

## A Pocket-sized Mid-infrared Emitter Improves Shelf-life, Sensory Qualities and Health Benefits of Dairy Products

T Umakanthan<sup>1\*</sup>, Madhu Mathi<sup>1</sup>, U Umadevi<sup>2</sup>

<sup>1</sup>Department of Dairy, Veterinary Hospital, Tamil Nadu, India; <sup>2</sup>Department of Botany, The Standard Fireworks Rajaratnam College for Women, Tamil Nadu, India

## ABSTRACT

Commercialization of dairy products compelled the use of preservatives, additives and complex processing and packaging technologies to enhance their shelf-life which is vital for dairy industry's survival, logistical and export capabilities; these techniques also impart negative effects on the sensory attributes. Whereas, sensory attributes, shelf-life and minimum processing are the qualities expected by consumers. In this research, we invented an economical pocket-sized emitter called as 2-6 µm Mid-infrared Generating Atomizer (MIRGA). This was sprayed externally over the packaged dairy products. 2-6 µm mid-infrared has resulted in chemical bond and molecular changes in the dairy products and thus enhanced their shelf-life by 30%-1450%, flavor and sensory attributes. Sensory panel tests and laboratory analyses of the dairy products demonstrated the mid-infrared induced chemical bond and molecular level changes that resulted in substitution of harmful moieties by favorable compounds which would prevent and help manage some diseases of dairy origin.

Keywords: Dairy products; 2-6 µm mid-infrared; Shelf life; Sensory attributes; Enhancement; Health benefits

## INTRODUCTION

Dairy products are highly perishable and holistic nutritional food; extension of their shelf-life is mandatory. Many methods such as the use of heat treatment, preservatives, acidifiers and emulsifiers, ultraviolet irradiation and combination of such methods are in use [1]. These methods affect the natural aroma and sensory attributes of dairy products and also possess cumulative poisoning effect, especially to the babies, children and elders [2-5]. Stringent food safety act in future has adversity on the survival of commercial dairy industry unless an affordable technology is evolved.

Infrared irradiation is on increasing trend in the food industry. It is proven to be a successful alternate for the pasteurization of liquid foods such as honey, milk, beer and orange juice [6]. But, to the best of our knowledge, no study has been done using mid-infrared on the enrichment of dairy products. Therefore, to extend the shelf-life and simultaneously preserve the sensory qualities of dairy products, we have chosen mid-Infrared (mid-IR) portion of the electromagnetic spectrum. Mid-IR is the fingerprint region at which most of the earthly molecules have their vibrational frequency [7]. Mid-IR spectrum is biologically safe, can penetrate any intervening media [8], and is absorbed by all organic compounds [9]. We have designed a pocket-sized 2-6  $\mu$ m mid-IR emitter briefly called as Mid-IR Generating Atomizer (MIRGA), and the objective of this study is to investigate the application of MIRGA on packaged dairy products such as liquid milk, butter milk, curd, butter, ghee, milk powder and cheese powder/cheese. MIRGA when sprayed externally over the packaged dairy products has generated 2-6 µm mid-IR which acted on the inside dairy products. We have performed various instrumentations to probe the structural and chemical composition details of these products before and after MIRGA sprayings and reported the corresponding increase in shelf-life and sensory attributes relative to the number of sprayings. We compare to demonstrate that the MIRGA technology is more advantageous than the existing methods and discuss its application on preserving features and quality of dairy products in detail. MIRGA is found to be a potential option for the dairy industry with operational easiness, safety and cost-effectiveness.

## MATERIALS AND METHODS

## Dairy products

Raw milk was procured from a local milk vendor. Chilled and boiled milk were respectively prepared by chilling and boiling the raw milk in home. Pasteurized milk, buttermilk, curd, butter, ghee, milk powder and cheese powder were obtained from a local supermarket. Each dairy product was repackaged into multiple smaller quantities of about 200 mL/200 gm in sterile polythene sachets of >51 micron thickness, and used in this study.

Correspondence to: Umakanthan T, Department of Dairy, Veterinary Hospital, Tamil Nadu, India, E-mail: rkbuma@gmail.com

Received: 18-Jan-2024, Manuscript No. ADR-24-29254; Editor assigned: 22-Jan-2024, PreQC No. ADR-24-29254 (PQ); Reviewed: 05-Feb-2024, QC No. ADR-24-29254; Revised: 12-Feb-2024, Manuscript No. ADR-24-29254 (R); Published: 19-Feb-2024, DOI: 10.35248/2329-888X.23.11.647

Citation: Umakanthan T, Mathi M, Umadevi U (2024) A Pocket-sized Mid-infrared Emitter Improves Shelf-life, Sensory Qualities and Health Benefits of Dairy Products. J Adv Dairy Res. 14:647.

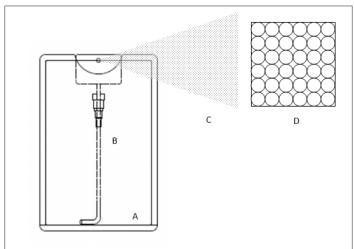
**Copyright:** © 2024 Umakanthan T, et al. This is an open-access article distributed under the terms of the Creative Commons Attribution License, which permits unrestricted use, distribution, and reproduction in any medium, provided the original author and source are credited.

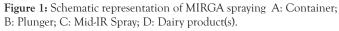
## Mid-IR emitter apparatus-'MIRGA'

MIRGA [10], is a 20 ml pocket sized atomizer (Figure S1) containing inorganic water based solution in which approximately two sextillion cations and three sextillion anions are suspended in water in their fundamental state and can move as free particles. It has a very little background frequency of detectable disintegration which is less than that of cosmic events whereas even humans have more radioactivity (around 10 microns). While spraying the emitter/ atomizer, the water-based ionic solution gets excited/charged, which in turn lead to oscillation among the imbalanced ions [11], resulting in the emission of mid-IR [12,13], and the accelerated ions in the sprayed ionic clouds collide among themselves which also generated mid-IR energy [14]. Depending on pressure (vary with the user) applied to plunger, every spraying generates 2-6 µm mid-IR (Figure S2). Design of the MIRGA and emission of 2-6 µm mid-IR has been presented in detail by [15,16]. Every time spraying emits 0.06 ml which contains approximately seven quintillion cations and eleven quintillion anions.

The inorganic compounds used in the generation of MIR are a perspective for biomedical applications [17,18]. It is also a new synthesis method for preparation of functional material (2-6  $\mu$ m mid-IR) [19-21]. It is well known that the combination of different compounds, which have excellent electronic properties, leads to new composite materials, which have earned great technological interest in recent years [22].

MIRGA spraying was externally done from a distance 0.25 to 0.50 meter over the packaged dairy products (Figure 1). This distance allowed the MIRGA solution to form ion clouds, which oscillated and generated 2-6  $\mu$ m mid-IR; closer spraying did not generate the mid-IR. The 2-6  $\mu$ m mid-IR penetrated the intervening packaging material and acted on the inside dairy products [23].





## Experimental design

13 packets of the raw milk each containing 200 mL was taken. Out of the 13 packets, one was marked "C" (control) and the remaining 12 packets were numbered from 1 to 12. MIRGA spraying was given from a distance of 0.25 to 0.50 meter externally over the milk packets on either sides; number of spraying corresponded to the packet number, i.e., packet #1 received one spraying on one side, packet #2 received 2 sprayings i.e., 1 spraying on either side, packet #3 received 3 sprayings i.e., 1 spraying on a side and 2 sprayings on another side, and so on; control packet received no spraying.

## OPEN OACCESS Freely available online

More number of spraying, i.e., 12 spraying for milk, was expected to approximate natural denaturing of dairy characteristics due to the input of excess mid-IR energy. In a few dairy products, fewer or more than 12 packets and 12 sprayings were tested. Curd and buttermilk required respectively up to 14 and 16 sprayings; whereas butter, ghee and milk powder required 10 sprayings and the cheese powder required 7 sprayings. Therefore, the control and trailed sample size (number of packets and sprayings) varied either more or less with respect to enhanced or reduced sensory qualities and with longer or shorter shelf-life of the dairy product.

## Shelf life and sensory tests

The sprayed and control packets were kept at room temperature (average 38°C) and similar batch of another 13 packets in refrigeration temperature. Periodically, the control and trial packets were opened and subjected to trained expert panel (n=6) and nontrained consumer panel (n=25) tests. The acceptability index used was a hedonic scale with a 9 point nominal structure: 1-Dislike extremely, 2-Dislike very much, 3-Dislike moderately, 4-Dislike slightly, 5-Neither like nor dislike, 6-Like slightly, 7-Like moderately, 8-Like very much, 9-Like extremely [24,25]. The experiment was explained to the trained panelists. They were asked to rate their preference of sensory evaluation for different traits, using the hedonic scale. The consumer panel was requested to describe the results in the fewest words with maximum clarity. The control and samples with enhanced sensory attributes, and with reduced sensory attributes were subjected to further instrumentation analyses. In every trial for every dairy product, we selected and tested one particular brand of the dairy product, and later for accuracy other brands of the dairy product were tested; brands and batches were tested individually, and never mixed.

#### Physicochemical tests

The control and MIRGA sprayed samples were subjected to the following instrumentations and analysed for the physicochemical changes caused by MIRGA spraying. Gas Chromatography-Mass Spectrometry (GC-MS) and Liquid Chromatography-Mass Spectrometry (LC-MS) were used to identify chemical compound transformations; Fourier Transform Infrared Spectroscopy (FTIR) for chemical bond changes; Powder X-ray Diffraction (PXRD) for molecular structural changes; Proton Nuclear Magnetic Resonance (1H-NMR) for nuclear resonances; Transmission Electron Microscopy (TEM) for configuration changes; and 3-Dimensional fluorescence spectroscopy for contour and signal-to-noise ratio. (Supplementary text part I).

Same experimental design, sensory evaluation and physicochemical analyses followed for all other dairy products *viz.*, pasteurized milk, buttermilk, curd, butter, ghee, milk powder and cheese powder.

## RESULTS

## Shelf-life and sensory evaluation

The control samples were found to have regular taste and the enhanced sensory attributes, if any, were observable in less than 2 minutes. As the spraying number increased, the flavor, sensory attributes and shelf-life were gradually found to be enhanced. And after certain number of sprayings, the aforementioned parameters started to decrease in performance. Both these enhanced and decreased sensory attributes were observable in about 1-2 minutes. From (Tables 1 and 2), number of MIRGA sprayings required to enhance the sensory quality is 5 for liquid milk (raw, boiled, chilled and pasteurized milk), 10 for buttermilk, 4 for curd, milk powder

## OPEN OACCESS Freely available online

and ghee, 2 for cheese powder and butter, at both the room and refrigeration temperatures. Whereas, number of sprayings that decreased the sensory qualities ranged from 10 to 16, except for cheese powder in which 7 sprayings decreased its sensory quality. Sensory scoring was based on a 9-point hedonic scale as specified by.

It is also observed that the dairy products having less water content (butter, ghee, milk powder and cheese powder) required less number of MIRGA spraying for enhancement/reduction in their sensory qualities when compared to the products with more water content (liquid milk, butter milk and curd). On contrary, Table 3 showed that, the sprayed dairy products with less water content had acquired an extended shelf-life of 30%-100% at room temperature and 30%-64% under refrigeration; and those with more water content had 200%-450% and 90%-1450% extended shelf-life respectively at room temperature and refrigeration.

Table 1: Sensory scoring by trained expert panel.

Sl.No	Sample		Hedonic score <sup>*</sup> for control samples		Trial samples					
		Quantity of sample –			Spraying	Hedonic score <sup>*</sup>		Spraying	Hedonic score <sup>*</sup>	
		sampie	Taste	Flavor	number	Taste	Aroma	number	Taste	Aroma
1	Raw milk	200 mL	5	5	5	7	8	12	2	1
2	Boiled raw milk	200 mL	5	5	5	7	8	12	1	1
3	Chilled raw milk	200 mL	5	5	5	7	8	12	2	2
4	Pasteurized milk	200 mL	5	5	5	7	8	12	1	2
5	Butter milk	200 mL	5	5	10	5	5	16	3	2
6	Curd	100 gm	5	5	4	7	6	14	1	1
7	Butter	100 gm	5	5	2	6	6	10	2	1
8	Ghee	100 gm	5	5	4	6	7	10	1	2
9	Milk powder	100 gm	5	5	4	7	8	10	1	2
10	Cheese powder	50 gm	5	5	2	7	7	7	1	1

Note: '9: Point hedonic scale details; 1: Dislike extremely; 2: Dislike very much; 3: Dislike moderately; 4: Dislike slightly; 5: Neither like nor dislike; 6: Like slightly; 7: Like moderately; 8: Like very much; 9: Like extremely.

 Table 2: Sensory qualities description by non-trained consumer-panel.

SI.No	Dairy product	Sensory qualities as described by non-trained consumer-panel				
1.	Milk (raw milk, boiled raw milk, chilled raw milk and pasteurized milk)	Control-regular taste and flavor 2 sprayed-sweetness, taste and flavor slightly enhanced than control 5 sprayed-sweetness, taste and flavor significantly enhanced than control 12 sprayed-undesirable taste and flavor				
2.	Buttermilk	Control-regular taste and flavor 4 sprayed-taste and flavor reduced than control 10 sprayed-taste and flavor enhanced, desirable sourness sensed than control 16 sprayed-undesirable taste and flavor				
3.	Curd	Control-regular taste, sourness 4 sprayed-taste (very tastier) enhanced than control 8 sprayed-undesirable taste 14 sprayed-undesirable taste				
4.	Butter	Control-regular taste, flavor and texture 2 sprayed-taste, flavor enhanced; became smooth and creamy 10 sprayed-taste, flavor reduced; became very soft (consistency lost)				
5.	Ghee	Control-regular flavor and taste 4 sprayed-flavor and taste enhanced 10 sprayed-became tasteless				
6.	Milk powder	Control-Natural taste 4 sprayed-sweetness enhanced 10 sprayed-sweetness reduced				
7.	Cheese powder/ cheese	Control-regular taste 2 sprayed sample-saltiness reduced, sourness reduced, flavor enhanced 7 sprayed sample-tasteless, flavor reduced, saltiness reduced				

#### Table 3: Shelf life enhancement of dairy products.

	Name of the sample	At room t	temperature (ave	rage 38°C)	At refrigerated condition			
OL M		No. of days remain unspoiled			No. of days remain unspoiled			
Sl.No.		Control (Non sprayed)	Trial (Sprayed)	_ % of shelf life enhanced	Control (Non sprayed)	Trial (Sprayed)	_ % of shelf life enhanced	
1	Raw milk	<1	02-Mar	300	2	28	1300	
2	Boiled milk	<1	4	300	03-Apr	30	650	
3	Chilled milk*	1	02-Apr	200	2	31	1450	
4	Pasteurized milk	<4	8	200	14- Dec	51	260	
5	Butter milk	2	6	200	15-21	43	100	
6	Curd	2	11	450	14-21	40	90	
7	Butter	7	14	100	14-28	46	64	
8	Ghee	80-95	106-125	30	250-306	332-400	30	
9	Cheese powder	<1	02-Mar	100	50-110	80-180	61	

Note: Chilled milk: Raw milk sold in chilled condition without any heating/pasteurization by the local vendors; Shelf life % calculation: Trial days (minus) maximum control days=X; % of Shelf-life enhanced=X/control days \*100.

#### Physicochemical analyses from instrumentation data

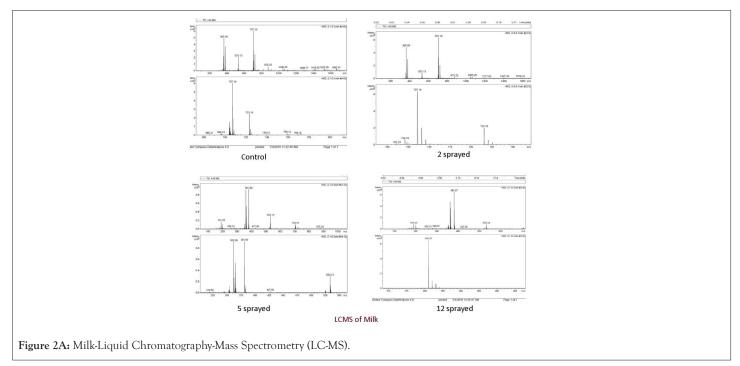
#### Milk instrumentations results

**LCMS-Milk:** Compared to control, lactose (m/z 365.09) concentration in 5 sprayed sample is 5 times lower due to breakdown of lactose into glucose and galactose, hence sweetness increased; but lactose completely disappeared in 12 sprayed sample. The aroma is due to kampferols and dihydroxy kaempferols (m/z 533.13), which are degraded in 12 sprayed, hence aroma reduction. The casein (m/z 707.19) and milk lipids (m/z>875.23) are degraded in 5 and 12 sprayed samples, hence 5 sprayed milk is better than other samples (Figure 2A) (Table S1).

**FTIR-Milk:** The first difference involved the  $CH_2$  absorption band at approximately 2911-2846 cm<sup>-1</sup>, which is related to the acyl chain on fatty acids. As expected, the degree of absorption for this band correlates with the fat quantity in each sample, with a higher fat

content resulting in higher IR absorption. The intensity of this peak is more in 2 and 5 sprayed samples. The peak at 1620 cm<sup>-1</sup> and 1544 cm<sup>-1</sup> is for amide I and II, respectively present in protein or due to -C=O from proteins. The peak at 1159 cm<sup>-1</sup> and 1076 cm<sup>-1</sup> is due to lactose. This peak intensity is more in control and 2 sprayed samples. The peak at 1017 cm<sup>-1</sup> is due to C=O from polysaccharide. This peak is very intense in control and is reduced in 12 sprayed samples. The broad peak at 3400 cm<sup>-1</sup> is due to -N-H from proteins and -O-H. The peak at this region is broad in control and narrow in other samples (Figure 2B).

**Proton NMR–Milk:** In control, lactose (3.5 to 5.5 ppm) peaks are uniform but in the sprayed samples the lactose is degraded which have many smaller peaks and lot of environment changes found in lactose region. In control, hydroxyl butyrate (1.2 ppm) is very low but found gradually increasing with increasing number of sprayings (Figure 2C).



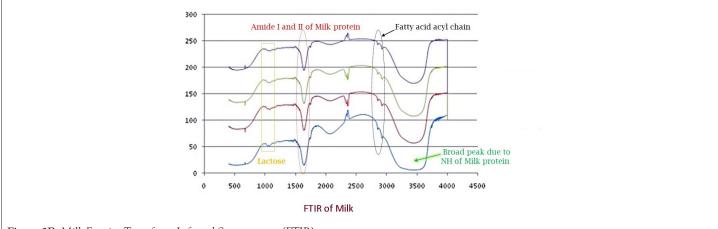
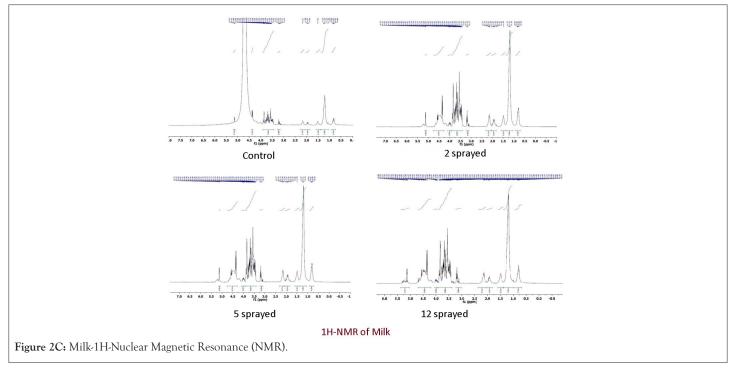


Figure 2B: Milk-Fourier Transform Infrared Spectroscopy (FTIR).



## Buttermilk instrumentation results

**LCMS-Butter milk:** Signals of characteristic Triacylglycerols (TAGs) containing both short and medium/long chain fatty acids were present in mass region m/z 500-800. Diacylglycerol fragment ions, monoacylglycerol fragment ions and acylium ions [RCO]<sup>+</sup> fragment ions can be observed in a lower mass region. Lactose (m/z 365.09) concentration in the control, 4 and 10 sprayed samples is similar, but 7 times lower in 16 sprayed sample which indicates the breakdown of the lactose into glucose and galactose and fragments of glucose and galactose. However, lactic acid in buttermilk makes its lactose content easier to digest, and thus recommended for lactose intolerant patients [26], (Figure 3A) (Table S2).

Buttermilk aroma is due to kampferols and dihydroxykaempferols (m/z 533.13) which is present in all sample but completely degraded in 16 sprayed sample, resulting in abrogation of aroma. The casein (m/z 707.19) is intact in all samples except in 16 sprayed sample. Slight degradation of triglycerides is also observed in 4 and 10 sprayed samples but complete degradation in 16 sprayed sample (Table S2). Overall, 10 sprayed buttermilk is chemically better when compared to the other buttermilk samples.

FTIR-Butter milk: The first difference involved the -CHstr band

at approximately 2966-2887 cm<sup>-1</sup>, which is related to the acyl chain on fatty acids. The intensity of this peak is more in 16 sprayed sample and low in control, 4 and 10 sprayed samples. The peak at 1620 cm<sup>-1</sup> is due to -C=O str from proteins. However, this peak is shifted to 1586 cm<sup>-1</sup> in control. Hence in control -C=O is in conjugation with olifinic or phenyl group which is removed upon spraying in other samples. The peak at 1373 cm<sup>-1</sup> and 1050 cm<sup>-1</sup> is found only in 16 sprayed sample. But not present in control and other samples. This is due to -C-O str of anhydride produced in 16 sprayed sample. The broad peak at 3300-3500 cm<sup>-1</sup> is due to -O-H. This peak is very broad in control and appears at 3360 cm<sup>-1</sup> due to intermolecular hydrogen bonding [O-H...O] and narrow, shifted in 16 sprayed sample due to free -O-H groups (Figure 3B).

**Proton NMR-Butter milk:** Lactose (3.5 to 5.5 ppm) is in the similar molecular environment in all the four buttermilk samples. Hydroxybutyrate (1.2 ppm) concentration is lowest in 4 sprayed sample followed by 16 sprayed but highest in 10 sprayed sample (Figure 3C). Butyrate and hydroxybutyrate generally contributes to the characteristic smell of the buttermilk. Therefore, 10 sprayed sample has acquired health benefits as described in the milk's proton NMR result.

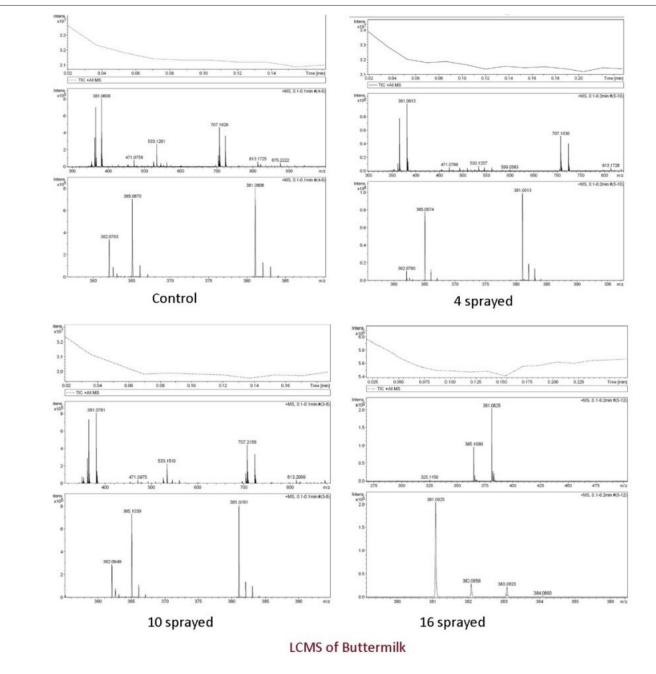
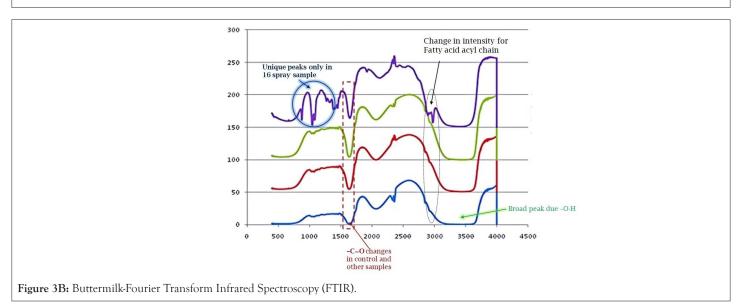
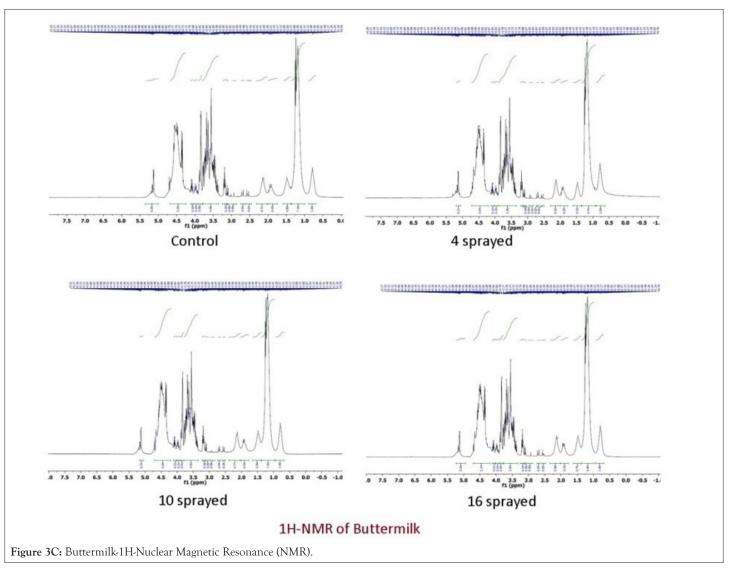


Figure 3A: Buttermilk-Liquid Chromatography-Mass Spectrometry (LC-MS).





#### Curd Instrumentation results

LCMS-Curd: The ESI mass spectrum of control sample showed that the peak at m/z 381.0854, m/z 203.0558,m/z533.1632, m/z 707.2308 corresponding to [M+H]+. The ESI mass spectrum of sample 4 sprayed sample showed peak at m/z 381.0880, m/z 281.0775, m/z 471.1135 m/z 533.1687, m/z 723.2308 corresponding to [M+H]+. The ESI mass spectrum of 8 sprayed sample showed peak at m/z 191.0463, m/z 281.0880, m/z 362.1114, m/z 364.1086, 365.1184, m/z 427.0907, m/z 533.1754, m/z 704.2391, m/z 813.2436 corresponding to [M+H]\*. Similarly the ESI mass spectrum of sample 14 sprayed sample showed major peak at m/z 191.0464, m/z 281.0819, m/z 362.1138, m/z 364.1107, 365.1188, m/z 427.0912, 471.1195, m/z 533.1754, m/z 704.2408, m/z 813.2436 corresponding to [M+H]\*.

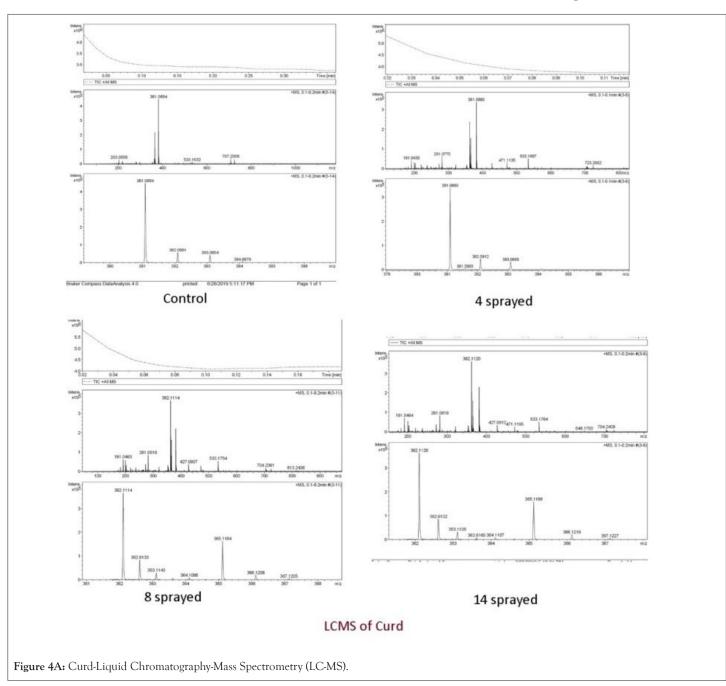
The presence of additional peaks in 8 sprayed sample at m/z 813.2436 indicates the production of longer chain high molecular weight fatty acids. Similarly the additional peaks at, m/z 427.0912, 471.1195, m/z 533.1754, in 14 sprayed sample shows that conjugation occurred and high molecular weight compounds are formed. In control and 4 sprayed samples, the peak at m/z 381.0854 is found. But in 8 and 14 sprayed samples, peak is noticed at m/z 365.1184, hence there is breakage of -OH bond (Figure 4A) (Table S3).

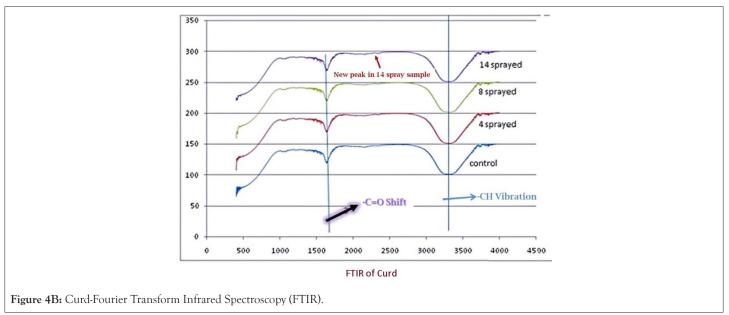
**FTIR-Curd:** The peak at 1600 cm<sup>-1</sup> is due to -c=0 str which is shifted to 1610 cm<sup>-1</sup> in 14 sprayed sample. A new peak arises at 2260 cm<sup>-1</sup> in this sample, which is not found in other samples.

The peak at 1600 cm<sup>-1</sup> is more intense in 8 sprayed sample and weak in 4 sprayed sample. The weak vibration at 1375 cm<sup>-1</sup> relates to the presence of a methylene group and a tertiary carbon atom (-CH(CH3)-). Control and the samples show the weak multiple bands in the area from 1200 to 1000 cm<sup>-1</sup>. It is evident that the 4 sprayed sample shows weaker vibration of carbonyl group in comparison to control, and 14 sprayed sample has a stronger vibration. These can be explained by the fact that there is different structural environment in these samples.

The increased area of the C-H environment indicates lypolysis of the fatty acids present in curd, this would result in the formation of short chain fatty acids which are volatile and contribute to the aroma of the curd. There is no shift of the carbonyl peak in 4 sprayed sample, indicating that the resulted short chain fatty acids are in the similar chemical environment as control causing increased aroma and taste (Figure 4B).

In 8 and 14 sprayed samples there is increased area of the C-H environment which also indicates that intensive lypolysis is occurring. However, there is slight shift of the carbonyl peak compared to the control and 4 sprayed samples indicating a change in environment of carbonyl (Figure 4B). This is due to the formation of acid anhydrides or unwanted fusion of the short chain fatty acids with the other components of the curd. This is the reason of the observed decreased aroma and taste of the 8 and 14 sprayed samples compared to the 4 sprayed and control curd samples.





#### Butter instrumentation results

**GCMS-Butter:** Control sample contains short, medium and long chain fatty acids and mixture of saturated and unsaturated fatty acids. The major peak of palmitic acid was found followed by oleic acid peak (8-Octadecenoic acid). The medium chain fatty acid such as undecanoic acid and myristic acid were also found in control sample. In 2 sprayed sample, there was increase in peak of oleic acid and stearic acid with decrease in palmitic acid, undecanoic acid and myristic acid. There was also no peak of any short chain fatty acid in 2 sprayed sample. Additionally, there was new peak of Phthalic acid and 9-octadecenoic acid, 2 phenyl-1, 3-dixalan-4-yl methyl ester, cis. On other hand, 10 sprayed sample has shown unique peak 12-Octadecenoic acid (Oleic acid) with increase in Octadecanoic acid (stearic acid). The palmitic acid remained nearly constant as compared to control while there was slight decrease in myristic acid (Figure 5A) (Table S4).

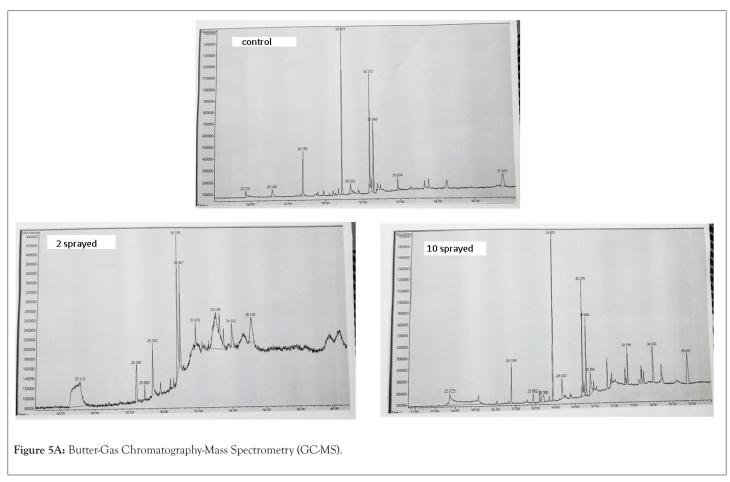
**FTIR-Butter:** Control sample shows a broad band between 3650-3200 cm<sup>-1</sup> which is attributable to O-H stretching of alcohols and carboxylic acids. A more defined, multiple band appears between 2800-3050 cm<sup>-1</sup>, which are typical of the C-H stretching of saturated moieties, like alkanes. A very intense peak is observed around 1745 cm<sup>-1</sup>, attributable to the stretching of C=O bonds. Below 1500 cm<sup>-1</sup> the fingerprint region is located. This region is usually very crowded and it is difficult to identify peaks accurately. 2 sprayed sample shows a spectrum with a significantly lower transmittance

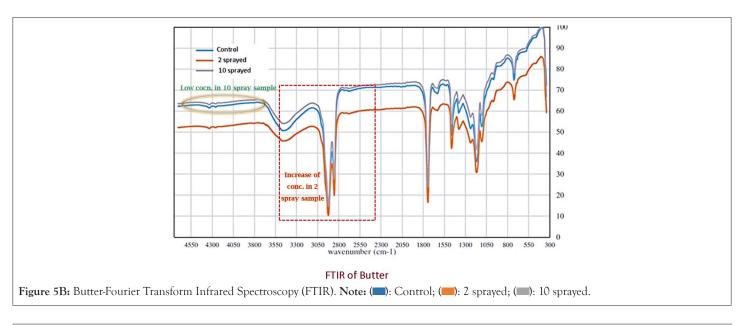
## OPEN OACCESS Freely available online

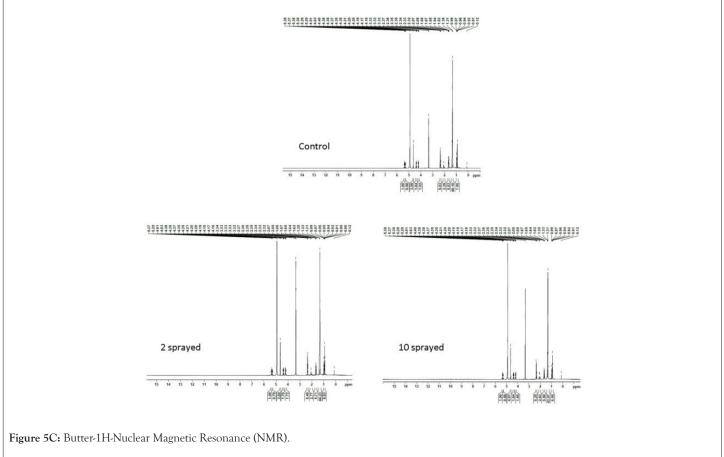
(more absorption), pointing to an increased concentration of the compounds giving rise to the observed bands. 10 sprayed sample shows a spectrum with a lower transmittance compared with the control sample, and remarkably lower than the transmittance of the 2 sprayed sample. This indicates that a reduction in the concentration of the compounds originating the bands and peaks occurs in the 10 sprayed sample compared to the control and, especially, compared to 2 sprayed sample (Table S5).

The number and position of the peaks and bands of the 3 samples are almost the same. But the most remarkable change is the variation in the transmittance (absorption) that is directly related to the concentration of the compounds, essentially, butanedione, 2,3-pentanedione, and 3-hydroxybutanone (Figure 5B). Changes observed in aroma and taste of butter samples is directly related to variations in the concentration of these compounds. Changes in texture are more related to the spatial organization of triacylglycerides and fatty acids, which is related to chemical changes in their structures.

**Proton NMR-Butter:** The number of CH<sub>3</sub> (and CH<sub>3</sub> aromatic) groups is more or less the same in all samples. However, number of CH<sub>2</sub> groups clearly increases upon spraying (the trend is 10 sprayed >2 sprayed >control) (Figure 5C). This suggests changing in the butter structure upon the spraying (Supplementary text part II (a)) (Table S6).







#### Ghee instrumentation results

**FTIR-Ghee:** Sprayed samples show typical absorptions of control sample, but a discrete intensity increase in O-H band (3464-3471 cm<sup>-1</sup>) observed, assigned to the formation of free fatty acids, a discrete displacement of the bands at 1597 (1581 cm<sup>-1</sup>) of the cis -CH=CH-functional group of the oleic acid (C18:1 cis n-9) and Csp3-H at 1103 (1111 cm<sup>-1</sup>) indicating a intermolecular modification in the sprayed samples (Figure 6A) (Table S7A and S7B).

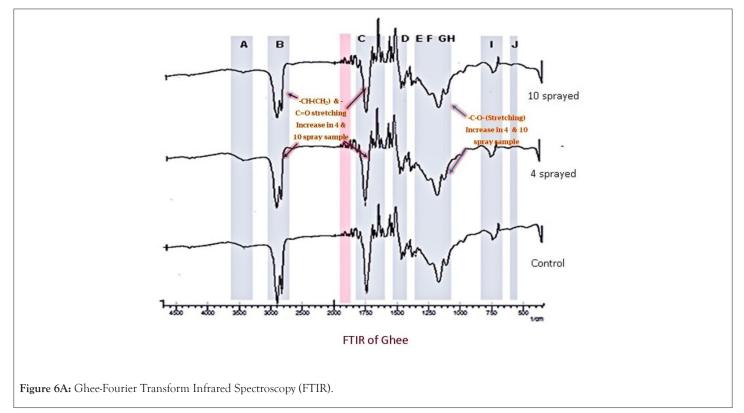
The  $-CH-(CH_2)$  and -CH-bond stretch of long chain fatty acid decreased in 4 sprayed ghee, but increased in 10 sprayed sample suggesting the fact that this has involvement for aroma enhancement. The 4 and 10 sprayings always have shown increase

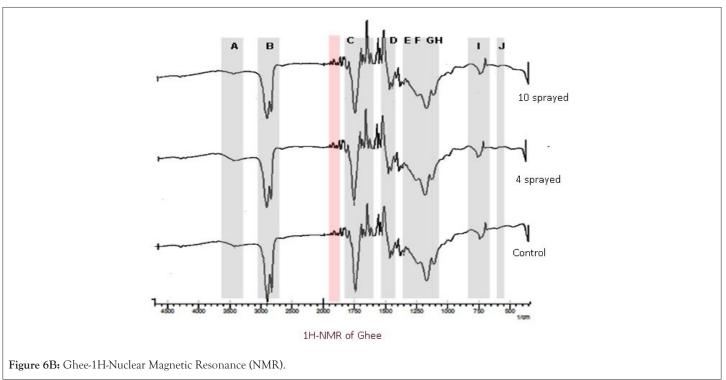
in -C=O and -C-O stretch as compared to control sample. 10 sprayed sample not shown change for trans-fatty acid content but there was increase in 4 sprayed sample.

**Proton NMR-Ghee:** Minor molecular environment variations of methyls and esters groups caused by MIRGA sprayings were found (Supplementary text part II (b)) (Figure 6B) (Tables S8A and S8B).

#### Nutritional value analysis

From Table 4, four spraying has reduced the fat and carbohydrate content and increased the protein content, than in control. While 10 spraying has reduced the fat content, and increased the carbohydrate and protein content.





### Table 4: Nutritional value analysis-ghee.

No of sprayings given	Protein (%)	Carbohydrate (%)	Lipid (%)
Control	0.1	0.23	0.52
4 sprayed	0.67	0.16	0.45
10 sprayed	1.07	0.21	0.44

## Milk powder instrumentation results

**GCMS-Milk powder:** Control sample contains majorly 1-Tridecene and Oleic acid. In 4 sprayed sample, there was major peak of 6-Octadecenoic acid and 1-Tridecene; also there was additional peak of cis-Vaccenic acid, which are responsible for enhancement of sweetness characters. On other hand, 10 sprayed sample has decrease in peak of 13-Octadecenal and 13-Hexyloxacyclotridec-10-en-2-one, but 1-Tridecene peak was found to be increased. This explained the reduction of sweetness of milk powder after 10 spraying. 6-Octadecenoic acid most abundant in 4 sprayed sample followed by 10 sprayed and control samples (Figure 7A) (Table S9).

**FTIR-Milk powder:** There is C=O bond stretching near 2340 cm<sup>-1</sup> and broad O-H at 3400 cm<sup>-1</sup> in 10 sprayed sample which implicates the prescence of a C=O from fatty acid (Figure 7B).

**PXRD-Milk powder:** PXRD shows a single broader peak at 19.5°. The area under the peak in the range 14° to 26° is: Control 91844; 4 sprayed 98534; and 10 sprayed 100182. MIRGA spraying has caused significant improvements in the crystallinity of the powders, and also lactose crystallization observed. The progress of the lactose crystallization is observed from the increasing intensities of the peaks at 19.5° (Figure 7C) (Table S10). The broad peak is due to the presence of polycrystalline substance. Crystallinity is a favorable factor which is always welcomed by the consumer.

**Proton NMR-Milk powder:** The total integrated area for sugar at 3-6 ppm range is around 62.32 and 67.69 in control and 4 sprayed samples respectively. This increased sugar content is responsible for the enhanced sweetness in 4 sprayed sample. In 10 sprayed, the

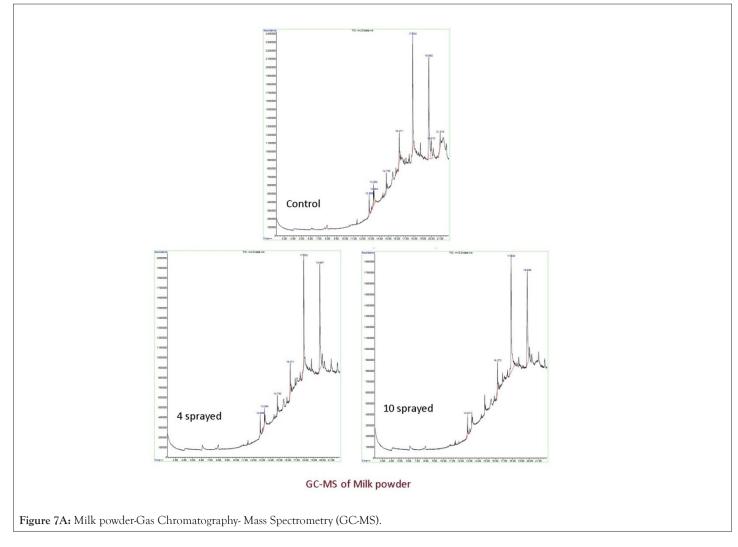
## OPEN OACCESS Freely available online

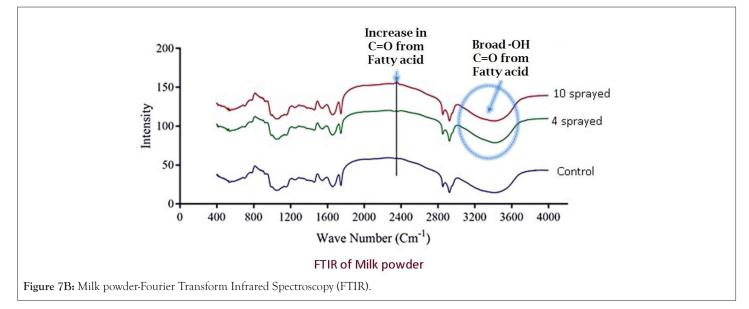
total integrated area is around 62.12, i.e., less than that of control, which is the reason for reduced sweetness in 10 sprayed sample (Supplementary text part II (c)) (Figure 7D) (Table S11).

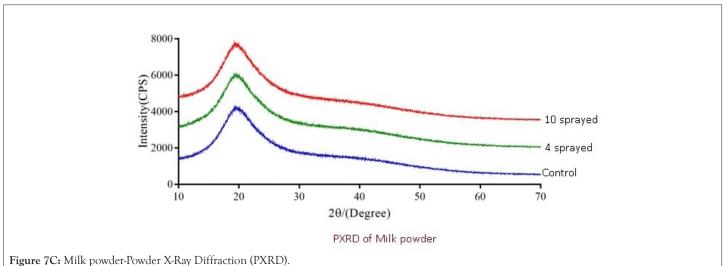
HR-TEM-Milk powder: Control, 4 sprayed and 10 sprayed samples showed spherical particles of about 400 nm in size, without agglomeration, no significant differences were found (Figure 7E). Whereas, elemental analysis has brought up the following differences: Copper content is 0.90%, 0.53% and 1.15% in the control, 4 sprayed and 10 sprayed milk powder samples respectively (Table S12). Reduced and increased copper content than in the control have caused better and poor tastes respectively in the 4 spraved and 10 spraved samples. Calcium has increased progressively with increased number of sprayings. The analysis revealed carbon and oxygen which are due to the fats, proteins and carbohydrates, and additionally the presence of copper, calcium, phosphorus, and sulfur. Calcium is a typical element presence in milk in the form of calcium phosphate. And sulfur is detected and is due to the presence of methionine and other sulfur components of proteins.

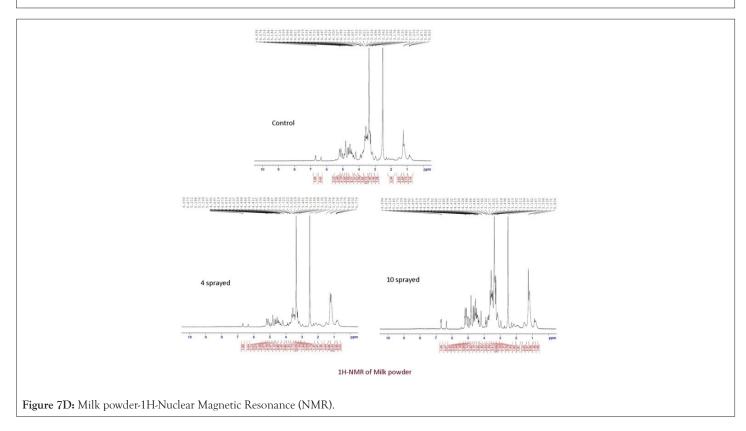
### 3-Dimensional fluorescence spectroscopy-milk powder

The samples are fluorescence active. There is more electron density in contour region in the fluorescence diagram of 10 sprayed than 4 sprayed sample. The increased fluorescence intensity after spraying is because of the unfolding of the protein exposing the tryptophan residues. There is excitation at Ex 290 nm and emission peak around 440 nm.This excitation dependent emission due to distribution of different particle size emission (Figure 7F).









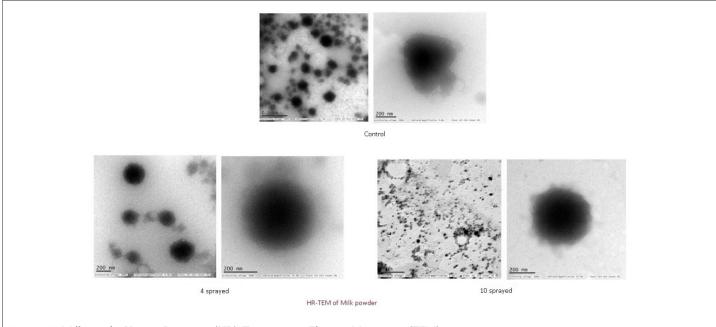
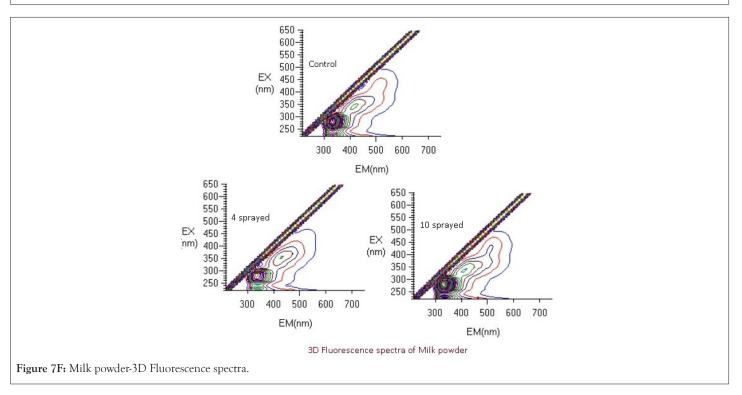


Figure 7E: Milk powder-Human Resources (HR) -Transmission Electron Microscopy (TEM).



#### Cheese powder instrumentation results

**FTIR-Cheese powder:** Peak 3402 cm<sup>-1</sup> is responsible for the presence of the O-H, it is a part of water peak and OH group from carbohydrates; compared to control, square of this peak is bigger by 18% and 34% in 2 and 7 sprayed samples respectively. Peak 2952 cm<sup>-1</sup> is responsible for the presence of the C-H and is characteristic of carbon chain; square of this peak is bigger by 50% and 92% respectively in 2 and 7 sprayed samples. Peak 2855 cm<sup>-1</sup> is responsible for the presence of the C-H and is characteristic of carbon chain; square of the C-H and is characteristic of carbon chain; square of the C-H and is characteristic of carbon chain; square of the C-H and is characteristic of carbon chain; square of the greak is bigger by 47% and 76% in 2 and 7 sprayed samples respectively; Peak 1748 cm<sup>-1</sup> is responsible for the presence of the C=O which is determined as part of lipids; square of this peak is bigger by 54% and 87% in 2 and 7 sprayed sample. Peak 1630 cm<sup>-1</sup> responsible for the presence of the N-H here is a part of proteins; square of this peak is smaller by 34% and 55%

respectively in 2 and 7 sprayed samples. Peak 1118 cm<sup>-1</sup> responsible for the presence of the C-O group is a part of carbonhydrates; square of this peak is bigger by 8% and smaller by 28% in 2 and 7 sprayed samples respectively. Peak 1026 cm<sup>-1</sup> responsible for the presence of the C-O group is determined as part of carbonhydrates; square of this peak is bigger by 33% and 7% in 2 and 7 sprayed samples respectively (Figure 8A).

In general, the peak areas increased in 2 and 7 sprayed samples. It is noted that, in which the amount of moisture is directly proportional. Also, spraying resulted in splitting of protein structures and carbohydrate breaking.

**PXRD-Cheese powder:** All three samples have one broad peak centred on doublet at 2 $\Theta$ =doublet 18° and 19° with no apparent change in breadth observed. Separation of doublet peak is most prominent in 7 sprayed followed by control. Merging of these two

#### OPEN OACCESS Freely available online

peaks occur at 2 sprayed. The separation of two peaks indicate occurrence of two different reflecting planes in the crystal. Here the crystal structure becomes more defined. The intense peak at  $31.80^{\circ}$  of 2 sprayed decreased in terms of its ratio with the intensity of the broad peak compared to control and 7 sprayed. The decrease of this peak's intensity allowed formation of minority crystalline phases as shown by the presence of peak on  $2\Theta$ =13.29° and occurrence of several peaks beyond 50.00°. The peak around 13° is slightly visible in 7 sprayed (Figure 8B).

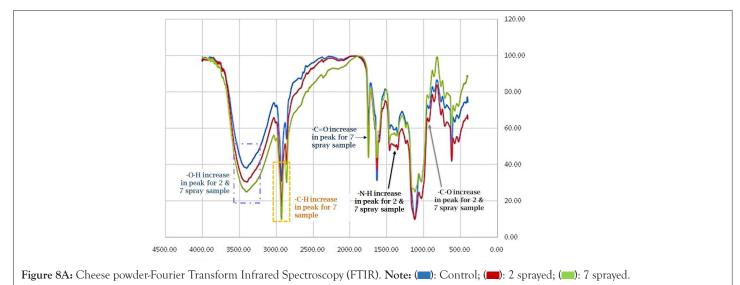
**Proton NMR-Cheese powder:** Control sample shows a profile compatible with a fat-rich sample with certain content of sugars and other minor compounds. Some lipids and minor compounds are responsible for the organoleptic features of cheese. In this context, the 2 sprayed sample shows an increase in some signals which is related to flavor and taste, and a reduction in other signals that can be related to the sourness of cheese. In 7 sprayed sample, decrease observed in some signals is related with the loss of flavor and taste (Supplementary text part II (d)) (Figure 8C).

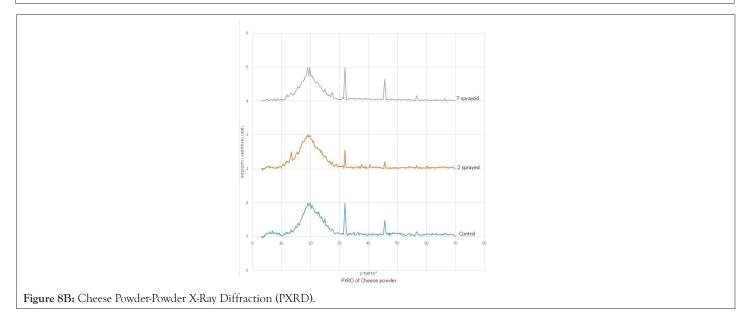
**TEM-Cheese powder:** Control sample has semi-spherical particles (main diameter) 0.4-1  $\mu$ m, nanoparticles 10-40 nm and nano aggregates 20 × 15 nm<sup>2</sup> (average). Large semi spherical particles (mainly nano aggregates), are characterized by a darker area in the centre due to mass contrast.

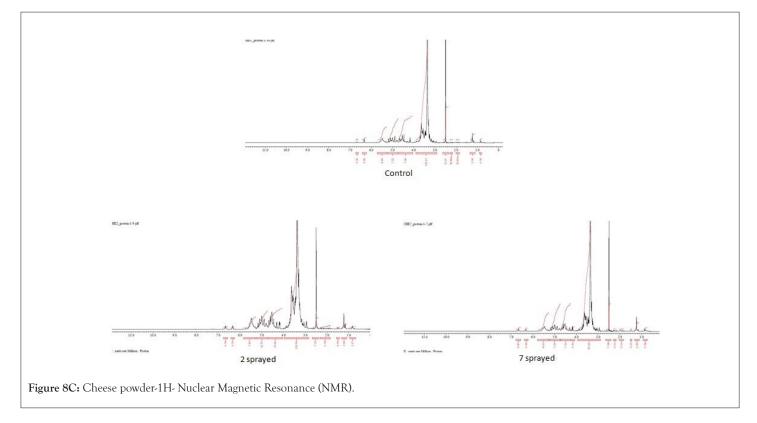
In the 2 sprayed sample, nano-sized aggregates are absent, semi-spherical particles size are about 1-2  $\mu$ m and some of them seem to have lost symmetry in mass distribution within particle. The said characteristics in darker area are shifted to the left and the circular contour is lost, with respect to "regular" particles. Nanoparticle size is reduced to 1-20 nm and less homogenous areal distribution than in the control.

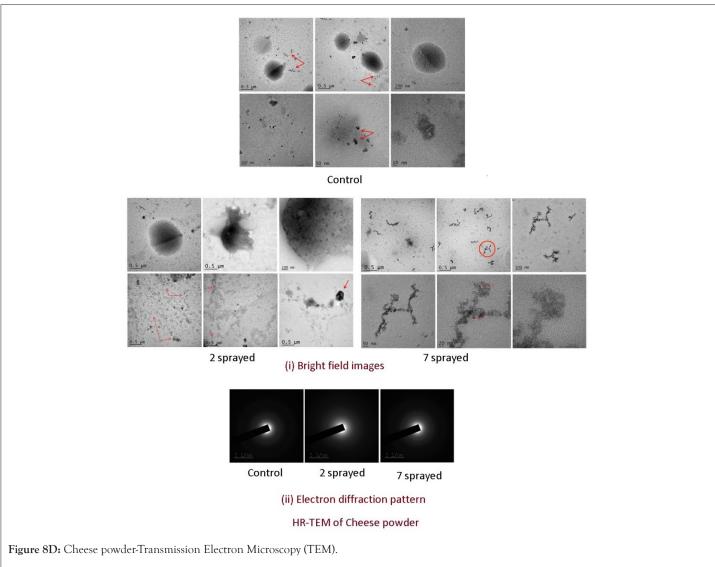
For the 7 sprayed sample, matrix structure of the control sample appeared to be completely lost, semi-spherical particle and nano particles are absent. Although aggregates are still observed, but the shape is very different than control sample, being chain like size perimeter for small ones 20-50 nm and 0.2-1  $\mu$ m for large ones. The chain like aggregates appear to be formed by sequence of individual units, linked by chemical bonds, similarly to fresh soot chains formed by tail pipe emission of vehicles.

Increasing number of spraying from 2 to 7 have caused significant alterations of the sample texture which concerns mainly the 7 sprayed sample and, to minor extent, the 2 sprayed sample. More specifically, 2 spraying affected the size, shape and mass distribution of semi-spherical particles, and the size (smaller in the 2-sprayed sample) and areal distribution (uneven in the 2 sprayed sample) of dark nanoparticles. In 7 sprayed sample, there is significant loss of the original sample structure. While 2 sprayed sample remained comparable to control, in the 7 sprayed sample, a completely different matrix structure is observed (Figure 8D).









## DISCUSSION

## Milk

(Raw milk, boiled raw milk, chilled raw milk and pasteurized milk)

Milk shelf life is influenced by various factors such as moisture and oxygen uptake, by the use of specialized heat exchangers and hygienic packaging technologies [27]. The addition of Plantaricin FB-2 extends the shelf life but has its own limitations [28]. Nonthermal technologies *viz.*, UV-C treatment, infrared treatment with 880-940 nm and ultrahigh temperatures also enhances the shelf life but affects the organoleptic properties of milk [29-31].

Five-times MIRGA sprayed milk samples have shown significantly enhanced sweetness, taste and flavor than the control and 2 sprayed samples. Whereas, excessive number of sprayings i.e., 12 spraying has led to undesirable taste and flavor of the milk. In raw milk, compared to the control, 5 MIRGA spraying has extended the shelf-life of raw milk by 300% at room temperature and by 1300% at refrigeration. Likewise, 5 sprayed-boiled milk, chilled milk and pasteurized milk samples-have shown extended shelf life by 300%, 200% and 200% respectively at room temperature; and by 650%, 1450% and 260% at refrigeration.

To infer from the physicochemical results, 5 sprayed is the best milk in relation to metabolite composition. Lactose degraded in to glucose and galactose and increased the sweetness of the milk [32-34], hence suitable for lactose intolerant patients. Lactose intolerance disease is quite commonly found in human beings these days. Lactose free dairy products are becoming popular and fetch high commercial value [35]. Manufacturing of lactose free products is a hard task to industries and is costlier [36,37]. Thus, a feasible solution is to simply apply MIRGA spraying to the milk products in order to make it lactose free.

The spraying of mid-IR also resulted in production of butyrate. Butyrate, a short chain of fatty acid and a potent anti-inflammatory, is a fuel for intestinal epithelial cells with vast health benefits against cholesterol, obesity, and cardiac and intestinal diseases [38]. It has many health benefits as cited by [39-41]. Research is yet challenging to concentrate and isolate butyrate [42,43], but MIRGA spraying is shown to be an effective method to concentrate butyrate content in dairy products. Besides these benefits, concurrent enhancement of sweetness, taste, flavor and shelf-life with MIRGA are also notable.

Milk, being the primary source for all other dairy products and dairy foods, superior quality and minimal processing is expected. Comparing to the present complex, costlier and limitation-rich technologies (Table 5), MIRGA is a viable method to simultaneously enrich the milk's shelf-life, sensory attributes and health benefits.

 Table 5: Existing technologies to increase the shelf life of milk.

SI.No	Techniques	Shelf life	Practical drawbacks
1.	HTST	10-14 days	Little cooked flavor
2.	UHT	180 days	Definite cooked flavor
3.	Extended shelf- life (ESL) or ultra-pasteurized	30-60 days	Mild cooked flavor
4.	Bactofugation	Up to 21 days	Insufficient shelf life extension
5.	Microfiltration	Up to 30 days	Insufficient shelf life extension

## OPEN OACCESS Freely available online

6.	UV-C treatment to Pasteurized trim milk	Up to 53 days	Off flavors in milk products. UV is an ionizing radiation, hence needs safety and specialized operating skill.
7.	MIRGA (2-6 μm mid-IR)	2-51 days	No drawbacks occurred; but sweetness, taste and flavor significantly increased.

## Buttermilk

Shelf life of buttermilk as of now is enhanced only by refrigeration techniques. Among the control, 4 sprayed, 10 sprayed and 16 sprayed buttermilk samples, 10 sprayed sample has shown enhanced taste and flavor and desirable sourness. At the same time, 10 sprayed sample has acquired extended shelf life by 200% and 100% at room temperature and refrigeration respectively, than the control.

To sum up from the instrument tests, 10 sprayed sample is more preferable with respect to metabolite composition *viz.*, increased hydroxybutyrate and aroma enhancing compounds, slightly degraded triglycerides, and unaltered lactic acid which however is easily digestible. Most important is that the 10 sprayed sample has higher concentration of hydroxybutyrate which is a potent antiinflammatory metabolite and will be better for the patients with Inflammatory Bowel Disease and Irritable Bowel Syndrome.

## Curd

Four sprayed curd sample has exhibited enhanced taste [44]. The consumer panel has described it as 'very tastier' than the control, 8 sprayed and 14 sprayed samples. Consequently, shelf life of the 4 sprayed curd extended by 450% and 90% at room temperature and refrigeration respectively. The result is more than the recently evaluated antimicrobial protein-based edible coating to the curd by [45].

Physicochemical tests showed that 4 sprayed curd has metabolic profile of short chain fatty acids similar to that of control, but with increased taste and aroma, hence preferable. Short chain fatty acids are known to have anti-inflammatory, antitumorigenic, and antimicrobial effects [46]. Whereas, in 8 and 14 sprayed samples, excess number of sprayings led to undesirable metabolites generation.

Fermented milk products-butter milk and curd-hold an important commercial place in dairy industry. MIRGA, as a cost-effective and health-promoting tool, fortifies the curd and buttermilk besides increasing their shelf-life and sensory qualities.

#### Butter

2 sprayed butter sample evinced enhanced taste and flavor. The consumer panel has defined the consistency of 2 sprayed butter sample as having preferable smooth and creamy consistency. More than 2 spraying has further softened the consistency of butter, and 10 sprayed sample was found to have lost its consistency and appeared very soft. At room temperature, 2 sprayed butter has 100% more shelf life than the control, and in refrigeration, has 64% more shelf life, than the control.

Butter is a fat rich dairy product, with some fatty acids being beneficial and some being non-beneficial to health. Stearic acid, a saturated fatty acid, has favorable effects on blood-lipid concentrations, and its biological effects are quite different to those of palmitic and myristic acids [47]. Myristic acid and palmitic acid have hypercholesterolemic effect, whereas stearic acid is essentially neutral [48]. It is notable from the physicochemical analyses that 2 spraying has selectively increased the stearic acid content by two fold, but reduced the palmitic acid level by 3 fold and completely removed the myristic acid. Application of MIRGA has induced manifold increase in the oleic acid which is infamously known to decrease the risk of coronary heart disease, cardio-metabolic risk, obesity, type-2 diabetes and hypertension; recent findings also have suggested potential protective effects on the promotion and progression of several human cancers [49].

MIRGA spraying has led to chemical changes as put forth in FTIR and NMR spectral analysis, this is consistent with the findings of [50], who employed gamma irradiation at doses up to 10 kGy and found no negative impact on sensory quality attributes of sheep butter. Whereas, in this study, 2-6  $\mu$ m mid-infrared from MIRGA has additionally given positive influence on sensory attributes, and the 2 sprayed butter sample has enriched fatty acid profile than the other samples.

### Ghee

Four sprayed ghee sample has enhanced taste and flavor than the control, and upon 10 spraying the ghee became tasteless. 4 sprayed ghee has 30% extended shelf-life at both room and refrigeration temperatures.

In dairy sector, ghee flavour is enhanced by enzyme modified ghee flavour technique through the addition of chemicals but is less effective [51]. To enhance the shelf-life of ghee, chemical antioxidants are added. Whereas, traditionally used natural antioxidants like guava leaf extract and betal/tulsi leaves were proved to be best [52]. As the high cholesterol content of ghee possess health risks, low cholesterol ghee are prepared using adsorbents  $\beta$ -cyclodextrin [53]. The disadvantages of these methods are, laborious and timeconsuming and involve coherent extraction of ghee's other natural nutritious and health promoters, e.g., 75% vitamin-D including flavour are removed in cholesterol reducing process [54], therefore a better technology in this aspect is still a vital need.

By and large, the above disadvantages were overcame by 4 MIRGA sprayings which resulted in ghee's improved flavour and taste, 30% shelf-life enhancement and increased dairy-based trans-fatty acids of ruminant origin which are hepato and cardioprotective [55]. Among the dairy products trialed, ghee samples were found to have the least extension in their shelf life with MIRGA.

Ghee, a type of clarified butter, has more fat content than the butter itself, and was considered as a cause of cardiovascular diseases. Therefore, nutritional content, targeting the fat, was estimated. 4 spraying has reduced the fat content by 15.6% and increased the protein content by many folds (Table S13), and is recommendable for reducing Low-Density Lipoproteins (LDL) associated risks.

Unlike other dairy products, the quality of ghee has stronger and direct influence on ghee-made food stuffs. Therefore, 200 gms of control, 4 and 10-sprayed ghee were used to prepare potato chips with 50 gm of potato. And it is found that the quantity requirement of 4 and 10-sprayed ghee was 34%-71% less when compared to the non-sprayed ghee quantity required to make the potato chips (Table S9). Thus it is expected that, with use of the 4 sprayed ghee, quantity of ghee required for cooking dishes could be reduced than the usual, but still impart desirable taste and aroma to the dishes.

#### Milk powder

On contrary to the liquid milk in which 2 sprayings were sufficient to enhance the sweetness, milk powder required upto 4 MIRGA

## OPEN ORCESS Freely available online

sprayings. Shelf life trial was not conducted with milk powder as they usually have prolonged shelf life of 6-12 months.

However, considering the powdered nature, the milk powder samples were studied using TEM in which elemental analyses evidenced the quantitative changes among the trace elements viz, Cu, Ca, P and Sulfur, caused by 2-6  $\mu$ m mid-IR treatment. This is in accordance with the changes in trace elements between UHT milk and OH (overheated) milk reported by [56]. In the present study, remarkable changes were obtained with the Cu and Ca. 4 spraying has reduced the Cu content by 1.6 fold [57], has presented similar reduction of Cu in UHT processed milk than in the raw milk. Meanwhile, Ca content was increased by 4.4 folds in 4 sprayed sample.

In a nutshell, 4 MIRGA sprayings have reduced copper content, and increased the sweetness, crystallinity, calcium content and also the 6-Octadecenoic acid, (Z)-which is an anti-cancerous, anti-bacterial, immune-stimulant and anti-inflammatory compound [58-60].

## Cheese powder/cheese

The addition of enhancers such as yeast extracts are being used to enhance the cheese flavor [61]. Using MIRGA two sprayed cheese powder and firm cheese has enhanced flavor, and reduced saltiness and sourness; and 7 sprayed samples were of poor sensory qualities. Available technologies to enhance the keeping quality of cheese include hyperbaric storage, edible coatings with active compounds, non-thermal techniques, hurdle technology, smart packaging, natural additives and inoculation of specific culture [62-65]. Cheese powder, like milk powder, having longer shelf life is not trialed for shelf life; instead, 2 sprayed hard cheese sample was trailed and found to have acquired 100% and 61% more shelf life in room and refrigeration temperatures, than the control.

In general, to enhance the shelf-life of cheese, ginger/garlic extract, herbs, chemical preservative, modified atmospheric packaging, active/edible coating, high pressure and integrated methods are applied. These methods are expensive and have influence on the chemistry and sensory attributes of cheese, further research for a better alternative is expected [66-68]. Two times MIRGA sprayed cheese powder/firm cheese has attained an improved flavor, extended shelf-life, and chemical compound level changes, thus making it more palatable and stable than control.

## Action of 2-6 µm mid-IR on the dairy products

Analysis of all the instrumentation data indicates that 2-6 µm mid-IR has altered the chemical bond, structural morphology and chemical compound transformation including chemical shifts, intermolecular modification, etc. The chemistry behind is, in our Earth, everything is made up of atoms and bound together by chemical bonds [69,70]. The atoms constantly vibrate in certain frequency and the vibrational frequencies of most of the earth molecules lie in the mid-IR region [71,72], has reported the absorption wavelengths of chemical groups of waters, sugars, lipids and proteins of food components to be from 2.7 to 5.92 µm [73]. Thus, the generated 2-6 µm mid-IR from MIRGA have passed through the packaged material and got absorbed by the inside constituent molecules of dairy-products . This interaction induced changes in the electronic, vibrational, and rotational states of atoms and molecules, internal chemical bonds are altered [74,75], ultimately causing changes in structure, chemical modifications [76,77]. Thus favorable changes in the physicochemical characters of dairy products are attained [78]. It is notable that, every spraying imparted its unique changes in the chemistry and sensory characteristics of dairy products.

## OPEN OACCESS Freely available online

Similar desirable results in coffee, tea, cocoa and edible salts were achieved using MIRGA spraying by [79,80].

# Proposed role of MIRGA in energy conservation in dairy industry

In dairy industry, energy involved in the conversion of raw milk to consumable dairy product is majorly spent in the form of electricity, and 50%-60% of total electricity is spent for refrigeration purposes [81]. A study has forecasted the increase in cost of electrical energy and will consequently impact on the cost of dairy products, and suggested that even a slight change in practice will lead to appreciable energy conservation. Considering these, the shelf-life enhancement of the dairy products achieved with MIRGA spraying is noteworthy, i.e., from 30% to >1000%, therefore MIRGA will remarkably aid in cutting down the energy costs throughout the supply chain of transportation-chilling-processing-market.

Invention background, definition, technique of mid-IR generation from MIRGA, toxicological study on MIRGA, safety of the MIRGA sprayed usables and primeval and future scope of MIRGA have been described [82,83]. (Supplementary text part III).

## Pros and cons

The main advantages are:

- Infrared irradiation is non-ionizing and is considered advantages due to its higher thermal efficiency, fast heating rate, reduced processing time and energy consumption, improved product quality and food safety, and being environment friendly.
- The mid-IR has to be applied only externally over the packaged dairy products, with no direct contact to the inside content.
- MIRGA by generating specific wavelength of 2-6 µm has overthrown the major limitation of wide spectral range offered by the existing infrared emitters [84].
- MIRGA is a versatile, simple and compact equipment, with low manufacturing cost i.e. 300 sprayings emitting MIRGA costs only around USD 0.35. There is no operational/maintenance cost required.
- No special skills are required to operate MIRGA and is easy-tohandle (like a perfume body spray) for users from industry to consumer level.
- Dairy products are preserved with extended shelf life in a single MIRGA treatment and no repeated/periodical treatment is required.
- Changes in nutritional value and health benefits of sprayed dairy products are favorable6 and can be advocated to avoid dairy-associated diseases.
- The applied 2-6 µm mid-IR was not found to have leached the polythene packaging material of different micron-level thicknesses.
- No short term/long term adverse effects were noted upon consumption of MIRGA sprayed dairy foods.

The only limitation felt is that excessive MIRGA sprayings found to have negatively influenced the shelf life and sensory attributes of dairy products, so care should be taken to avoid over use of MIRGA.

## CONCLUSION

In order to prevent the dairy products from storage and reduce the losses to dairy industry in terms of electricity consumption for cold storage, application of 2-6  $\mu$ m mid-IR generated from MIRGA has promising results in extending their shelf-life with enhanced consumer sensory acceptance, and also to manage and prevent dairy-associated diseases such as lactose intolerance, irritable bowel syndrome and cardio-metabolic risks. With the increasing demand for quality rich, healthier dairy products, mid-infrared irradiation may serve as a suitable means of strengthening the sustainable food production and food safety. There are definitely more avenues that exist than experimented up to now, potentially open for future research using the MIRGA platform.

## ACKNOWLEDGEMENT

Authors thank multi-Faculty scientists of different labs, institutions, universities, etc., around the world for their technical guidance and help; also thank Dr.George Tranter, Chiralabs Ltd., Begbroke Centre for Innovation and Enterprise, Oxfordshire, UK; Dr.Jan IC Vermaak, Manager of Engineering, Nuclear Science Center-Texas A&M University, USA; Dr.TakashiroAkitsu, Professor, Department of Chemistry, Faculty of Science, Tokyo University of Science, Japan; Dr.Kam-Hung Low, X-ray Facility manager, Department of Chemistry, The University of Hong Kong; Ms. Satitaphorn Sriphuttha, Tokyo University of Science, Japan; Ms. Shiho Murakami, Mr. Kanai and other Spectroscope specialists of Hitachi High-Tech, Japan; Mr. Gary Powell, Lightwind corporation, Petaluma, California, USA; Dr. Ramakrishnan, Head, Indian Veterinary Research Institute, Mukteshwar, India; Dr. R Prabhakaran, Assistant Professor, Department of Chemistry, Bharathiar University, Coimbatore, India; Kolabtree experts; all sensory expert panel members, untrained consumer panel members; and we would also like to apologize to all scientists and other helpers around the world who are not cited here.

## FUNDING

This study received no specific funding.

## CONFLICT OF INTEREST

In accordance with the journal's policy and our ethical obligation as researchers, we submit that the authors Dr.Umakanthan and Dr.Madhu Mathi are the inventors and patentee of Indian patent for MIRGA (under-patent no.: 401387) which is a major material employed in this study.

## AUTHOR CONTRIBUTION

Umakanthan: Conceptualization, Methodology, Project administration, Resources, Supervision, Funding; Madhu Mathi: Investigation, Data curation, Visualization, Writing-Original draft preparation; Umakanthan and Madhu Mathi: Validation, Writing-Reviewing and Editing.

## DATA AND MATERIALS AVAILABILITY

The datasets generated during this study are available in the manuscript and supporting information file.

## ETHICS STATEMENT

The work does not involve use of human or animal subjects.

## REFERENCES

- 1. Ansari JA, Ismail M, Farid M. Extension of shelf life of pasteurized trim milk using ultraviolet treatment. J Food Saf. 2020;40(2):127-168.
- 2. Jo Y, Benoist DM, Barbano DM, Drake MA. Flavor and flavor chemistry differences among milks processed by high-temperature, short-time pasteurization or ultra-pasteurization. J Dairy Sci. 2018;101(5):3812-3828.
- 3. Karlsson MA, Langton M, Innings F, Malmgren B, Höjer A, Wikström M,et al. Changes in stability and shelf-life of ultra-high temperature treated milk during long term storage at different temperatures. Heliyon.

2019;5(9).

- Singh P, Gandhi N. Milk preservatives and adulterants: Processing, regulatory and safety issues. Food Rev Int. 2015;31(3):236-261.
- Cadwallader KR, Singh TK. Flavours and off-flavours in milk and dairy products. Adv Dairy Chem. 2009:631-90.
- Nazari SH. Impact of using infrared irradiation energy in food processing. Energy Educ Sci Technol Part A. 2014;32:1387-1396.
- Waynant RW, Ilev IK, Gannot I. Mid-infrared laser applications in medicine and biology. Philos Trans R Soc London. 200;359:635-644.
- 8. Pereira MF, Shulika O. Terahertz and mid infrared radiation: Generation, detection and applications. 2011.
- 9. Girard J. Principles of environmental chemistry. Jones Bartlett Pub. 2013.
- Aboud SA, Altemimi AB, RS Al-HiIphy A, Yi-Chen L, Cacciola F. A comprehensive review on infrared heating applications in food processing. Molecules. 2019;24(22):4125.
- 11. Sanders RH. Revealing the heart of the galaxy. 2014.
- 12. Verheest F. Waves in dusty space plasmas. 2000.
- 13. Keping S, Yu G. Recent developments in applied electrostatics. 2004.
- 14. Fauchais PL, Heberlein JVR, Boulos MI. Thermal spray fundamentals from powder to part. 2014.
- Umakanthan, Mathi M. Decaffeination and improvement of taste, flavor and health safety of coffee and tea using mid-infrared wavelength rays. Heliyon. 2022;8(11).
- Umakanthan T, Mathi M. Quantitative reduction of heavy metals and caffeine in cocoa using mid-infrared spectrum irradiation. J Indian Chem Soc. 2023;100(1):100861.
- Tishkevich DI, Korolkov IV, Kozlovskiy AL, Anisovich M, Vinnik DA, Ermekova AE, et al. Immobilization of boron-rich compound on Fe3O4 nanoparticles: Stability and cytotoxicity. J Alloys Compd. 2019;797:573-581.
- Dukenbayev K, Korolkov I V, Tishkevich D I, Kozlovskiy A L, Trukhanov S V, Gorin Y G, et al. Fe3O4 nanoparticles for complex targeted delivery and boron neutron capture therapy, Nanomaterials. Nanomaterials (Basal). 2019;9(4):494.
- Kozlovskiy AL, Alina A, Zdorovets MV. Study of the effect of ion irradiation on increasing the photocatalytic activity of WO3 microparticles. J Mater Sci Mater Electron. 2021;32:3863-3877.
- El-Shater RE, Shimy HE, Saafan SA, Darwish MA, Zhou D, Trukhanov AV, et.al. Synthesis, characterization, and magnetic properties of Mn nanoferrites. J Alloys Compd. 2022;928:166954.
- Kozlovskiy AL, Zdorovets MV. Effect of doping of Ce4+/3+ on optical, strength and shielding properties of (0.5x)TeO2-0.25MoO-0.25Bi2O3xCeO2 glasses. Mater Chem Phys. 2021;263:124444.
- Almessiere MA, Algarou NA, Slimani Y, Sadaqat A, Baykal A, Manikandan A, et al. Investigation of exchange coupling and microwave properties of hard/soft (SrNi0.02Zr0.01Fe11.96O19)/(CoFe2O4)x nanocomposites. Mat Today Nano. 2022;18:100186.
- Krishnakumar T. Application of microwave heating in food industry. ResearchGate. 2019.
- Everitt, M. Consumer-targeted sensory quality. Trends Food Sci technol. 2009;117-128.
- Wichchukit, S, O'Mahony M. The 9-point hedonic scale and hedonic ranking in food science: Some reappraisals and alternatives. J Sci Food Agric. 2014;95(11):2167-2178.
- Alm, L. Effect of fermentation on lactose, glucose, and galactose content in milk and suitability of fermented milk products for lactose intolerant individuals. J Dairy Sci. 1982;65(3):346-352.

### OPEN OACCESS Freely available online

- Pasagadi.A. S, Prakash.A. K, Harthikote Veerendrasimha.V. S, Geethambika. S. B, Franklin. M. E. E, Pushpadass.H. A. Shelflife prediction of milk-millet powders. J Food Process Engineering. 2023;46(10).
- Yujun J, Xinyan Y, Haonan J, Xiaohan F, Fang T, Yang S, et.al. Shelf-life prediction and chemical characteristics analysis of milk formula during storage. Lwt. 2021;144:111268.
- 29. Ansari JA, Ismail M, Farid M. Extension of shelf life of pasteurized trim milk using ultraviolet treatment. J Food Saf. 2020;40(2):12768.
- Rosidi A, Muhammad HH. Application of infrared with different waves and its effect on organoleptic of dairy milk. J Anim Husb. 2021;6(1).
- Nivedita, D. UHT Treatment of Milk and Dairy Products. J Dairy Res. 2018.
- 32. Takami SH, Hejazian S, Shendi EG. Overall evaluation of produced flavored low lactose milk powder for lactose intolerants. Mod Appl Sci. 2016;10(3):149-154.
- Suri S, Kumar V, Prasad R, Tanwar B, Goyal A, Kaur S, et al. Considerations for development of lactose-free food. J Nutr Intermed Metab. 2019;15:27-34.
- Torres JK, Stephani R, Tavares GM, de Carvalho AF, Costa RG, de Almeida CE, et al. Technological aspects of lactose-hydrolyzed milk powder. Food Res Int. 2017;101:45-53.
- Dekker PJ, Koenders D, Bruins MJ. Lactose-free dairy products: Market developments, production, nutrition and health benefits. Nutrients. 2019;11(3):551.
- Szilagyi A, Ishayek N. Lactose intolerance, dairy avoidance, and treatment options. Nutrients. 2018;10(12):1994.
- Fialho TL, Martins E, Silveira AC, Silva CR, Perrone ÍT, Schuck P, et al. Lactose hydrolyzed milk powder: Thermodynamic characterization of the drying process. Dry Technol. 2018;36(8):922-931.
- Cavaleri F, Bashar E. Potential synergies of β-Hydroxybutyrate and butyrate on the modulation of metabolism, inflammation, cognition, and general health. J Nutr Metab. 2018.
- Olga L, van Diepen JA, Chichlowski M, Petry CJ, Vervoort J, Dunger DB, et al. Butyrate in human milk: Associations with milk microbiota, milk intake volume, and infant growth. Nutrients. 2023;15(4):916.
- Słupecka-Ziemilska M, Pierzynowski SG, Szczurek P, Pierzynowska K, Wychowański P, Seklecka B, et al. Milk formula enriched with sodium butyrate influences small intestine contractility in neonatal pigs. Nutrients. 2022;14(20):4301.
- Paparo L, Nocerino R, Ciaglia E, Di Scala C, de Caro C, Russo R, et al. Butyrate as a bioactive human milk protective component against food allergy. Allergy. 2021;76(5):1398-1415.
- Conway V, Gauthier SF, Pouliot Y. Buttermilk: much more than a source of milk phospholipids. Anim Front. 2014;4(2):44-51.
- de Freitas Mascarello A, Pinto GI, de Araújo IS, Caragnato LK, da Silva AL, dos Santos LF et al. Technological and biological properties of buttermilk: A mini review. IntechOpen. 2019.
- 44. Ahlawat A, Basak S, Ananthanarayan L. Optimization of spray-dried probiotic buttermilk powder using response surface methodology and evaluation of its shelf stability. J Food Process Preserv. 2022;46(11):e16928.
- 45. Wang C, Zhou S, Du Q, Qin W, Wu D, Raheem D, et al. Shelf life prediction and food safety risk assessment of an innovative whole soybean curd based on predictive models. J Food Sci Technol. 2019;56:4233:4241.
- Stanley JC. Stearic acid or palmitic acid as a substitute for trans fatty acids. Lipid Technol. 2009;21(8-9):195-198.
- 47. Snook JT, Park S, Williams G, Tsai YH, Lee N. Effect of synthetic triglycerides of myristic, palmitic, and stearic acid on serum lipoprotein

- Arsic A, Stojanovic A, Mikic M. Oleic acid-health benefits and status in plasma phospholipids in the Serbian population. Serb J Exp Clin Res. 2019;20(2):3-8.
- Santos IC, Pinto J, Pimenta AI, Madureira J, Matos P, Viegas C, et al. Use of gamma radiation in sheep butter manufacturing process for shelf-life extension. Int. Dairy J. 2017;71:43-49.
- 50. Debnath PP, Ghosh BC. Enzyme modified ghee flavour: Application and shelf life studies. Int J Pure Appl Biosci. 2018;6:822-826.
- Tan J, McKenzie C, Potamitis M, Thorburn AN, Mackay CR, Macia L. The role of short-chain fatty acids in health and disease. Adv Immunol. 2014;121(1):91-119.
- Aditya N, Divya MP. A study on incorporation of natural antioxidants for the shelf life extension of ghee. Int J Eng Res App. 2018;8(1):10-16.
- Bhatia P, Sharma V, Arora S, Rao PS. Effect of cholesterol removal on compositional and the physicochemical characteristics of anhydrous cow milk fat (cow ghee). Int J Food Prop. 2019;22(1):1-8.
- Tahir MN, Bokhari SA, Adnan A. Cholesterol extraction from ghee using glass beads functionalized with beta cyclodextrin. J Food Sci Technol. 2015;52(2):1040-1046.
- 55. Mozaffarian D. Natural transfat, dairy fat, partially hydrogenated oils, and cardiometabolic health: The Ludwigshafen risk and cardiovascular health study. Eur Heart J. 2015;37(13):1079-1081.
- Seiquer I, Andrade DC, Haro A, Navarro MP. Assessing the effects of severe heat treatment of milk on calcium bioavailability: In vitro and in vivo studies. J Dairy Sci. 2010;93(12):5635-5643.
- 57. Akhtar S, Ismail T, Riaz M, Shahbaz M, Amin K. Minerals and heavy metals in raw and ultra heat treated commercial milks in Pakistan. Int J Food Allied Sci. 2015;1:18-24.
- Abubakar M, Majinda R. GC-MS Analysis and Preliminary Antimicrobial Activity of Albizia adianthifolia (Schumach) and Pterocarpus angolensis (DC). Medicines. 2016;3(1):3.
- Campos SH, Souza PR, Peghini B, Silva JD, Cardoso C. An overview of the modulatory effects of oleic acid in health and disease. Mini Rev Med Chem. 2013;13(2):201-210.
- 60. European Patent Office. Application number: 99204230.9. Date of publication: 28.06.2000 Bulletin 2000/26.
- Duarte RV, Lopes-da-Silva JA, Gomes AM, Delgadillo I, Barba FJ, Saraiva JA. Improving fresh cheese shelf-life through hyperbaric storage at variable room temperature. J Food Sci. 2023;88(1):391-402.
- 62. Prajapati P, Garg M, Singh N, Chopra R, Mittal A. Extension of shelf life of paneer-an indian variety of soft cheese: A review. Asian J Dairy Food Res. 2022;1(1):1-9.
- Li Z, Hui L, Yunzhi C, Zhengjian Z, Xiaojun M. Extension of cheese shelf-life using chitosan-procyanidin composite film with high antioxidant activity. 2021;1(1):410-418.
- Zappia A, Branca ML, Piscopo A, Poiana M. Shelf life extension of mozzarella cheese packed in preserving liquid with calcium lactate and bergamot juice concentrate. J Dairy Res. 2020;87(4):474.479.

## 65. Junior BR, Tribust AA. Use of nisin and bioprotective lactic cultures to extend the shelf life of sheep and goat cheese whey. Food bioscience

OPEN ACCESS Freely available online

- to extend the shelf life of sheep and goat cheese whey. Food bioscience. 2022;102096.
- 66. Kongo JM, Malcata. Cheese: Processing and sensory properties. 2016
- Jalilzadeh A, Tuncturk Y, Hesari J. Extension shelf life of cheese: A review. Int J Dairy Sci. 2015;10(1):44-60.
- 68. Ritota M, Manzi P. Natural preservatives from plant in cheese making. Animals. 2020;10(4):749.
- Ganesan, PC. Pendulum power of guidance and pyramid power for success. Sura books (pvt) ltd. 2003;8.
- Alberts B, Johnson A, Lewis J, Raff M, Roberts K, Walter P et al. The chemical components of a cell. Nat. Rev Mol Cell Biol. 2002.
- Toor F, Jackson S, Shang X, Arafin S, Yang H. Mid-infrared lasers for medical applications: Introduction to the feature issue. Biomed Opt Express. 2018;9(12):6255-6257.
- Rosenthal I. Electromagnetic radiations in food science. Advances in food research. 1953;(3)119-196.
- Krishnamurthy K, Khurana HK, Soojin J, Irudayaraj J, Demirci A. Infrared heating in food processing: an overview. compr rev food sci f. 2008;7(1):2-13.
- 74. Tsai SR, Hamblin MR. Biological effects and medical applications of infrared radiation. J Photochem Photobiol B. 2017;170:197-207.
- Flynn GW, Parmenter CS, Wodtke AM. Vibrational energy transfer. J Phys Chem. 1996;100(31):12817-12838.
- Atkins P, De Paula J. Elements of physical chemistry. Oxford University Press, USA; 2013.
- Datta SN, Trindle CO, Illas F. Theoretical and computational aspects of magnetic organic molecules. Sci World J; 2013.
- Esmaeili K. An overview of Viremedy as an information-including medicine, and a call for a Funded Study. J Complement Med Res. 2023;14(6):81-122.
- Umakanthan T, Mathi M. Increasing saltiness of salts (NaCl) using midinfrared radiation to reduce the health hazards. Food Sci Nutr. 2023.
- Umakanthan, Madhu Mathi, 2023d. Potentiation of Siddha medicine using Muppu (Universal Potentiator). Int j pharm res appl. 2023; 8(4); 2070-2084.
- Prabhakar PK, Srivastav PP, Murari K. Energy consumption during manufacturing of different dairy products in a commercial dairy plant: A case study. Asian J Dairy Food Res. 2015;34(2):98-103.
- 82. Mijailovic N, Nesler A, Perazzolli M, Aït Barka E, Aziz A. Rare sugars: recent advances and their potential role in sustainable crop protection. Molecules. 2021;26(6):1720.
- Rysstad G, Kolstad J. Extended shelf life milk advances in technology. Int J Dairy Technol. 2006;59(2):85-96.
- Kapadiya DB, Aparnathi KD. Evaluation of commonly used herbs to enhance shelf life of ghee against oxidative deterioration. J Food Process Preserv. 2018;42(7):e13658.