



Sensory Quality and Shelf-life Enhancement of Vegetables Irradiated with Mid- infrared

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Authors' contributions

This work was carried out in collaboration among all authors. Author UT did the conceptualization, methodology, supervision, validation of the study. Author MM did the data curation, investigation, visualization, writing - original draft preparation of the manuscript. Author UU did the project administration, resources of the manuscript. Authors UU and MM did the writing- reviewing and editing of the manuscript. All authors read and approved the final manuscript.

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ABSTRACT

Vegetables play a highly significant role in our daily dietary intake. The inherent perishability of vegetables restricts their exportation and necessitates early consumption. Consequently, ongoing research endeavors aim to develop technologies that can extend the shelf life of vegetables. In this study, we subjected vegetables to irradiation using a mid-infrared ray with a wavelength of 2-6 μm . To achieve this, we utilized a recently invented atomizer device termed MIRGA (Mid-infrared generating atomizer). During spraying of MIRGA, the contained chemical solution is expelled at a determined plunger pressure, leading to the oscillation of ions in the sprayed mist, thereby

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generating 2-6 μm mid-infrared. This mid-infrared energy exerts its influence on the chemical bonds within the vegetable molecules, resulting in improvements in taste, aroma, overall sensory satisfaction, and shelf life. Notably, the shelf life of vegetables increased by a range of 16-900% and 9-314% (duration) when stored at room temperature and refrigerated, respectively. These beneficial alterations, which are mediated by the 2-6 μm mid-infrared ray, are substantiated through a variety of instrumentation and sensory expert panel results that elucidate the physicochemical characteristics of the vegetables. Employing MIRGA in various domains of food technology represents a plausible and viable option.

Keywords: MIRGA; 2-6 μm mid-infrared; vegetables; irradiation; shelf life; sensory attributes; enhancement; economical.

1. INTRODUCTION

Vegetables are vital for a healthy diet as they provide essential nutrients, dietary fiber, and phytochemicals, while reducing the risk of diseases and medical conditions [1]. They also contribute to the sensory appeal of a diet through their taste and flavors [2]. Important taste compounds in vegetables include organic acids, free sugars, and amino acids, which give vegetables their sour, sweet, and umami tastes [3]. The consumption of fruits and vegetables is recommended by organizations like FAO/WHO to prevent chronic noncommunicable diseases and mitigate micronutrient deficiencies [4]. However, despite their importance, the intake of fruits and vegetables remains low for a majority of the global population [5]. Owing to the lack of proper storage technology, daily tons of vegetables are thrown away, and the exact quantity calculation is very hard. Around 2.5 decades ago, we consumed "in-season" fresh fruits and vegetables. Trade and commerce now forced to develop an extended shelf life and possibly an enhanced sensory attributes. Although shelf life is extended by conventional, physiological, biochemical and biotechnological techniques, the results are not consistent. Therefore, there is a need for research and action to make vegetables more available, accessible, and desirable through various mechanisms at different levels.

To improve the taste of vegetables, various methods can be used. One method involves using non-continuous light irradiation and a light source illumination device, which can enhance the taste of vegetables in an artificial light type plant factory [6]. Another approach is to use vegetable powder, which can be added to food products to improve their taste, flavor, and texture [7]. Additionally, a body taste improver, such as a long-chain highly unsaturated fatty acid, can be used in vegetable fats to enhance

their taste [8]. Another method is to add sucralose after frying or baking the vegetables or the food in which the vegetables are used, which can improve their flavour [9]. Furthermore, organic production of vegetables has been found to improve their quality, taste, and flavor by increasing their dry-matter content, vitamin C, phenolic compounds, and protein content [10].

The shelf life of vegetables can be improved through various methods such as the use of edible coatings, bio preservation, and biotechnological approaches. Edible coatings, made from substances like Aloe vera gel and Hibiscus rosa-sinensis mucilage, create a protective layer on the surface of fruits and vegetables, controlling the exchange of gases and metabolic processes to extend freshness [11,12]. Bio preservation involves the use of microorganisms or plants to enhance shelf life, with lactic acid bacteria bacteriocin and plant-derived peptides showing antimicrobial effects [13,24]. Biotechnological approaches can improve the nutritional quality and shelf life of fruits and vegetables, addressing issues like ripening dynamics and perishability [15]. These approaches have the potential to enhance quality and shelf life, but require engagement from various stakeholders to address safety, commercialization, and environmental considerations. Modified atmosphere packaging (MAP) technology, combined with edible film coating, is another method to improve shelf life by minimizing permeability and preventing microbial degradation. Overall, a combination of these methods can help improve the shelf life of vegetables.

Overcoming the aforementioned technologies, in this research, we invented a 2-6 μm mid-infrared generating atomizer (MIRGA) to simultaneously improve the sensory qualities as well as the shelf life of vegetables in an easy and economical way. Mid-infrared (MIR) refers to the wavelength

range in the electromagnetic spectrum that lies between the near-infrared and far-infrared regions. The electromagnetic radiation from the sun, which includes mid-infrared wavelengths, plays a significant role in the development and evolution of living organisms [16]. This region is vital and interesting for many applications since that region coincides with the internal vibration of most molecules [17]. Thus the 2-6 μm mid-infrared has been studied for its action and effect on the vegetables and the results are presented.

2. MATERIALS AND METHODS

2.1 MIRGA Equipment

MIRGA (patent no.: 401387) is a 20 ml pocket sized atomizer (Pic 1) (*Supplementary file – Fig. F1*) containing inorganic water based solution in which approximately two sextillion cations and three sextillion anions are contained. During spraying, depending on pressure (vary with the user) applied to plunger, every spraying generates 2-6 μm mid-IR. Design of the MIRGA and emission of 2-6 μm mid-IR has been presented in detail by Umakanthan et al. [18], Umakanthan et al. [19], Umakanthan et al. [20], Umakanthan et al. [21]. Every time spraying emits 0.06ml which contains approximately seven quintillion cations and eleven quintillion anions. (*details about MIRGA available in supplementary text T1*)

The inorganic compounds used in the generation of MIR are a perspective for biomedical applications [22,23]. It is also a new synthesis method for preparation of functional material (2-6

μm mid-IR) [24,25]. 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 [26,27].

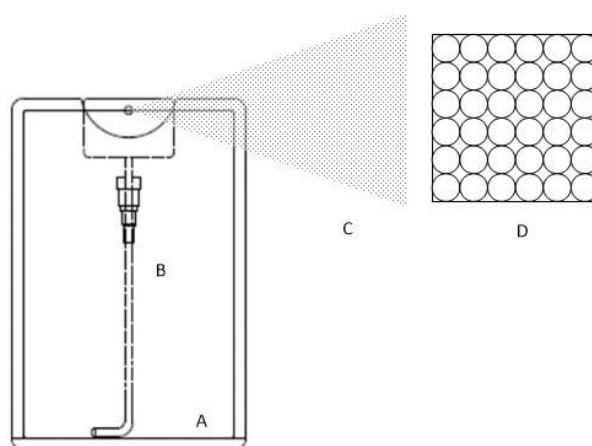
2.2 MIRGA Experimental Set-up

The distance between the spraying orifice of the MIRGA and the packaged vegetable sample is at a range of 0.25 m to 0.50 m. This distance is essential for the MIRGA sprayed chemical solution to form ion clouds and oscillations, and to generate 2-6 μm mid-IR. The MIRGA spray can penetrate the intervening packaged material to act on the vegetables inside. Close spraying does not generate 2-6 μm mid-IR energy. (*Method of MIRGA spraying in Supplementary file – video V1*)

2.3 Sample Preparation

Fresh vegetable samples and powder vegetable samples were obtained from a local market. The fresh vegetable samples were carrot (*Dacus carota*), beetroot (*Beta vulgaris*) and bitter melon (*Momordica charantia*). In addition, powdered vegetable samples of carrot, beetroot, and bitter melon were tested. The fresh vegetable samples and powder vegetable samples were treated with MIRGA spray treatments in two separate trials (trial I and trial II), which were repeated 6 independent times.

Microbial analysis done for the presence of coliforms and *Salmonella* sp. (method 2000.15 and method 967.6).



Pic. 1. Schematic of MIRGA equipment. The components of the equipment are (A) container; (B) plunger; (C) mid-IR spray which is used to treat the vegetable sample; (D) vegetables packet that contains the testing sample

2.4 Sensory Quality, Aroma Evaluation and Shelf-life Study

The sensory evaluation of the fresh vegetable samples was scored by an expert sensory panel (no.: 6). The sensory panel used an acceptability index based on 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 [28,29]. The shelf-life study consisted of evaluating the acceptability index of the fresh vegetable samples and the powder vegetable samples every day. After the shelf-study was finished, the sensory evaluation results were used to calculate the percentage of self-life increase.

2.5 Trial I: Treatment of Fresh Vegetable Samples with MIRGA Spraying

Two hundred and fifty grams (250 g) of each fresh vegetable sample was weighed and packed in 50 individual polyethene bags of different thicknesses (upto 70 μm). Air was removed manually by the operator, then the samples were sealed with cellophane. There were 400 packets in total (8 vegetables * 50 bags) to be tested. The control was Non-MIRGA sprayed packets for each vegetable, which accounted for 10 individual polyethene bags out of the 50 (10 x 8 = 80).

20 individual polyethene bags for each fresh vegetable received one MIRGA spray treatment, and another 20 bags received two MIRGA spray treatments (8 x 40 = 320). Each fresh vegetable sample from the control (5 samples), one MIRGA spray treatment (10 samples), and two MIRGA spray treatment (10 samples) groups were stored at room temperature (approximately 32°C), and at refrigeration temperatures (4°C). The fresh vegetable samples were daily subjected to sensory scoring.

2.6 Trial II: Treatment of Vegetable Powder Samples with MIRGA Spraying

Powder vegetable samples were packed in polyethene (>51 μm thickness). The control was Non- MIRGA sprayed packets for each powder vegetable (carrot, beetroot, and bitter gourd). Powder vegetable samples were sprayed with MIRGA. After every spraying, powder taken out, sensory evaluation done and samples taken for analysis. The spraying was until the powders' natural characteristics were nearly or completely lost (i.e. unpalatable). More spraying was

performed in comparison to the fresh vegetable samples because extra energy is needed to denature the natural characteristics. The number of spray treatments varied depending on the powder vegetable sample used.

Preliminary analysis demonstrated the number of MIRGA spray treatments that resulted in an increase in palatability, and unpalatability. In the case of powder carrot samples, two MIRGA spray treatments made it more palatable, and seven MIRGA spray treatments made it unpalatable. For beetroot, two MIRGA spray treatments resulted in a more palatable sample, and eight MIRGA spray treatments resulted in unpalatable samples. Finally, bitter gourd became more palatable after four MIRGA spray treatments and unpalatable after twelve spray treatments. So the control, more palatable and unpalatable samples were subjected to various instrumentations.

2.7 Effect of MIRGA spraying on Physicochemical Properties of Vegetable Samples

Analytical studies were conducted to evaluate the effect of MIRGA on the physicochemical properties of the vegetables. Gas chromatography mass spectroscopy (GC-MS) was employed to verify the transformation of chemical compounds. Changes in the chemical bonds, structures, and configuration were analysed via Fourier-transform infrared spectroscopy (FTIR), transmission electron microscopy (TEM), and powder X-ray diffraction (PXRD), respectively. Finally, changes in proton resonances were studied by proton nuclear magnetic resonance (^1H - NMR) assays [30,31,32,33].

GCMS: Instrument: Agilent 7890A GC with 5975C MS system. Column: HP-5. Ionization: EI (70 eV). Method: General_1_HP5_80_DEG.M. MSD: Single Quad.

FT-IR: A small quantity of the sample is added to KBr in the ratio 1:100 approximately. The matrix is grind for 3-4 minutes using mortar and pestle. The fine powder is transferred into 13 mm diameter die and made into a pellet using a hydraulic press by applying a pressure of 7 tonnes. The fine pellet is subjected to FTIR analysis using universal pellet holder. (a single drop of oil is poured on the KBr pellet in case of liquid samples). Infrared spectral data were collected on Thermo Avtar 370 FTIR spectrometer Spectra are collected over a range

of 4000– 400 cm^{-1} at 4 cm^{-1} resolution with an interferogram of 32 scans.

PXRD: The sample is smeared over low back ground sample holder (amorphous silica holder) and fixed on the sample stage in goniometer. The instrument is set with B-B geometry. The current and voltage is set to 40 mV and 35 mA and data has been collected. Instrument make: Bruker Model D8 Advance Goniometer: theta/2 theta.

TEM: An extremely small amount of material is suspended in water/ethanol (just enough to obtain slightly turbid solution). The solution is homogenised using ultrasonicator to disperse the particles, a drop of the solution is then pipetted out and cast the drop on carbon-coated grids of 200 mesh the grid is dried and fixed in the specimen holder. Instrument Make: Jeol Model JM 2100.

¹H-NMR: The experiments were done on a 600 MHz NMR spectrometer (ECZR Series, JEOL, JAPAN) using a 3.2mm CPMAS probe at 150MHz frequency. All the samples were run at 18 KHz spinning speed at Room Temp and with a delay of 5sec.

3. RESULTS AND DISCUSSION

3.1 Microbial Analysis Revealed that the Vegetables are Free of Pathogens

Trial I: Treatment of fresh vegetable samples with MIRGA spraying

The sensory quality, aroma, and shelf-life were enhanced in the fresh vegetable samples in comparison to control samples. Therefore, MIRGA spray treatment resulted in more palatable fresh vegetables, irrespective of stored condition (Table 1).

Table 2 shows that, fresh vegetables acquired an enhanced shelf-life of 16-900% at room temperature and 9-314% in refrigeration.

Trial II: Treatment of vegetable powder samples with MIRGA spraying

The sensory panel experts concluded that powder vegetable samples treated by MIRGA spraying twice (carrot and beetroot) and four times (bitter gourd) observed an increase in their aroma and sensory quality. Samples that received seven (carrot), eight (beetroot), and twelve (bitter gourd) treatments with MIRGA spraying, resulted in a decrease in their aroma and sensory quality to unpalatable levels. These

sensory attribute changes were perceived in 1-2 minutes after spraying. (Table 3).

It is observed that two sprayings derived favorable results in carrot and beetroot, whereas four MIRGA sprayings are found effective in bittergourd to achieve the desirable qualities. Excessive number of spraying beyond this are found to have negatively influenced the results leading to unfavorable sensory characteristics in the vegetables.

3.2 Instrumentation Results with Vegetable Powders (Raw Data of Instrumentations in Supplementary File – Data D1)

3.2.1 Transformation of Chemical Compounds via GC-MS Analysis

(a) Carrot Powder: Control sample contains Z-11-Pentadecanol, 3-Hydroxy Dodecanoic acid as major peaks. In 2 sprayed sample, there was new peak of 13-Heptadecyn-1-ol and 3,3,5 Trimethyl Cyclohexanone but there was no peak of Z-11-Pentadecanol. Additionally, there was new peak of n-Hexadecanoic acid, Octadecanedioic acid and Bicyclohexyl, 4 - phenyl. These differences are responsible for enhancement in sweetness and aroma. On other hand, 7 sprayed sample has shown unique peak E-z-Octadecadecenoic-1-ol and it was a major peak. There was no peak of 3-Hydroxy Dodecanoic acid but decrease in Z-11-Pentadecanol as compared to control sample. There was newer peak of n-Hexadecanoic acid and 6-Dimethyl (chloromethyl)silyloxy Tetradecane. Overall, these attributed to the reduction of sweetness and taste in 7 sprayed sample.

(b) Beetroot Powder: Control sample contains Phytosterols, such as Stigmasterol, Stigmasterol, Estra-1, 3,5 (10)- trien-17 β -ol, and other molecules such as 13-Docosenamide, (Z), Oxiraneoctanoic acid, 3- octyl-, cis, etc. The phytosterol peaks are unique peaks for Beetroot samples, which are disappearing after spraying. In 2 sprayed sample, there was the unique peak of dl- Glyceraldehyde, 1-Nitro-2-propanol, 7-Methyl-Z-tetradecen-1-ol acetate, which was not present in the control. Further, there was decrease in peak of Estra-1,3,5(10)-trien-17 β -ol in 2 sprayed sample. 8 sprayed sample has shown a unique peak of Monoethanolamine, n-Hexadecanoic acid, Deoxyspergualin, etc., which is responsible for tastelessness, reduction in sweetness and aroma, took time to solubilize. In

addition, there was great increase in peak of 2-Amino-1,3-propanediol as compared to control sample. 9-Octadecenamide and 1,7-Dioxo-10-thia-4,13-diazacyclopentadeca-5,9,12-trione are two peaks which have appeared after spraying.

(c) Bitter Gourd Powder: The control bitter gourd powder contained Methyl 10-Methylundecanoate, Hexadecanoic acid methyl ester, Pentadecanoic acid, 14-Methyl-,methyl ester, n-Hexadecanoic acid, 11-Octadecenoic acid methyl ester, 17-Octadecynoic acid, Octadecanoic acid methyl ester, Pentadecanoic acid, and Octadecanoic acid compounds. After four MIRGA spray treatments, there were new peaks that were not present in control, including Methyl 10-Methylundecanoate and 17-Octadecynoic acid. However, there were unique peaks of Hexadecanoic acid, methyl ester in samples that received 4 MIRGA spray treatments, and Pentadecanoic acid in samples that received 12 MIRGA spray treatments, compared to control. Interestingly, there was a disappearance of 11-Octadecenoic acid methyl ester from the GC-MS spectra of bitter gourd powder due to MIRGA spray treated samples. It was observed that MIRGA spray treatments also caused a gradual increase in the peak of n-Hexadecanoic acid and a gradual decrease in the peak of Octadecanoic acid.

3.2.2 Chemical bonds analysis via FTIR

(a) Carrot Powder: The FTIR spectra of control carrot powder displayed a big peak was observed at 3396cm^{-1} , which was specific to carbohydrate and OH-groups from water. Two more peaks were observed: a peak at 2925cm^{-1} , specific to C-H bonds from organic compounds, and a peak at 1632cm^{-1} , which resembled N-H bonds (near C=O) from proteins. In carrot powder samples that were MIRGA spray treated 2 times and 7 times, a small increase in the absorbance in all areas was observed compared to in control. The increased absorbance was probably due to increasing the concentration of a substance (carbohydrates, OH-groups, organic compounds, or proteins) in the compaction matrix. In samples that were MIRGA spray treated 2 times, the absorbance was lower at 1632cm^{-1} and 618cm^{-1} , which was probably caused by a lower protein concentration. The enhanced sweetness was associated with the breakdown of a portion of polysaccharides into monosaccharides [34].

(b) Beetroot Powder: A big peak was observed at 3392cm^{-1} , for the FTIR spectra of control beetroot powder, which was specific to

carbohydrate and OH-groups from water. Two more peaks were observed: a peak at 2929cm^{-1} , specific to C-H bonds from organic compounds, and a peak at 1631cm^{-1} , similar to N-H bonds (near C=O) from proteins. All peaks became smaller after 2 MIRGA spray treatments, compared to the control and 8 sprayed samples. 8 sprayed samples were almost identical to the control samples, except for peaks at 1631cm^{-1} and 618cm^{-1} . An explanation for this behaviour is due to protein reduction caused by the MIRGA spray treatments.

(c) Bitter Gourd Powder: The control bitter gourd powder had a big peak of OH-groups at 3406cm^{-1} . This peak was specific to carbohydrates and OH-groups from water. Absorbance at this wavelength increased when samples received 4 MIRGA spray treatments and decreased when samples received eight MIRGA spray treatments. It could be proposed that absorbance increased after four treatments because MIRGA spraying caused an increase in the concentration of required substances (carbohydrates and OH-groups), and the reduced absorbance after 8 treatments resulted from water evaporation. An increase in the peaks at 2925cm^{-1} , from C-H bonds of organic compounds, and at 1741cm^{-1} , possibly C=O bonds from carbohydrates, were observed for samples that received 4 MIRGA spray treatments. The increase in the peaks at 2925cm^{-1} and 1741cm^{-1} were more pronounced in the samples that received eight MIRGA spray treatments. Therefore, the concentration of carbohydrates seemed to increase with a higher number of MIRGA spray treatments. The opposite trend was observed for proteins. A peak at 1631cm^{-1} , similar to N-H bonds (near C=O) from proteins, decreased slightly after 8 MIRGA spray treatments, and it was highly reduced after 4 MIRGA spray treatments.

3.2.3 Particle structure analysis via TEM

(a) Carrot Powder: As shown in Fig. 3b, carrot powder samples which received 2 MIRGA spray treatments showed minor changes of morphology and matrix structure, and negligible or no changes of the atomic arrangement, compared to control samples. In contrast, carrot samples that received 7 MIRGA spray treatments were visibly affected in terms of morphology and matrix structure, compared to control samples. Therefore, as the number of MIRGA spray treatments increased, more noticeable changes may occur in the morphology and matrix of carrot powder

Table 1. Sensory profiling of fresh vegetables

No. of MIRGA sprayings	Fresh vegetables															
	Lady's finger		pumpkin		Beans		Carrot		Brinjal		Bottle gourd		Beetroot		Bitter gourd	
	Taste	Aroma	Taste	Aroma	Taste	Aroma	Taste	Aroma	Taste	Aroma	Taste	Aroma	Taste	Aroma	Taste	Aroma
Control	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5
1	7	6	7	5	7	5	8	6	7	6	6	5	7	6	6	7
2	8	6	8	6	8	6	8	6	8	6	8	6	7	7	6	6

Table 2. Shelf-life study of fresh vegetable samples based on the comparison of the number of days that the samples remained unspoiled at room temperature (app. 32 °C) and refrigeration temperature (4 °C) (standard deviation ± one day)

Number of MIRGA spray treatments	Fresh vegetable sample	Number of days unspoiled at room temperature (app. 32 °C)	Shelf-life increase at room temperature (%)	Number of days unspoiled at refrigerated temperature (4°C)	Shelf-life increase at refrigeration temperature (%)
0 (control)	Lady's finger	6	-	15	-
0 (control)	Pumpkin (sliced)	2	-	10	-
0 (control)	Carrot	3	-	10	-
0 (control)	Beans	5	-	10	-
0 (control)	Beetroot	5	-	14	-
0 (control)	Brinjal	4	-	7	-
0 (control)	Bottle guard	3	-	10	-
0 (control)	Bitter guard	3	-	7	-
1	Lady's finger	24	16	42	180
1	Pumpkin (sliced)	12	71	20	100
1	Carrot	30	900	38	280
1	Beans	15	200	22	120
1	Beetroot	7	40	65	9
1	Brinjal	15	275	29	314
1	Bottle guard	25	733	37	270
1	Bitter guard	13	333	17	142
2	Lady's finger	13	116	28	86
2	Pumpkin	9	350	16	300
2	Carrot	21	600	30	200
2	Beans	19	280	21	110
2	Beetroot	9	80	24	71
2	Brinjal	8	100	11	57
2	Bottle guard	19	533	32	220
2	Bitter guard	10	233	16	128

Table 3. Sensory profiling of vegetable powders

No. of MIRGA sprayings	Vegetable powders					
	Carrot powder		Beetroot powder		Bitter gourd powder	
	Taste	aroma	Taste	aroma	Taste	Aroma
Control	5	5	5	5	5	5
1	7	6	6	5	5	5
2	8	7	8	7	5	6
3	8	6	7	7	7	7
4	5	5	6	5	9	8
5	3	5	5	5	7	7
6	3	3	5	5	7	6
7	2	3	3	4	5	5
8			2	4	5	4
9					4	3
10					2	3
11					1	3
12					1	1

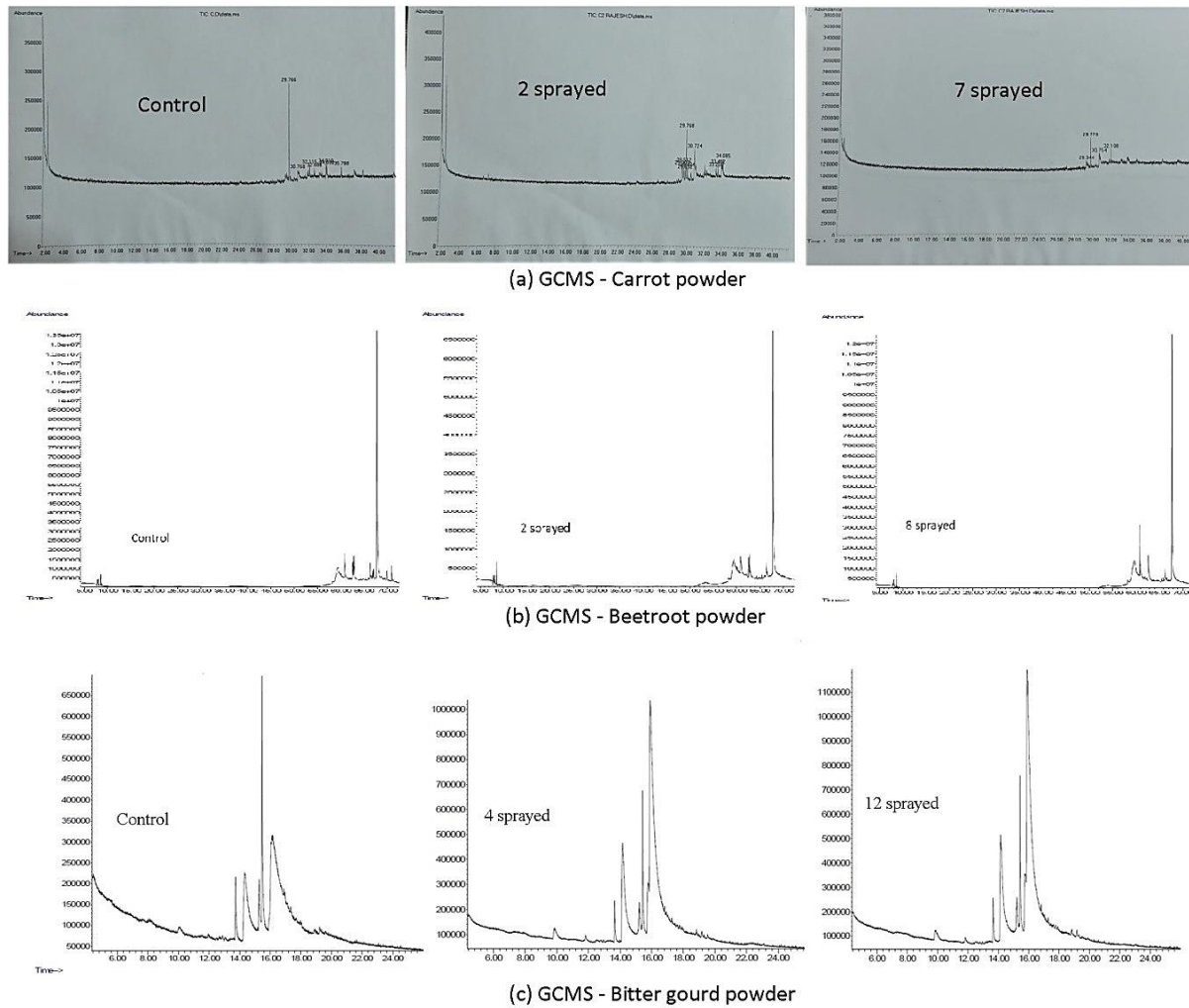
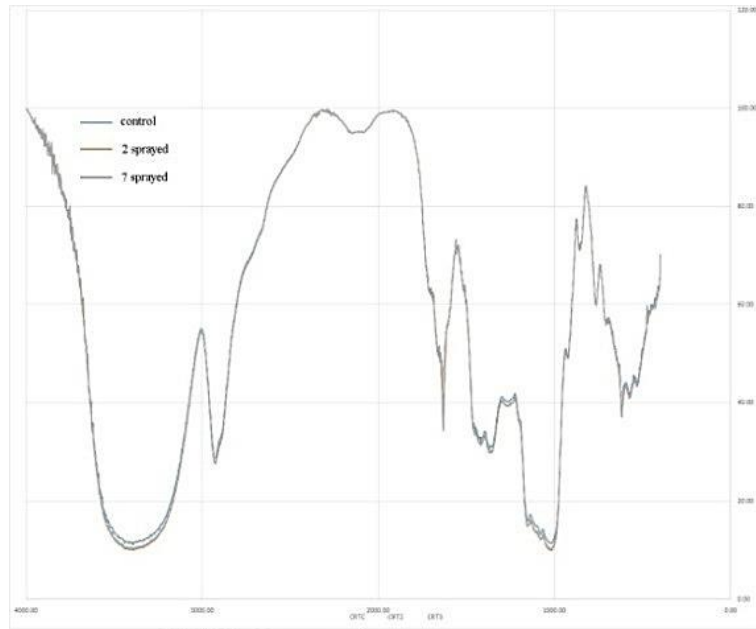
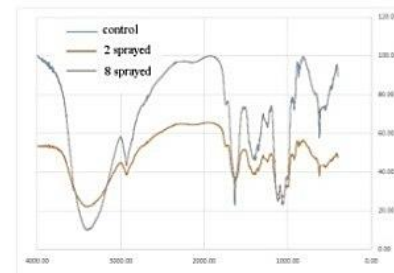


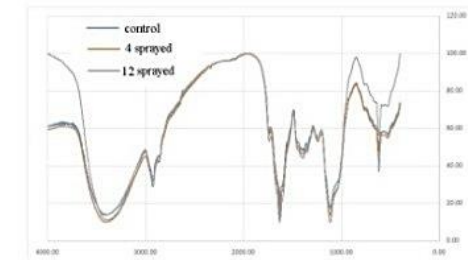
Fig. 1. GC-MS chromatograph of powder(a) beetroot powder, and (b) bitter gourd treated with MIRGA spraying



(a) FTIR - Carrot powder



(b) FTIR - Beetroot powder



(c) FT-IR Bitter gourd powder

Fig. 2. FTIR spectra of (a) carrot powder, (b) beetroot powder, and (c) bitter gourd powder treated with MIRGA spraying

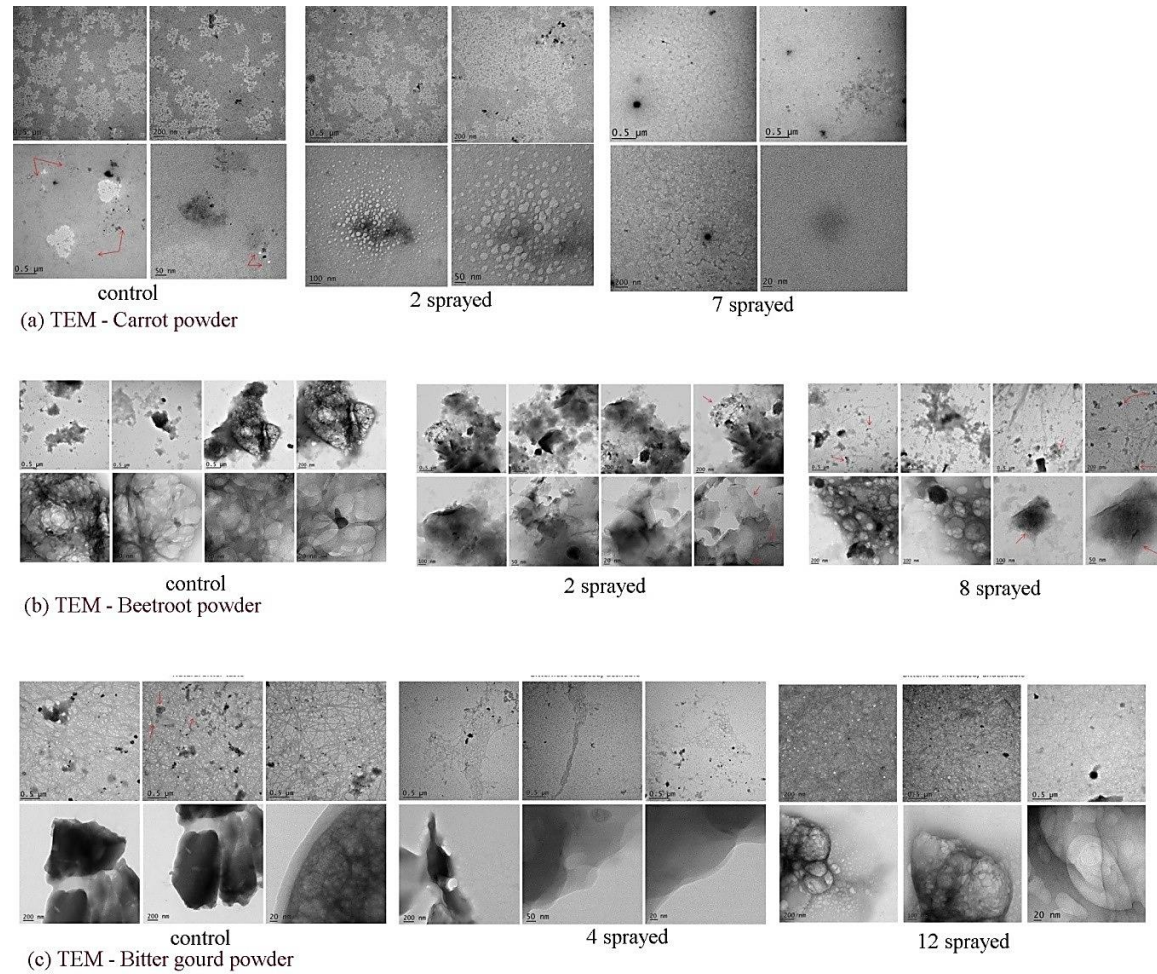
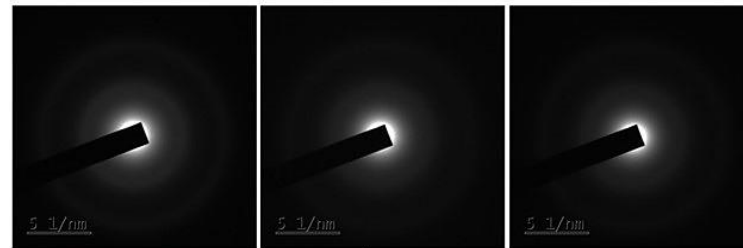
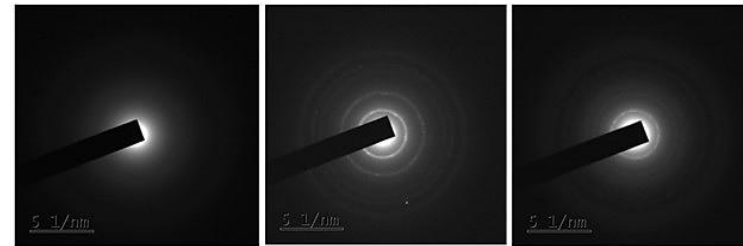


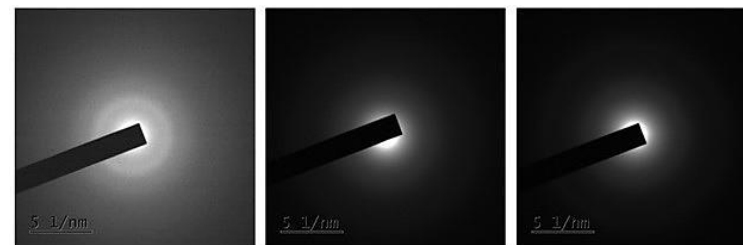
Fig.3a.TEM - Bright field images of (a) carrot powder, (b) beetroot powder, and (c) bitter melon powder treated with MIRGA spraying.



(a) Carrot powder



(b) Beetroot powder



(c) Bitter gourd powder

Fig.3b.TEM –Electron diffraction patterns of (a) carrot powder, (b) beetroot powder, and (c) bitter gourd powder treated with MIRGA spraying

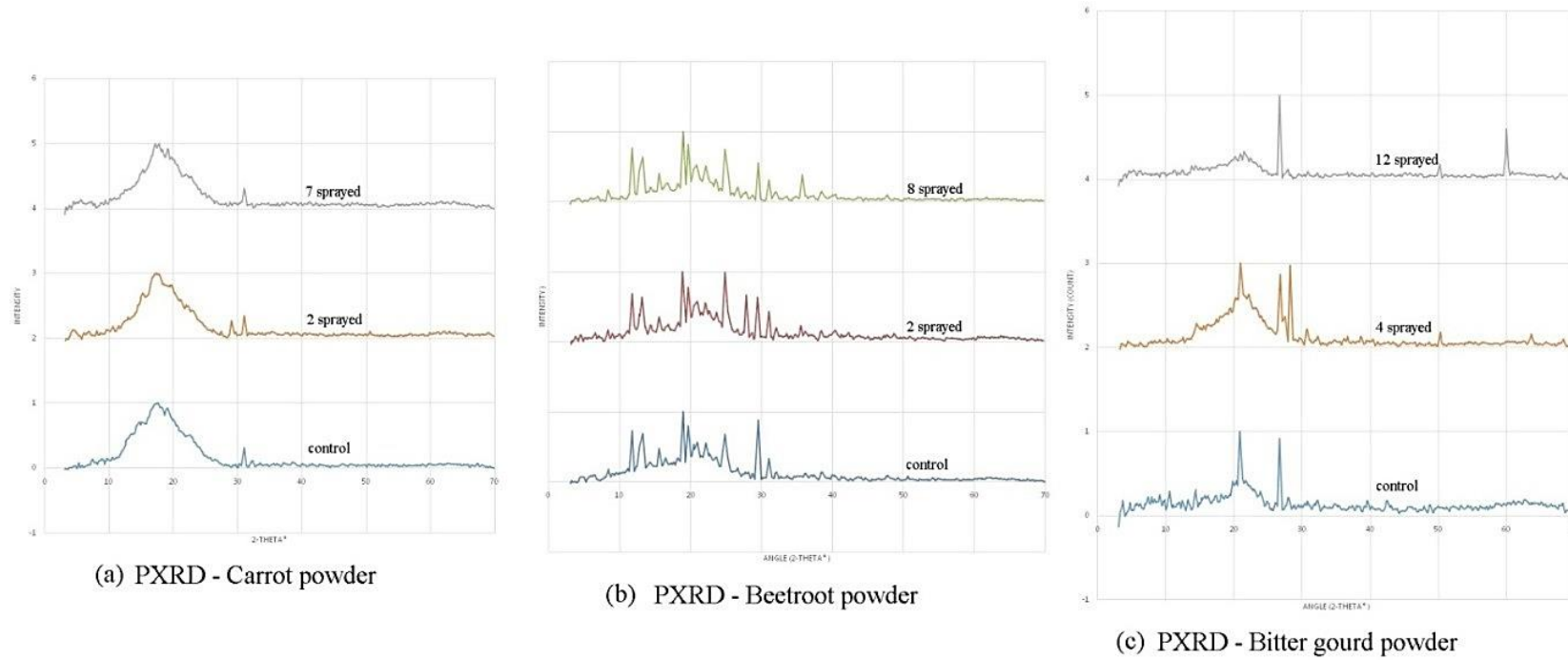


Fig. 4. PXRD spectra of (a) carrot powder, (b) beetroot powder, and (c) bitter melon powder treated with MIRGA spraying

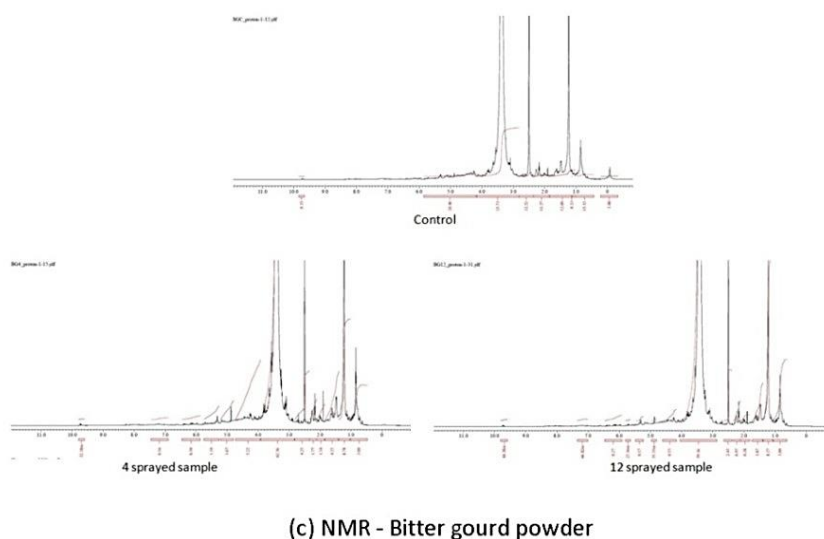
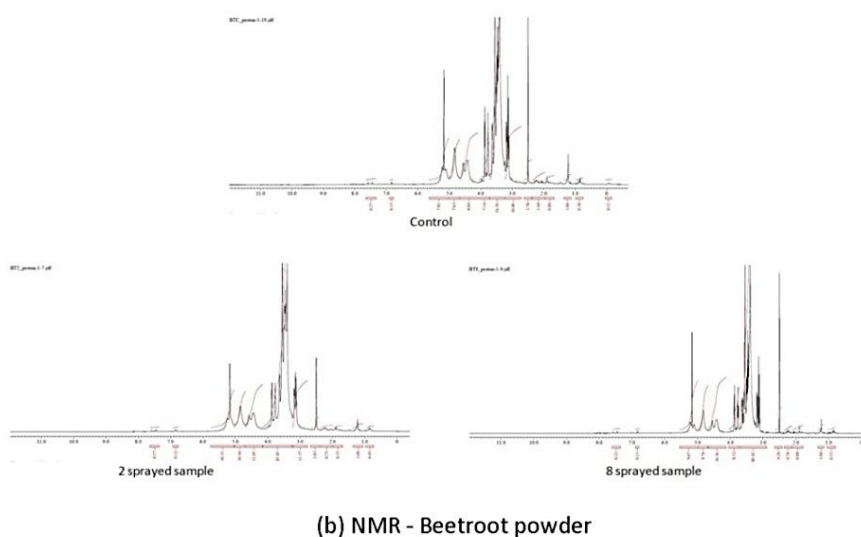
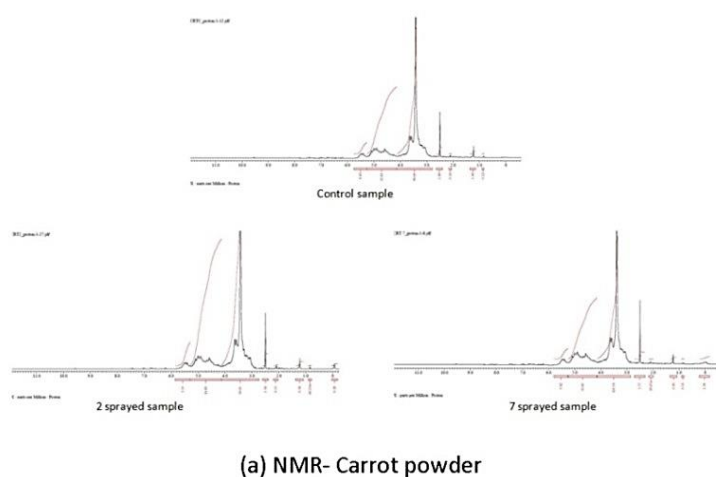


Fig 5. H-NMR spectra of (a) carrot powder, (b) beetroot powder, and (c) bitter gourd powder treated with MIRGA spraying

(b) Beetroot Powder: MIRGA spraying had different effects on the size and shape of matrix components of beetroot powder, therefore it changed the overall sample structure. In Fig. 3a, the main matrix components of the beetroot control samples can be identified as amorphous-shaped fragments, but in the samples that received 2 MIRGA spray treatments the fragments were clustered in large aggregates, and in the samples that received 7 MIRGA spray treatments the size of the fragment was noticeably smaller.

(c) Bitter Gourd Powder: Alterations in the structure, matrix components in numerosity, shape, and morphology were observed on bitter gourd powder samples that received MIRGA spray treatments compared to control. However, the atomic arrangement was only slightly affected as seen in Fig. 3a. Peculiarly, 4 MIRGA spray treatment caused more evident changes than 12 MIRGA spray treatments.

3.2.4 Chemical configuration analysis via PXRD

(a) Carrot Powder: As seen in Fig. 4, all three carrot powder samples showed a large volume (%) of amorphous phases reflected by one broad peak centred on 17°. A general trend was observed where the broad peak shifted to the right with an increased number of MIRGA spray treatments. The control samples and the samples that received 2 MIRGA sprayed sample showed broader and overlapping peaks (10° - 28°) compared to samples that were MIRGA spray treated 7 times. The prominent peak at 31° had a fixed location in all the control and sprayed samples, even as the number of MIRGA spraying increased. Samples that received 2 MIRGA spraying had one more peak at 29.07°, which suggested an increase of the crystalline phase. Most peaks below 31° in all the samples agree with the values presented by Rocha et al. [35].

(b) Beetroot Powder: The spectra of beetroot powder showed good signal-to-noise ratios, allowing the identification of several peaks. Control samples had the least intense peak reflection. Samples that were MIRGA spray treated 2 times presented more peaks in their spectra than samples that were MIRGA spray treated 8 times, although the latter showed a relatively intense peak at 35.59°. The peak around 24.80° is highest for samples that received 2 MIRGA spray treatments, followed by samples that received 8 MIRGA spray

treatments, and control samples had the smallest peak. In the case of beetroot powder that received 2 MIRGA spray treatments, new peaks at 27.81° and 35.59° were displayed compared to control samples. Samples that received 8 MIRGA spray treatments also showed new peaks at 35.69°, but with higher intensity than for samples that received 2 treatments

(c) Bitter Gourd Powder: Consistent with the literature [36,37], all bitter gourd powder samples had characteristic peaks at around 14°, 21°, and 26° for their pattern of XRD (Fig. 4). Control samples had the least number of prominent peaks among the three samples (control, and samples that received 4 or 12 MIRGA spray treatments). Samples that received 4 MIRGA spray treatments had the highest number of intense, narrow peaks, while the samples that received 12 MIRGA spray treatments showed the best signal-to-noise ratio. A considerable change in the presence of new peaks was observed in samples that received 4 MIRGA spray treatments, particularly at 28.25°, indicating the formation of a new crystalline phase in the samples. More drastic changes in the structure were observed in samples that received 12 MIRGA spray treatments, where the intensity of the peak in 21.49° was reduced, and the peak at 26.66° became the most prominent, compared to control samples and samples that received 4 MIRGA spray treatments. Samples that received 12 MIRGA spray treatments had the largest volume (%) crystalline phase among the three samples (control, and samples that received 4 or 12 MIRGA spray treatments). Control samples and samples that received 4 MIRGA spray treatments had a similar structure. The peak centred around $2\theta = 26^\circ$ in each of the three samples (control, and samples that received 4 or 12 MIRGA spray treatments) changed in terms of breadth and intensity, indicating a change in crystallinity as the number of MIRGA spray treatments increased.

3.2.5 Nuclear resonance analysis via ¹H-NMR

(a) Carrot Powder: Sweetness and aroma of carrot powder depended on sugars and terpenoid compounds. The control samples showed a profile compatible with a sugar-rich composition and other minor components. The interpretation of ¹H-NMR spectra was challenging due to signal overlap. However, it is possible to infer from Fig. 5 that the MIRGA spray treatment affected the concentration, or the

integrity of molecules involved in the sweetness and aroma of carrot powder.

(b) Beetroot Powder: An increase of sweetness, aroma, and solubility was observed in samples that received 2 MIRGA spray treatments. Sweetness, aroma, and solubility were directly related to the increase in the concentration of sucrose, geosmin, and soluble compounds, respectively. The integral corresponding to the region of the signals coming from sucrose was higher than that from the control samples. Since geosmin is a minor component, its variation was hidden by the overlap of its signal with more intense signals. In the case of samples that received 8 MIRGA spray treatments, changes followed the opposite trend: a drastic reduction in the concentration of sucrose, geosmin, and soluble compounds was observed. Therefore, sweetness, aroma, and solubility were reduced.

(c) Bitter Gourd Powder: Bitter gourd powder samples that received 4 MIRGA spray treatments showed a reduction in the concentration of terpenoids and polyphenols, which could be attributed to a reduced bitterness. Terpenoids and polyphenols are involved in the bitter taste of bitter gourd powder, but they are composed of a breath of relatively complex molecular structures with different chemical groups. For this reason, it was difficult to interpret which variations in the NMR spectra correspond to changes in the concentration of molecules involved in the perception of bitterness.

In the case of samples that received 12 MIRGA spray treatments, an increase in bitter compounds was observed in the spectra. Paradoxically, all the integrals showed a reduction in bitter compounds for samples that received 12 MIRGA spray treatments, compared to the control samples. A possible explanation for this result would be that there were minor components in bitterness, and the augmentation effect that they have on the perception of bitterness was hidden by the overlap with signals coming from more concentrated components.

3.3 Action of Mid-IR on Vegetables

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 by Umakanthan et al. [18]

and Umakanthan et al. [21] (*detailed discussion on MIRGA available in supplementary text T2*).

Commonly vegetables shelf life is improved by treating vegetables with microorganism derived preservative, followed by preservative gas package [38], nanotechnology related strategies such as nano-zincdioxide and silver nanoparticles [39], edible coating with pulsed light treatment [40] and evaporation cooling system [41]. Infrared was found to influence the vegetables shelf life [42]. The sensory quality of vegetables is improved through seasonings [43]. Not in this study simultaneously enhanced the shelf life and sensory qualities by exposing vegetables to 2-6 μm mid-infrared.

At present, chemical preservatives and ultraviolet, microwave, pulsed electric fields, gamma and infrared radiations are used for food processing. Except infrared, other radiations are ionizing, hence lethal, uneconomical and non-friendly to user and ecology [44,45,46,47]. Thus the use of infrared in food processing is a future option [47]. Though infrared radiation use has many advantages than the conventional method but not without challenges and research [48]. It is concluded that MIRGA overcome the disadvantages of present food processing technologies and expected to convince the industry and consumer.

Similar desirable results in coffee, tea, cocoa, edible salts and terminalia were achieved using MIRGA spraying by Umakanthan et al. [18] Umakanthan et al. [19], Umakanthan et al., [49], Umakanthan et al. [21].

4. CONCLUSION

Through sensory trials and various instrumentations, we demonstrated that MIRGA spraying (from 0.25-0.50 meter) treatments altered the chemistry of polythene packaged vegetable samples and made them more desirable in terms of aroma, taste and shelf life enhancement. The impact of this study involves the development of a processing technology that could improve the aroma, sensory quality, and shelf-life of vegetables without genetic manipulation. In our experience with MIRGA spraying technology, it is possible to further enhance the taste, aroma, and shelf-life by altering the MIRGA formulation. As this study found no shortcomings in achieving the study objective, further studies are also ongoing to

reveal the use of MIRGA in other areas of food sciences.

DATA AND MATERIALS AVAILABILITY

All data is available in the manuscript and supplementary materials.

Supplementary file:

<https://docs.google.com/document/d/1Le3SwWx7o7j0fiUdvgI3pnMttYGCljcc/edit?usp=sharing&oid=111101387151809704391&rtpof=true&sd=true>

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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