highest for RB while the lowest for BM (control). It values were increased with increasing incorporation level of OSF-BW oloegel. It ranged from 15.64 to 18.41 and from17.38 to 24.31 with blend 60:40 and 50:50 respectively, this might due to a high content of phenolic compounds that is related with higher amount of olive oil in the formula oleogel (Nadeem et al. 2013).

Table 1. Chemical compositions and physical properties of modified healthy butter (Table Margarine)

PARAMTER	BM Breakfast Butter Control	RB Recommend Butter Control	MFFS:OSF-OLEOGEL BLENDS 60:40			50:50			*SE	**LSD
			OG1	OG2	OG3	OG4	OG5	OG6		
FAT CONTENT%	57.26^{C}	81.38^{B}	82.30^{A}	82.06^{A}	82.16^{A}	82.10^{A}	82.20^{A}	82.03^{A}	0.1	0.29
MIOSTURE %	40.79^{A}	16.24^{B}	16.13^{B}	16.43^{B}	16.20^{B}	16.37^{B}	16.26^{B}	16.23^{B}	0.11	0.34
SNF%	1.94^{B}	2.37^{A}	1.56^{BC}	1.50^{C}	1.64^{BC}	1.53^{C}	1.53^{C}	1.74^{BC}	0.14	0.41
IV	69.78^{A}	32.51^{G}	60.55^{F}	65.98^{C}	61.77^{E}	64.04^{D}	68.97^{B}	66.49^{C}	0.17	0.51
CHOLESTROL MG100MG	1105.36^{E}	270.69^{A}	123.97^{C}	126.19^{B}	121.45^{D}	106.23^{E}	109.82^{G}	103.05^{F}	0.73	2.20
SMP ◦C	32.23^{F}	35.40^{E}	44.76^{C}	45.30^{B}	44.83^{C}	44.00^{D}	46.03^{A}	44.26^{D}	0.14	0.41
OSI	13.88^{G}	28.28^{A}	18.41^{D}	15.64^{F}	18.25^{D}	21.35^{C}	17.38^{E}	24.31^{B}	0.147	0.44

Significantly a level of 5% of probability (p< 0.5)Means with the same letter are not significantly different
Standard Error=(SE+).Least Significant Difference (LSD).Mffs= Milk fat fractions RB= butter recommend made fromwhole butter oil.
OG1 and OG4 =l15:S15 :S25(7:2: 1) OG2andOG5=L15:L25:S15(7.5:1.5:1) OG3and OG6=l15:l25:s25(8:1:1)
Olive oil:Sesame oil:Flaxseed oil :Beeswax =OSF:BW Oleogels (6:3:1:1.76)ratio

Solid fat content (SFC)

SFC values show the changes in consistency and plasticity of food products at different temperatures. It is an essential measurement in bakery, confectionery and margarine industries. As shown table (2) and Fig (2) that the SFC was found to be the highest for RB, whereas the lowest content was observed in OG6 at 0 C which contained 13.64 g/100g. At 20 C. SFC was recorded between 6.21 to 5.77 (g/100g) when made TM oloegel at 60:40 blend and between 5.04 to 5.27 at 50:50 blend, whereas, MB control was 7.49 (g/100g) at this temperature. On the other hand , the lower SFC at 30 °C with OG sample 50:50 blend than that OG sample 60:50 blend .BM control had 1.01 g/100g SFC at 30 °C that its quality measures with rapid mutability in the mouth and firmness at room temperature (Chougui et al. 2015) .The SFC and SMP values are usually used for the estimation of products suitability for different assess. On the contrary, the fat was the lowest level of UFA had the highest SFC.which according with (Moziar et al.,1989) that texture and consistency are not correlated with the SFC, but instead with other factors such as the processing conditions and crystal lattice of the product.

Radical scavenging activity (RSA%) of fat blend used in margarine preparing

The antioxidant potential of modified butter was estimated using the radical scavenging index (RSA%). As shown in Fig. 3, the DPPH values increased significantly with increasing OGs in MB. The ability to inhibit oxidation was the highest value with 50:50 ratio.

Table 2. Solid fat content (g/100 g) of TM by partial replacement of milk fat fractions with oleogels.

TIME (MIN)	BM	RB	MFFR : OOSFOW RATIO 60:40			50:50			**LSD
			OG1	OG2	OG3	OG4	OG5	OG6	
0	40.61^{B}	57.80^{A}	19.01^{F}	15.40^{G}	13.64^{H}	23.18^{D}	24.44^{C}	21.47^{E}	0.515
10	18.77^{B}	48.13^{A}	12.38^{DE}	11.86^{DE}	9.76^{E}	12.02^{DE}	14.23^{DC}	16.34^{BC}	3.504
20	7.49^{B}	20.79^{A}	5.27^{E}	5.04^{E}	5.12^{E}	5.77^{D}	6.53^{C}	6.21^{C}	0.411
30	1.01^{F}	7.13^{A}	3.87^{D}	3.09^{E}	3.42^{E}	4.38^{C}	4.99^{B}	4.94^{B}	0.335
40	0.14^{E}	0.17^{E}	2.51^{C}	2.21^{D}	2.06^{D}	2.96^{B}	3.34^{A}	3.18^{BA}	0.230

Significantly at a level of 5% of probability (p < .05), Means with the same letter are not significantly different.
****Least Significant Difference (LSD)**
Control=Recombined butter made with whole butter oil, MFr in T1 and T4 =L15: S15:S25 (7: 2: 1) ratio, MFr in T2 and T5 =L15: L25:S25 (7.5: 1.5:1) ratio, MFr in T3 and T65 =L15: S15:S25 (8: 1: 1) ratio, Olive oil: Sesame oil : Flaxseed oil: Bee wax = O:S:F : W (6 : 3: 1: 1.76) ratio.

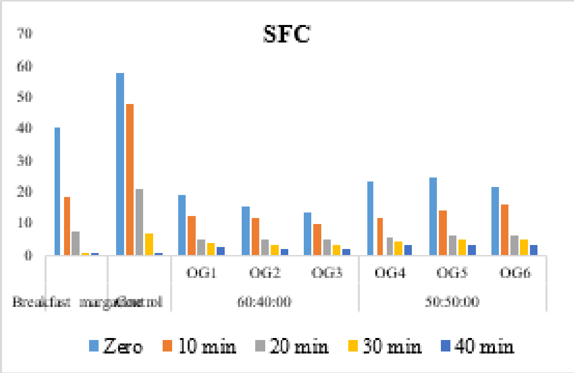

Fig 2. Solid fat content (g/100g) of modified butter by partial replacement of milk fat fractions with oleogels.

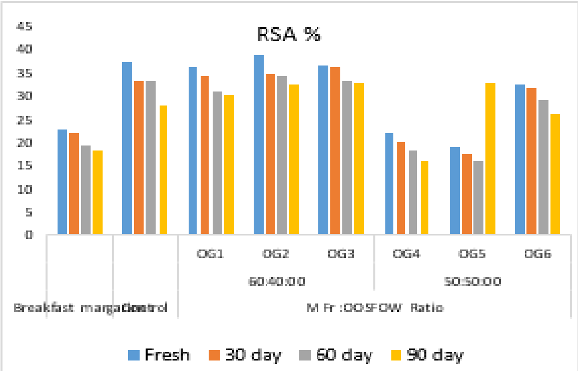

Fig. 3. Radical scavenging activity (RSA%) of TM by partial replacement of milk fractions with oleogels of different kinds vegetable oils- beeswax blends during storage periods at ~5˚C.

These results may be attributed to the sesame oil contains many bioactive compounds such as phytosterols, tocopherols and lignans including sesamin, sesamolin and sesamol which have antioxidation properties (El –Aziz et al. 2013). Also, as well known the flaxseed oil is a rich source of alpha-linolenic acid and some effective phytochemicals such as lignan, phenolic acids, and flavonoids that have many properties including antioxidant and anti-inflammatory (Hadeel et al. 2020).

Differential Scanning Calorimeter (DSC)

DSC provides a more detailed analysis of the melting behavior than slip melting point or solid fat content measurement. DSC analysis to understand the characteristic for these oloegel and it is very important to chemical nature and molecular composition of beewax and how this gelator is able to interact with the oil phase in both melting and crystallization processes. DSC used to study the crystallization and melting behaviors of BM, RB and MB. (table 3 and Fig. 4 ,5).. The crystallization of BM and BR were started at 14.81 and 24.53 C, respectively, while the crystallization for the MB were shifted to be from 28.18 - 15.45 °C. This may be due to the differentiation of physical properties of new fatty acids ratio. Crystallization and melting behaviors of vegetable oils and fats are important for functionality in many prepared food products and used in qualities and quantitative ways for the identification of vegetable oils and fats. The length of the FA increasing the chain leads to an increase in melting point, increasing the degree of unsaturation leads to a decrease in melting point, the TFA have higher melting point than the cis form (Abdolmaleki et al. 2022). The more increase of unsaturated fatty acid in MB with oleogel the more decrease of melting. The endothermic peak (melting point) started at -27.32 and 11.53 °C for both BM and RB respectively, while the endothermal melting peak started around -20.44 and -14.91 °C in the cases of made butter with oloegel. Also, all butter samples mixed with oleogel were decreased compare with RB and this attributed to the different fatty acid structure of modified butter (Table 2).

Table 3. Thermal Parameters of crystallization, melting and their enthalpy of TM made with different milk fat fractions and oloegel vegetable oils- bee wax blends.

CRYSTALLIZATION					
MFFR:OSFBW BLENDS		Temperature			
		T_{OnsetC}	T_{C1}	T_{C2}	Enthalpy ΔH_C (J/g)
BM		14.81^{G}	-4.46^{C}	-22.04^{B}	15.85^{H}
CONTROL		24.53^{B}	16.80^{A}	-1.93^{A}	65.33^{A}
60:40	OG1	21.44^{C}	-3.45^{BC}	….	43.55^{B}
	OG 2	28.18^{A}	-4.32^{C}	-25.20^{D}	27.62^{D}
	OG 3	16.37^{E}	-12.35^{D}	….	38.22^{C}
50:50	OG 4	16.31^{E}	-3.06^{B}	….	26.38^{E}
	OG 5	19.82^{D}	-3.83^{BC}	-24.48^{C}	22.15^{G}
	OG 6	15.45^{F}	-11.77^{D}	….	23.54^{F}
SE		0.115	0.348	0.010	0.018
LSD		0.346	1.044	0.069	0.056

MELTING						
MFFR:OSFBW BLENDS		T_{OnsetM}	T_{M1}	T_{M2}	T_{M3}	Enthalpy ΔH_M (J/g)
BM		-32.58^{H}	-27.32^{E}	-19.90^{D}	1.14^{B}	15.25^{F}
CONTROL		-29.21^{G}	11.53^{A}	17.06^{A}	35.76^{A}	69.26^{A}
60:40	OG1	-26.56^{E}	-21.36^{D}	8.75B^{C}	….	33.52^{B}
	OG2	-24.59^{C}	-20.44^{D}	-0.38^{C}	….	18.13^{E}
	OG3	-27.15^{F}	-17.39^{C}	-2.14^{C}	….	27.86^{C}
50:50	OG4	-25.25^{D}	-20.26^{D}	4.87^{B}	….	23.36^{D}
	OG5	-18.93^{A}	-14.91^{B}	-0.74^{C}	….	9.21^{H}
	OG6	-22.16^{B}	-17.54^{C}	6.43^{B}	….	11.62^{G}
SE		0.026	0.477	1.512	0.019	0.017
LSD		0.079	1.432	4.535	0.077	0.051

Significantly at a level of 5% of probability ($p < .05$), Means with the same letter are not significantly different.
***Standard Error (SE) **Least Significant Difference (LSD) Control=Recombined butter made with whole butter oil, MFFr in T1andT4 =L15: S15:S25 (7: 2: 1) ratio, MFFr in T2andT5 =L15: L25:S25 (7.5: 1.5:1) ratio, MFFr in T3andT6 =L15: S15:S25 (8: 1: 1) ratio, Olive oil: Sesame oil: Flaxseed oil: Bee wax = O: S: F: BW (6: 3: 1: 1.76) ratio.**

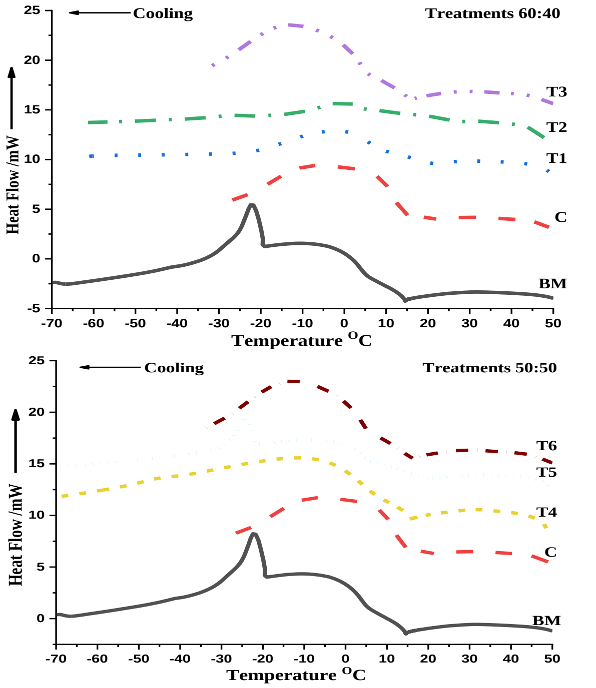

Fig. 4. DSC crystallization thermograms (10°C/min) by cooling from (50:-70°C) of recombined butter by partial replacement of milk fractions with oleogels of different kinds vegetable oils-bee Wax blends

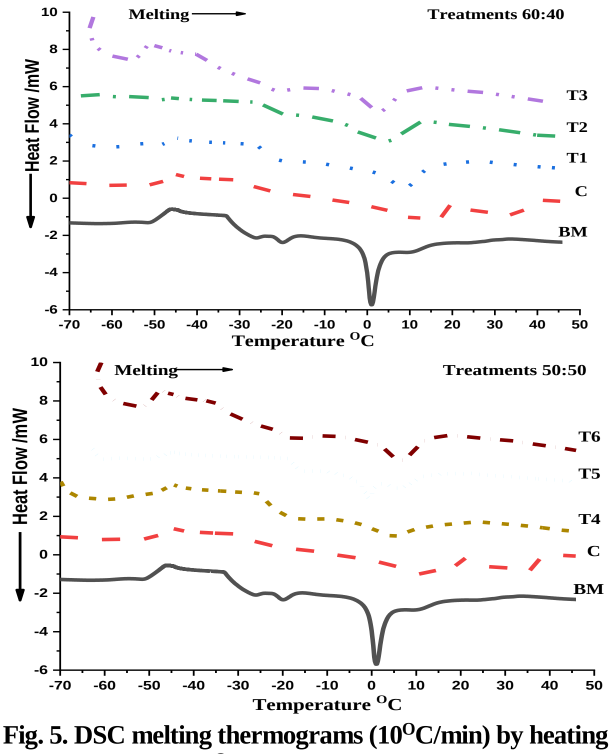

Fig. 5. DSC melting thermograms (10°C/min) by heating from (-70:50°C) of recombined butter by partial (50:50) replacement of milk fractions with oleogels of different kinds vegetable oils-bees wax blends

The decrease of crystallization and melting temperature with increase of gelator level which can be related with the amount of MB leading to the consequently stronger crystal networks as results with firmness and stickiness.

An expected, the OG2 and OG5 sample as compared with other sample which due to contained L25 in formula with higher MUFA content show in table.

As proportion of BW increased in oloegel from 40 to 50 % blending in formula, the enthalpy of crystallization and melting decreased in all treatments.

OG2 sample had the two crystallization points from -4.32 and -25.48 °C (at 40% oloegels), whereas, OG5 had-3.83 and -24.48°C (at 50% oloegels) and melting points from -20.4 C and -0.38 C(40%oloegels) to -14.91 and 0.74 C (50% oloegels).

As for, the crystallization point of OG1 and OG3 decreased (-3.45C) and -12.35 C at 40% oloegel while OG4 and OG6 being -3.06 C and -11.77 C at 50% oloegels. Concerning, the melting temperature for OG1 and OG3 have two peak that record -21.36, -and -17.39 C in peak one and 8.75, -2.14 C in peak two at 40% oloegel.

As the level of TM increased in oloegels, enthalpy temperature of crystallization and melting lower at 50%oloegels than 40% oloegels due to the higher USFA content in samples. Which may be attributed to the properties of beeswax and buffalo milk fat fractions.

This results were confirmed with Öğütcü et al. (2015) who reported that oloegels with BW have lower enthalpy and temperature of crystallization and melting than those carnauba wax. From these results indicated that thermal properties of oloegels may be affected by the concentration of oloegels and Mffr.

Accordingly, the melting of control sample RB (control) had three peak was 11.53,17.06 and 35.76 C in T_{m1}, T_m2 and T_{m3} respectively and their enthalpy was 69.26 J/G might due to the higher content in saturated fatty acids (SFA) and lower content in unsaturated fatty acids (USFA)as compared with BM and among of treatment.

CONCLUSION

Incorporating olive, sesame and flaxseed oils with beewax to obtained OGs using by partial replace to Mffs at 40:60 and 50:50 (W/W) in TM The oxidative stability was improvement that positive effect on nutritional quality for all samples compare with BM. balance of CƆ6:CƆ3 ratio of all OGs samples. The firmness and stickiness significantly decrease of TM. Using OGs with Mffs improvement the spreadability and overall acceptability of TM with 50:50 ratio (w/w). Thus, consumers are aware of the nutritional values of TM and they would be to prefer the OGs samples for a healthier formulation spread.

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انتاج مارجرين المائدة من مشتقات دهن اللبن الجاموسي مع أولوجيل

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الملخص

تهدف هذه الدراسةالي انتاج مارجرين المائده TM باستخدام اوليوجيل OGs الناتج بخلط زيت الزيتون وزيت السمسم وزيت الكتان مع شمع عسل النحل بنسبة 6 :3 :1: 1.76. تم اسبتدال مشتقات دهن اللبن الجاموسي Mffs باوليوجيل بنسبتين 60 : 40و 50: :50 (وزن/وزن) لإنتاج مارجرين المائدة صحي ومقارنته بزبد السوق. تم تقدير درجة الانصهار (SMP) –محتوي الدهون الصلبة(SFC) بالإضافة إلي التحليل الإحصائي لكل النتائج المتحصل عليها . وقد أظهرت النتائج ما رجرين المائدة. حدث أعلا انخفاض في SFC عند كل المستوين 60:40 و50:50(وزن /وزن). الاستبدال الجزئي لمشتقات الدهن الجاموسي خاصه للعينات OG4,OG5بالمقارنه بالعينات الأخرى. وتوصي هذه الدراسة باستخدام الاوليوجيل المحضر من خليط الزيوت النباتية مع شمع عسل النحل لإنتاج مارجرين المائدة

الكلمات الدالة : مارجرين المائدة ، أولوجل، شمع عسل.