SensoryQuality andShelf-lifeEnhancementofVegetablesIrradiatedwithMid-infrared

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 generating 2-6 µmmid-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 µmmid-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 **(Jody et al., 2021)**. They also contribute to the sensory appeal of a diet through their taste and flavors **(Sayma et al., 2023)**. Important taste compounds in vegetables include organic acids, free sugars, and amino acids, which give vegetables their sour, sweet, and umami tastes **(Silva, 2011)**. The consumption of fruits and vegetables is recommended by organizations like FAO/WHO to prevent chronic noncommunicable diseases and mitigate micronutrient deficiencies **(Hideki, 2009)**. However, despite their importance, the intake of fruits and vegetables remains low for a majority of the global population **(Kanika et al., 2015)**. 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 **(Chen et al., 2019)**. Another approach is to use vegetable powder, which can be added to food products to improve their taste, flavor, and texture **(Plijter, 2020)**. Additionally, a body taste improver, such as a long-chain highly unsaturated fatty acid, can be used in vegetable fats to enhance their taste **(Susumu et al., 2003)**. 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**(Ando et al., 2013)**. 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 **(Singh et al., 2019)**.

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 **(Chithra et al., 2022; Jashanveer et al., 2021)**. 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 **(Khalid et al., 2018; Vignesh et al., 2019)**. Biotechnological approaches can improve the nutritional quality and shelf life of fruits and vegetables, addressing issues like ripening dynamics and perishability **(Merve et al., 2017)**. 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.

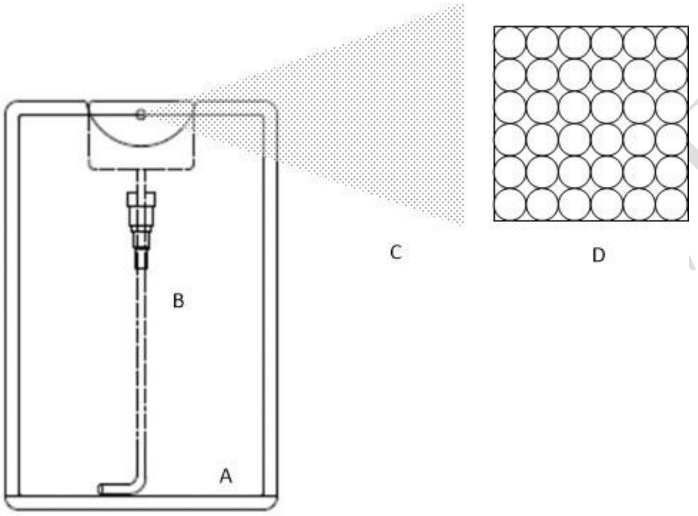
Overcoming the aforementioned technologies, in this research, we invented a 2-6 µm mid-infrared generating atomizer (MIRGA) to simultaneously improve the sensory qualities as well as the shelf life of vegetables in an easy and aconomical way. Mid-infrared (MIR) refers to the wavelength range in the electromagnetic 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 **(Charles, 2022)**. This region is vital and interesting for many applications since that region coincides with the internal vibration of most molecules **(CORDIS, 2022)**. Thus the 2-6 µm mid-inrfared has been studied for its action and effect on the vegetables and the results are presented.

2. MaterialsandMethods

2.1.MIRGAEquipment

MIRGA *(patent no.: 401387)* is a 20 ml pocket sized atomizer *(Pic 1) (Supplementary file – figure 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 2-6µm mid-IR. Design of the MIRGA and emission of 2-6µm mid-IRhas been presented in detail by **Umakanthan *et al.*, 2022a; Umakanthan *et al.*, 2022b; Umakanthan *et al.*, 2023c; Umakanthan *et al.*, 2023d.** 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 **(Tishkevich *et al.*, 2019; Dukenbayev*et al.*, 2019)**. It is also a new synthesis method for preparation of functional material (2-6 µm mid-IR) **(Kozlovskiy*et al.*, 2021; El-Shater *et al.*, 2022)**. 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**(Kozlovskiy and Zdorovets, *2021;* Almessiere*et al.*, 2022)**.



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.

2.2.MIRGAExperimentalSet-up

ThedistancebetweenthesprayingorificeoftheMIRGAandthepackagedvegetablesampleisat a range of 0.25 m to 0.50 m. This distance is essential for the MIRGA sprayed chemicalsolution 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 thevegetablesinside.Closesprayingdoesnotgenerate2-6µmmid-IRenergy.*(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. Thefresh vegetable samples were carrot (*Dacuscarrota),* beetroot(*Betavulgaris*) and bittergourd(*Momordicacharantia*). In addition, powdered vegetable samples of carrot, beetroot, and bitter gourd weretested. The fresh vegetable samples and powder vegetable samples were treated with MIRGAspray treatments in two separate trials (trial I and trial II), which were repeated 6 independenttimes.

Microbial analysis done for the presence of coliforms and *Salmonella* sp. (method 2000.15 andmethod967.6).

2.4.Sensory Quality, AromaEvaluationandShelf-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-pointnominalstructure1-Dislikeextremely,2-Dislikeverymuch,3-Dislikemoderately,4- Dislike slightly, 5 - Neither like nor dislike, 6 - Like slightly, 7 - Like moderately, 8 - Like verymuch, 9 - Like extremely **(Everitt, 2009; Wichchukit *et al*., 2014)**. The shelf-life study consisted of evaluating the acceptabilityindex of the fresh vegetable samples and the powder vegetable samples every day. After theshelf-study was finished, the sensory evaluation results were used to calculate the percentage ofself-lifeincrease.

2.5.Trial I:Treatmentoffreshvegetable sampleswith MIRGA spraying

Two hundred and fifty grams (250 g) of each fresh vegetable sample was weighed and packed in50 individual polyethene bags of different thicknesses (upto 70 µm). Air was removed manuallyby the operator, then the samples were sealed with cellophane. There were 400 packets in total (8vegetables * 50 bags) to be tested. The control was Non-MIRGA sprayed packets for eachvegetable, whichaccountedfor10individualpolyethene bagsoutofthe50(10x8= 80).

20 individual polyethene bags for each fresh vegetablereceived one MIRGA spray treatment,and another 20 bags received two MIRGA spray treatments (8 x 40 = 320). Each fresh vegetablesamplefromthecontrol(5samples),oneMIRGAspraytreatment(10samples),andtwoMIRGA spray treatment (10 samples) groups were stored at room temperature (approximately32°C), and at refrigeration temperatures (4°C). The fresh vegetable samples were daily subjectedtosensoryscoring.

2.6.Trial II:Treatmentofvegetable powdersampleswith MIRGAspraying

Powder vegetable samples were packed in polyethene (>51 µn thickness). The control was Non-MIRGA sprayed packets for each powder vegetable (carrot, beetroot, and bitter gourd). PowdervegetablesamplesweresprayedwithMIRGA.Aftereveryspraying,powdertakenout,sensoryevaluation done and samples taken for analysis. The spraying was until the powders' naturalcharacteristics were nearly or completely lost (i.e. unpalatable). More spraying was performed incomparison to the fresh vegetable samples because extra energy is needed to denature the naturalcharacteristics.Thenumber ofspray treatmentsvarieddepending onthepowdervegetablesampleused.

Preliminary analysis demonstrated the number of MIRGA spray treatments that resulted in anincrease in palatability, and unpalatability. In the case of powder carrot samples, two MIRGAspraytreatmentsmadeitmorepalatable,andsevenMIRGAspraytreatmentsmadeitunpalatable. For beetroot, two MIRGA spray treatments resulted in a more palatable sample, andeight MIRGA spray treatments resulted in unpalatable samples. Finally, bitter gourd becamemore palatable after four MIRGA spray treatments and unpalatable after twelve spray treatments.Sothecontrol,morepalatableandunpalatablesamplesweresubjectedtovariousinstrumentations.

2.7.EffectofMIRGAsprayingonPhysicochemicalPropertiesofVegetableSamples

Analytical studies were conducted to evaluate the effect of MIRGA on the physicochemicalproperties of the vegetables. Gas chromatography mass spectroscopy (GC-MS) was employed toverify the transformation of chemical compounds. Changesin the chemical bonds, structures,andconfigurationwereanalysedviaFourier-transforminfraredspectroscopy(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. **(Shanthamurthy et al., 2022; Bernd et al., 2022; Yulia et al., 2013; Koleleni et al., 2023)**

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

FT-IR: A small quantity of the sample is added to KBr in the ratio 1:100 approximately. Thematrix is grind for 3-4 minutes using mortar and pestle. The fine powder is transferred into 13mm diameter die and made into a pellet usinga hydraulic press by applying a pressure of 7tonnes. The fine pellet is subjected to FTIR analysis using universal pellet holder. (a single dropof oil is poured on the KBr pellet in case of liquid samples). Infrared spectral data were collectedon ThermoAvtar370FTIRspectrometerSpectraarecollectedoverarangeof4000–400cm^{-1}at 4cm^{-1}resolutionwithaninterferogramof32scans.

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. Thecurrent and voltage is set to 40 mV and 35 mAand data has been collected. Instrument make:BrukerModel D8AdvanceGoniometer: theta/2theta.

TEM: An extremely small amount of material is suspended in water/ethanol (just enough toobtain slightly turbid solution).The solution is homogenised using ultrasonicator to disperse theparticles, a drop of the solution is then pippeted out and cast the drop on carbon-coated grids of200 mesh the grid is dried and fixed in the specimen holder. Instrument Make: Jeol Model JM2100.

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 18KHzspinningspeedatRoomTempandwithadelayof5sec.

3. Results and Discussion

*3.1.Microbialanalysis*revealedthatthevegetablesarefreeofpathogens.

TrialI:TreatmentoffreshvegetablesampleswithMIRGAspraying

Thesensoryquality,aroma,andshelf-lifewereenhancedinthefreshvegetablesamplesincomparisontocontrolsamples.Therefore,MIRGAspraytreatmentresultedinmorepalatablefreshvegetables,irrespectiveofstoredcondition.*(Table 1)*

*Table2*showsthat,freshvegetablesacquiredanenhancedshelf-lifeof16-900%atroomtemperatureand9-314% inrefrigeration.

TrialII:TreatmentofvegetablepowdersampleswithMIRGAspraying

The sensory panel experts concluded that powder vegetable samples treated by MIRGA spraying twice (carrot and beetroot) and four times(bittergourd)observedanincreaseintheiraromaandsensoryquality.Samplesthatreceivedseven(carrot),eight(beetroot),andtwelve(bittergourd)treatmentswithMIRGAspraying,resultedinadecreaseintheiraromaandsensoryqualitytounpalatablelevels.Thesesensoryattributechangeswere perceivedin 1-2minutesafterspraying.*(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.Instrumentationresultswithvegetablepowders (raw data of instrumentations in Supplementary file – Data D1)

3.2.1. *TransformationofChemicalCompoundsviaGC-MSAnalysis*

(a) *Carrotpowder*

ControlsamplecontainsZ-11-Pentadecanol,3-HydroxyDodecanoicacidasmajorpeaks.In2sprayedsample,therewasnewpeakof13-Heptadecyn-1-oland3,3,5TrimethylCyclohexanonebuttherewasnopeakofZ-11-Pentadecanol.Additionally,therewasnewpeakofn-Hexadecanoicacid,OctadecanedioicacidandBicyclohexyl,4-phenyl.Thesedifferencesareresponsibleforenhancementinsweetnessandaroma.Onotherhand,7sprayed sample has shown unique peak E-z-Octadecadecenoi-1-ol and it was a major peak.Therewasnopeakof3-HydroxyDodecanoicacidbutdecreaseinZ-11-Pentadecanolascompared to control sample. There was newer peak of n-Hexadecanoic acid and 6-Diemthyl(chloromethyl)silyloxyTetradecane. Overall,theseattributed to thereduction ofsweetnessandtastein7sprayedsample.

(b) *Beetrootpowder*

Control sample contains Phytosterols, such as Stigmasterol, Stigmasterol, Estra-1, 3,5 (10)-trien-17β-ol, and other molecules such as 13-Docosenamide,(Z), Oxiraneoctanoicacid,3-octyl-,cis,etc.ThephytosterolpeaksareuniquepeaksforBeetrootsamples,whicharedisappearingafterspraying.In2sprayedsample,therewastheuniquepeakofdl-Glyceraldehyde,1-Nitro-2-propanol,7-Methyl-Z-tetradecen-1-olacetate,whichwasnotpresent in the control. Further, there was decrease in peak of Estra-1,3,5(10)-trien-17β-ol in 2sprayedsample.8sprayedsamplehasshownauniquepeakofMonoethanolamine,n-Hexadecanoic acid, Deoxyspergualin, etc., which is responsible for tastelessness, reduction insweetness and aroma, took time to solubilize. In addition, there was great increase in peak of2-Amino-1,3-propanediol as compared to control sample. 9-Octadecenamide and 1,7-Dioxa-10-thia-4,13-diazacyclopentadeca-5,9,12-trionearetwopeakswhichhaveappearedafterspraying.

(c) *Bittergourdpowder*

ThecontrolbittergourdpowdercontainedMethyl10-Methyl-undecanoate,Hexadecanoicacidmethylester,Pentadecanoicacid,14-Methyl-,methylester,n-Hexadecanoicacid,11-Octadecenoicacidmethylester,17-Octadecynoicacid,Octadecanoicacid methyl ester, Pentadecanoic acid, and Octadecanoic acid compounds. After four MIRGAspray treatments, there were new peaks that were not present in control, including Methyl 10-Methyl-undecanoateand17-Octadecynoicacid.However,therewereuniquepeaksofHexadecanoic acid, methyl ester in samples that received 4 MIRGA spray treatments, andPentadecanoicacidinsamplesthatreceived12MIRGAspraytreatments,comparedtocontrol.Interestingly,there was adisappearance of 11-Octadecenoic acid methyl ester fromtheGC-MSspectraof bitter gourdpowderduetoMIRGAspray treated samples.ItwasobservedthatMIRGAspraytreatmentsalsocausedagradualincreaseinthepeakofn-HexadecanoicacidandagradualdecreaseinthepeakofOctadecanoicacid.

3.2.2. ChemicalBondsAnalysisviaFTIR

(a) Carrotpowder

The FTIR spectra of control carrot powder displayed a big peak was observed at 3396cm^{-1},whichwasspecifictocarbohydrateandOH-groupsfromwater.Twomorepeakswereobserved: a peak at 2925 cm^{-1}, specific to C-H bonds from organic compounds, and a peak at1632 cm^{-1},whichresembled N-H bonds (near C=O)fromproteins. In carrot powder samplesthat were MIRGA spray treated 2 times and 7 times, a small increase in the absorbance in allareas was observed compared to in control. The increased absorbance was probably due toincreasing theconcentrationofasubstance(carbohydrates,OH-groups,organiccompounds,or proteins) in the compaction matrix. In samples that were MIRGA spray treated 2 times, theabsorbance was lower at1632cm^{-1}and 618 cm^{-1},which was probably caused by a lowerproteinconcentration.Theenhancedsweetnesswasassociatedwiththebreakdownofaportionofpolysaccharidesinto monosaccharides**(Shah *et al*., 1978)**.

(b) Beetrootpowder

Abigpeak wasobservedat 3392cm^{-1},fortheFTIRspectraofcontrolbeetrootpowder,whichwasspecifictocarbohydrateandOH-groupsfromwater.Twomorepeakswereobserved: a peak at 2929 cm^{-1}, specific to C-H bonds from organic compounds, and a peak at1631 cm^{-1}, similar to N-H bonds (near C=O) from proteins. All peaks became smaller after 2MIRGA spray treatments, compared to the control and 8 sprayed samples. 8 sprayed sampleswere almost identical to the control samples, except for peaks at 1631 cm^{-1} and 618 cm^{-1}. AnexplanationforthisbehaviourisduetoproteinreductioncausedbytheMIRGAspraytreatments.

(c) Bittergourdpowder

The control bitter gourd powder had abig peak of OH-groups at 3406 cm^{-1}. This peak wasspecifictocarbohydratesandOH-groupsfromwater.Absorbanceatthiswavelengthincreased when samples received 4 MIRGA spray treatments and decreased when samplesreceived eight MIRGA spray treatments. It could be proposed that absorbance increased afterfour treatments because MIRGA spraying caused an increase in the concentration of requiredsubstances (carbohydrates and OH-groups),andthereduced absorbanceafter 8 treatmentsresulted from water evaporation. An increase in the peaks at 2925 cm^{-1}, from C-H bonds oforganiccompounds,andat1741cm^{-1},possiblyC=Obondsfromcarbohydrates,wereobserved for samples that received 4 MIRGA spray treatments. The increase in the peaks at2925 cm^{-1}and 1741 cm^{-1}weremorepronounced in the samplesthatreceived eight MIRGAspray treatments.Therefore, theconcentration ofcarbohydrates seemed to increase with ahigher number of MIRGA spray treatments. The opposite trend was observed for proteins. Apeak at 1631 cm^{-1}, similar to N-H bonds (near C=O) from proteins, decreased slightly after 8MIRGAspraytreatments,anditwashighlyreducedafter4MIRGAspraytreatments.

3.2.3. ParticleStructureAnalysisviaTEM

(a) Carrotpowder

Asshowninfig.3b,carrotpowdersampleswhichreceived2MIRGAspraytreatmentsshowed minorchanges ofmorphology and matrix structure,and negligibleor nochanges oftheatomicarrangement,comparedtocontrolsamples.Incontrast,carrotsamplesthereceived 7 MIRGA spray treatments were visibly affected in terms of morphology and matrixstructure, compared to control samples. Therefore, as the number of MIRGA spray treatmentsincreased,morenoticeablechangesmayoccurinthemorphologyandmatrixofcarrotpowder

(b) Beetrootpowder

MIRGAsprayinghaddifferenteffectsonthesizeandshapeofmatrixcomponentsofbeetroot powder, therefore it changed the overall sample structure. In fig. 3a, the main matrixcomponentsofthebeetrootcontrolsamplescanbeidentifiedasamorphous-shapedfragments, but in the samples that received 2 MIRGA spray treatments the fragments wereclustered in large aggregates, and in the samples that received 7 MIRGA spray treatments thesizeofthefragmentwasnoticeably smaller.

(c) Bittergourdpowder

Alterations in the structure, matrix components in numerosity, shape, and morphology wereobserved on bitter gourd powder samples that received MIRGA spray treatments compared tocontrol.However,theatomicarrangementwasonlyslightlyaffectedasseeninFig.3a.Peculiarly, 4 MIRGA spray treatment caused more evident changes than 12 MIRGA spraytreatments.

3.2.4. ChemicalConfigurationAnalysisviaPXRD

(a) Carrotpowder

As seen in fig. 4, all three carrot powder samples showed a large volume (%) of amorphousphases reflected by one broad peak centred on 17°. A general trend was observed where thebroad peak shifted to the right with an increased number of MIRGA spray treatments. Thecontrol samples and the samples that received 2 MIRGA sprayed sample showed broader andoverlapping peaks (10° -28°) compared tosamples thatwere MIRGA spray treated7 times.Theprominent peak at31° had afixed location inallthe control and sprayedsamples,evenasthenumberof MIRGA spraying increased.Samples that received 2

MIRGAspraying hadone more peak at 29.07°,whichsuggested an increase ofthe crystalline phase. Most peaksbelow31°inallthesamplesagreewiththevaluespresentedby**Rocha*etal*.,2011**.

(b) Beetrootpowder

Thespectraofbeetrootpowdershowedgoodsignal-to-noiseratios,allowingtheidentification of several peaks. Control samples had the least intense peak reflection. SamplesthatwereMIRGAspraytreated2timespresentedmorepeaksintheirspectrathansamplesthat were MIRGA spray treated 8 times, although thelattershowed arelatively intensepeakat35.59°.Thepeakaround24.80°ishighestforsamplesthatreceived2MIRGAspraytreatments,followedbysamplesthatreceived8MIRGAspraytreatments,andcontrolsamples had the smallest peak. In the case of beetroot powder that received 2 MIRGA spraytreatments,newpeaks at27.81° and 35.59° weredisplayed compared tocontrol samples.Samples that received 8 MIRGA spray treatments also showed new peaks at 35.69°, but withhigherintensitythan forsamplesthatreceived2treatments

(c) Bittergourdpowder

Consistentwiththeliterature**(Mariselvi*et al*., 2017; Singh *et al*., 2017)**,allbittergourdpowdersampleshadcharacteristicspeaksat around 14°, 21°, and 26°for their pattern of XRD (Fig. 4). Control samples had the leastnumber of prominentpeaks among the three samples(control,andsamples that received 4 or12MIRGAspraytreatments).Samplesthatreceived4MIRGAspraytreatmentshadthehighest number of intense, narrow peaks, while the samples that received 12 MIRGA spraytreatmentsshowedthebest signal-to-noise ratio. A considerablechangein thepresence ofnew peaks was observed in samples that received 4 MIRGA spray treatments, particularly at28.25°,indicatingtheformationofanewcrystallinephaseinthesamples.Moredrasticchanges in the structure were observed in samples that received 12 MIRGA spray treatments,wheretheintensityofthepeakin21.49°wasreduced,andthepeakat26.66°becamethemostprominent,compared tocontrol samplesandsamplesthat received 4 MIRGAspraytreatments. Samples that received 12 MIRGA spray treatments had the largest volume (%)crystallinephaseamongthethreesamples(control,andsamplesthatreceived4or12MIRGAspraytreatments).Controlsamplesandsamplesthatreceived4MIRGAspraytreatments had a similar structure. The peak centred around $2\theta = 26°$ in each of the threesamples(control,and samples thatreceived4 or12 MIRGAspray treatments)changed interms of breadth and intensity, indicating a change in crystallinity as the number of MIRGAspraytreatmentsincreased.

3.2.5. NuclearResonanceAnalysisvia1H-NMR

(a) Carrotpowder

Sweetness and aroma of carrot powder depended on sugars and terpenoid compounds. Thecontrol samples showed a profile compatible with a sugar-rich composition and other minorcomponents. The interpretation of H1-NMR spectra was challenging due to signal overlap.However,itispossible to infer from Fig 5 thattheMIRGA spray treatment affected theconcentration, or the integrity of molecules involved in the sweetness and aroma of carrotpowder.

(b) Beetrootpowder

An increase of sweetness, aroma, and solubility was observed in samples that received 2MIRGAspraytreatments.Sweetness,aroma,andsolubilityweredirectlyrelatedtotheincrease in the concentration of sucrose, geosmin, and soluble compounds, respectively. Theintegral corresponding to the region of the signals coming from sucrose was higher than thatfrom the control samples.Since geosmin is a minor component,its variation was hidden bythe overlap of its signal with more intense signals. In the case of samples that received 8MIRGA spray treatments, changes followed the opposite trend: a drastic reduction in theconcentrationofsucrose,geosmin,andsolublecompoundswasobserved.Therefore,sweetness,aroma,andsolubilitywerereduced.

(c) Bittergourdpowder

Bittergourdpowdersamplesthatreceived4MIRGAspraytreatmentsshowedareductionintheconcentrationofterpenoidsandpolyphenols,whichcouldbeattributedtoareducedbitterness. 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 differentchemicalgroups.Forthisreason,itwasdifficulttointerpretwhichvariationsintheNMRspectra correspond to changes in the concentration of molecules involved in the perception ofbitterness.

Inthecaseofsamplesthatreceived12MIRGAspraytreatments,anincreaseinbittercompounds was observed in the spectra. Paradoxically, all the integrals showed a reduction inbitter compounds for samples that received 12 MIRGA spray treatments, compared tothecontrolsamples.Apossibleexplanationforthisresultwouldbethattherewereminorcomponents in bitterness, and the augmentation effect that they have on the perception ofbitternesswashiddenbytheoverlapwithsignalscomingfrommoreconcentratedcomponents.

*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*., 2022a**and **Umakanthan *et al*., 2023d***(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**(Prosekov et al., 2018)**, nanotechnology related strategies such as nano-zincdioxide and silver nanoparticles **(Wenchao et al., 2020)**, edible coating with pulsed light treatment **(Annachiara et al., 2021)**and evaporation cooling system**(Ayobami et al., 2023)**. Infrared was found to influence the vegetables shelf life**(Faisal et al., 2008)**. The sensory quality of vegetables is improved through seasonings**(Ulla et al., 2021)**. Not in this study simultaneously enhanced the shelf life and sensory qualities by exposing vegetables to 2-6 μmmid-infrared.

At present, chemical preservatives and ultraviolet,microwave,pulsed electricfields,gammaandinfrared radiations areused for foodprocessing.Exceptinfrared,otherradiationsareionizing,hencelethal,uneconomicalandnon-friendlytouserandecology**(Lopez-Malo*et al*., 2004; Gautam *et al*., 2016; Vasuja*et al*., 2018;**

Aboud *et al*., 2019).Thustheuseof infrared in food processing is a future option **(Aboud *et al*., 2019)**. Though infrared radiation use has manyadvantagesthantheconventionalmethodbutnotwithoutchallengesandresearch**(Das *et al*., 2014)**.ItisconcludedthatMIRGAovercomethedisadvantagesofpresentfoodprocessingtechnologiesandexpectedtoconvincetheindustryand consumer.

Similar desirable results in coffee, tea, cocoa, edible salts and terminalia were achieved using MIRGA spraying by **Umakanthan *et al*., 2022a; Umakanthan *et al*., 2022b; Umakanthan *et al*., 2023c; Umakanthan *et al*., 2023d.**

4. Conclusion

Through sensory trials and various instrumentations, we demonstrated that MIRGA spraying(from0.25-0.50meter)treatmentsalteredthechemistryofpolythenepackagedvegetablesamplesandmadethemmoredesirableintermsofaroma,tasteandshelflifeenhancement.Theimpact ofthis study involves the developmentofa processing technology that couldimprovethearoma,sensoryquality,andshelf-lifeofvegetableswithoutgeneticmanipulation.In outexperiencewithMIRGAsprayingtechnology,itispossible to further enhance the taste, aroma, and shelf-life by altering the MIRGAformulation. 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&ouid=111101387151809704391&rtpof=true&sd=true

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Table1:Sensoryprofilingoffreshvegetables

No. ofMIRGA sprayings	**Freshvegetables**															
	Lady’sfinger		**pumpkin**		**Beans**		**Carrot**		**Brinjal**		**Bottlegourd**		**Beetroot**		**Bittergourd**	
	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

Table2.Shelf-lifestudyoffreshvegetablesamplesbasedonthecomparisonofthenumber of days that the samples remained unspoiled at room temperature (app. 32 °C)andrefrigerationtemperature(4°C)(standarddeviation±oneday)

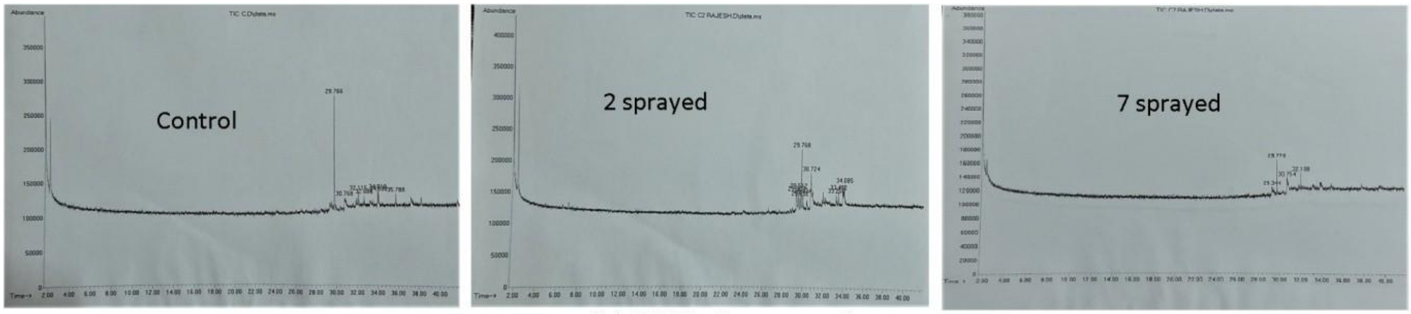
NumberofMIRGAspraytreatments	Freshvegetablesample	Numberofdaysunspoiledatroomtemperature (app.32°C)	Shelf-lifeincreaseatroomtemperature (%)	Numberofdaysunspoiledatrefrigeratedtemperature(4 °C)	Shelf-lifeincreaseatrefrigerationtemperature (%)
0(control)	Lady'sfinger	6	-	15	-
0(control)	Pumpkin(sliced)	2	-	10	-
0(control)	Carrot	3	-	10	-
0(control)	Beans	5	-	10	-
0(control)	Beetroot	5	-	14	-
0(control)	Brinjal	4	-	7	-
0(control)	Bottleguard	3	-	10	-
0(control)	Bitterguard	3	-	7	-
1	Lady'sfinger	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	Bottleguard	25	733	37	270
1	Bitterguard	13	333	17	142
2	Lady’sfinger	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	Bottleguard	19	533	32	220
2	Bitterguard	10	233	16	128

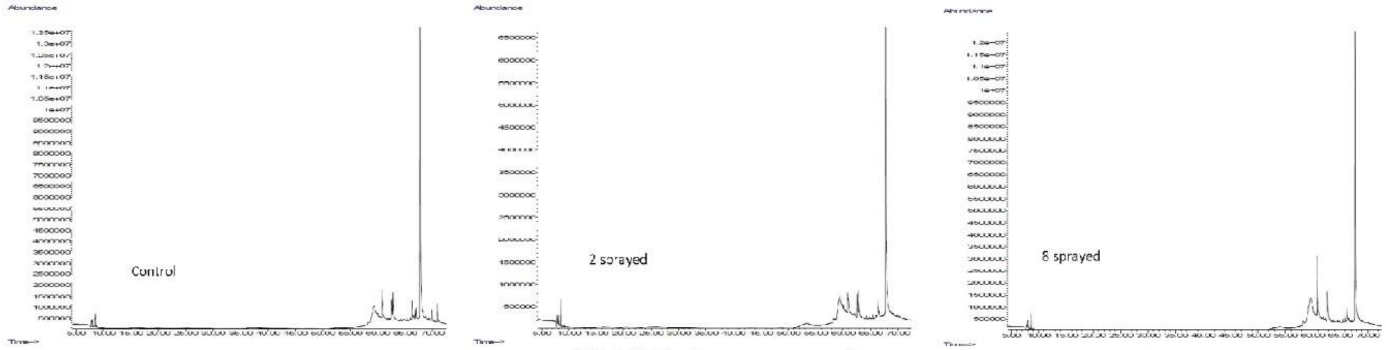
Table3:Sensoryprofilingofvegetable powders

No.ofMIRGAsprayings	Vegetablepowders					
	Carrotpowder		Beetrootpowder		Bittergourdpowder	
	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

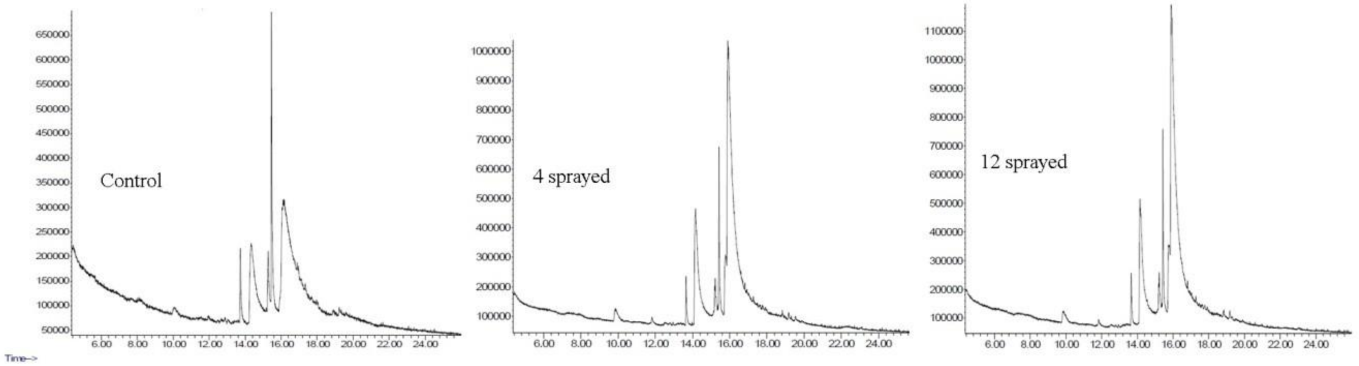
10					2	3
11					1	3
12					1	1



(a) GCMS - Carrot powder

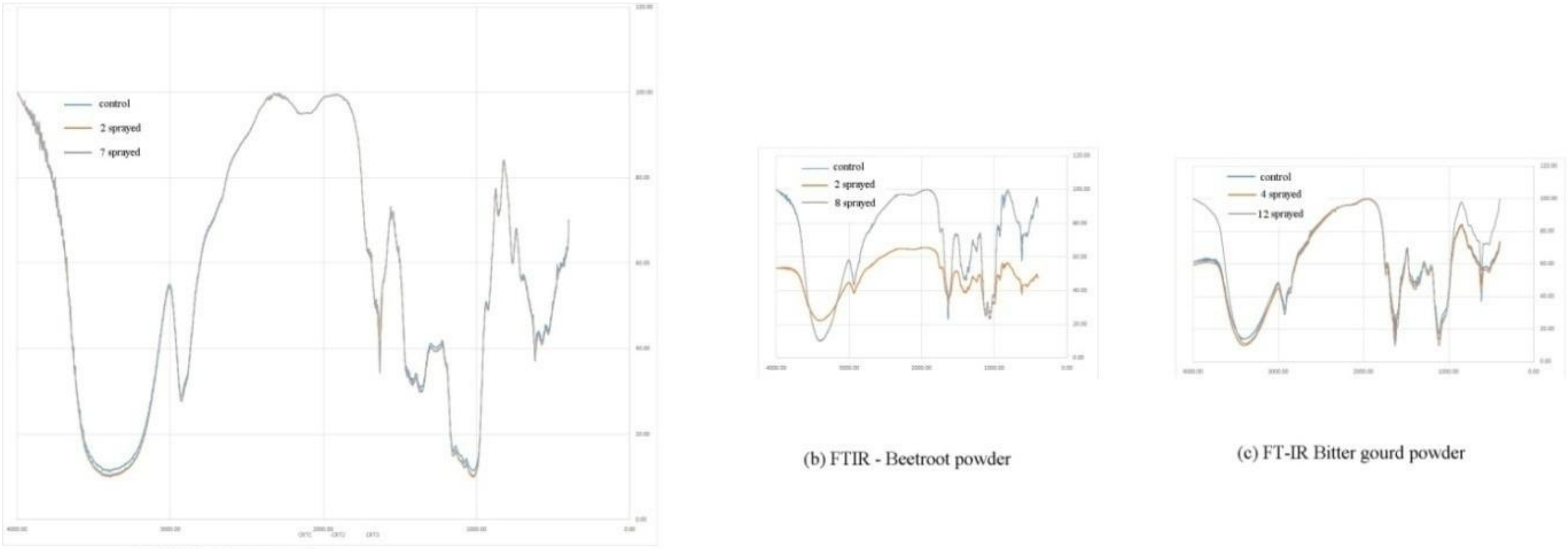


(b) GCMS - Beetroot powder



(c) GCMS - Bitter gourd powder

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

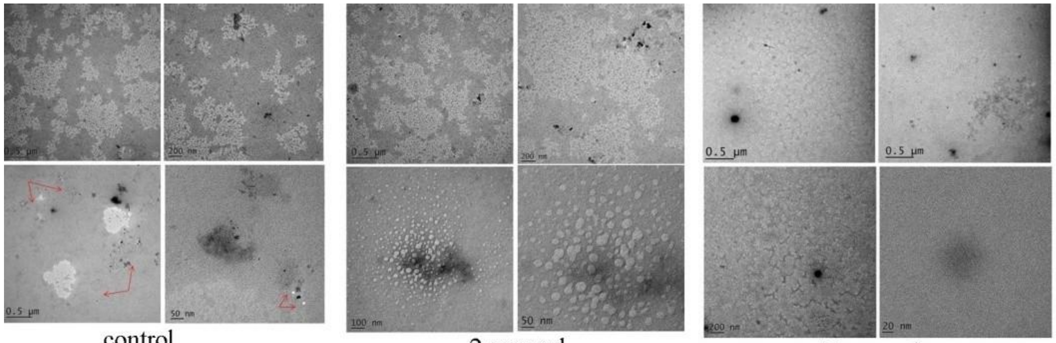


(a) FTIR - Carrot powder

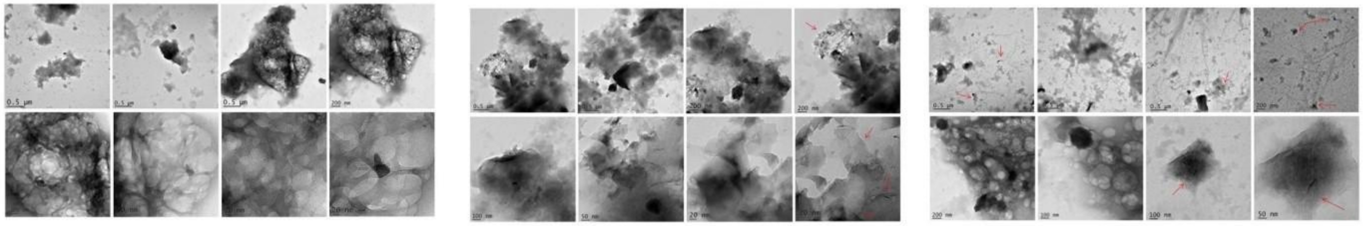
(b) FTIR - Beetroot powder

(c) FT-IR Bitter gourd powder

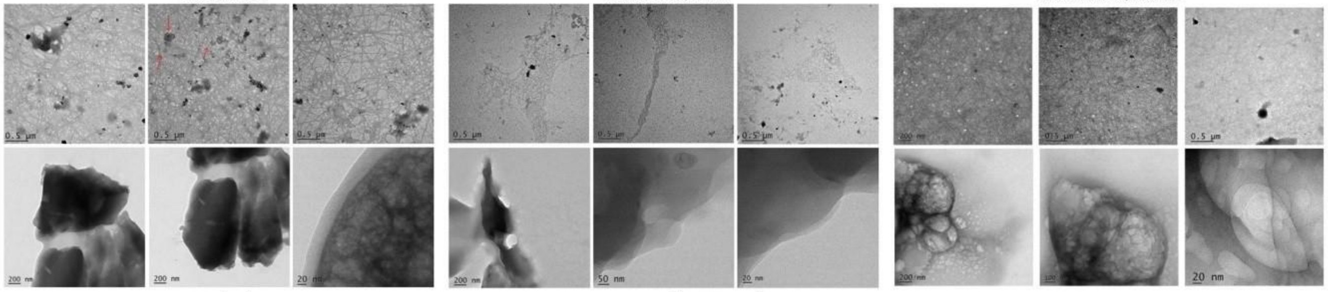
Fig.2.FTIRspectraof(a)carrotpowder,(b)beetrootpowder,and(c)bittergourdpowdertreated withMIRGAspraying.



(a) TEM - Carrot powder



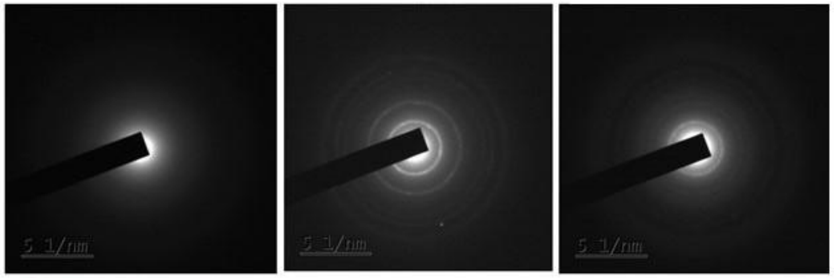
(b) TEM - Beetroot powder



(c) TEM - Bitter gourd powder

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

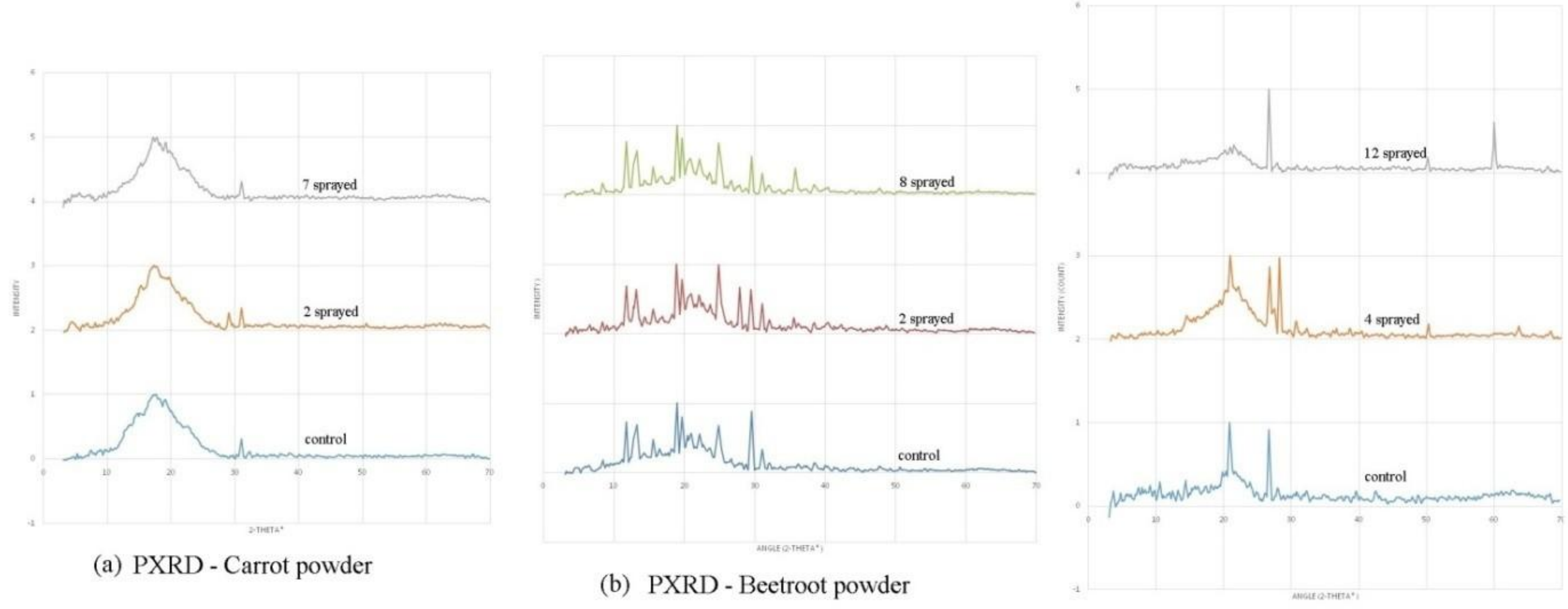
(a) Carrot powder



(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.

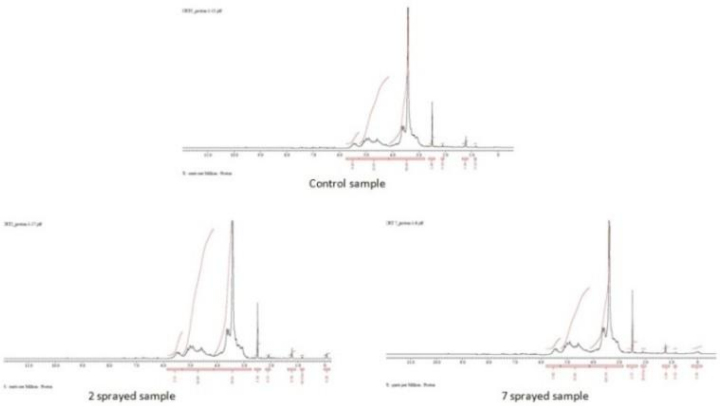


(a) PXRD - Carrot powder

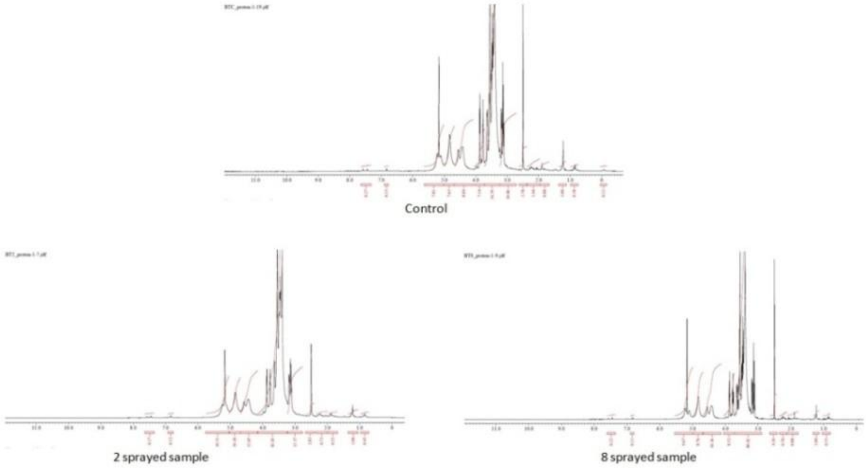
(b) PXRD - Beetroot powder

(c) PXRD - Bitter gourd powder

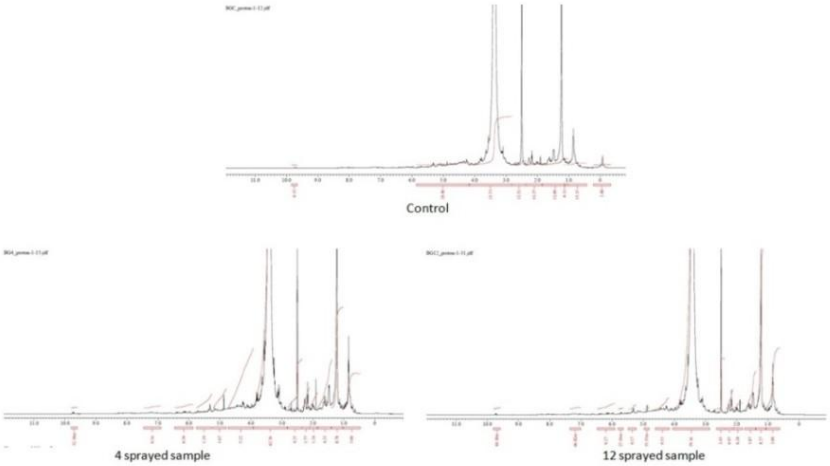
Fig4. PXRD spectra of (a) carrot powder, (b) beetroot powder, and (c) bitter gourd powder treated with MIRGA spraying.



(a) NMR- Carrot powder



(b) NMR - Beetroot powder



(c) NMR - Bitter gourd powder

Fig5.H-NMRspectraof(a)carrotpowder,(b)beetrootpowder,and(c)bittergourdpowdertreated withMIRGAspraying