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Development of an Antioxidant-Rich Sugar-Free Plantain Candy and Assessment of Its Shelf-Life in a Flexible Laminate

Running title: Nutraceutical Candy Having Synergy among L-tryptophan, Serotonin and Melatonin

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SUMMARY

Research background. Candy is a popular confection worldwide, and it would be of societal benefit if it could be converted into a source of antioxidant molecules, to eliminate their adverse health effects. The amount of antioxidants available even in fruit candies is questionable due to the high thermal processing losses they undergo and the presence of various food additives. Plantains (*Musa paradisiaca*) are less known as good sources of biotherapeutic antioxidants, namely L-tryptophan, serotonin, and melatonin, and consumption of this highly nutritious fruit is limited to underdeveloped and developing countries. The objectives of this current study are: to develop a functional antioxidant-rich sugar-free plantain-based candy with appreciable contents of the mentioned biomolecules in synergy; and to ensure its extended shelf-life without compromising its physicochemical properties and functionalities by wrapping it with a suitable packaging laminate.

Experimental approach. To accomplish the first objective, lyophilized plantain powder, sorbitol, and mannitol were used as base materials with minimal additives under minimal processing conditions

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to reduce processing losses. After development, the developed candies were evaluated for their sensory, proximate, physicochemical, and phytochemical properties, including the antioxidant synergy among the mentioned biomolecules. For the second objective, the candies were enclosed in two different flexible packaging laminates, and the optimal wrapper was determined based on the microbiological safety and sensory appeal of the packaged candies. Subsequently, the above-mentioned properties were assessed for the packaged (in the most suitable laminate) candies at regular time intervals during storage for assessment of their shelf-life.

Results and conclusions. The candy exhibited a characteristic flavour of plantain, uniform dark brown colour, rich mouthfeel, pleasant aroma, moderately hard texture, and moderate sweetness, along with high antioxidant activity and considerable content of L-tryptophan, serotonin, and melatonin (present as a synergistic consortium). During storage of the packaged candy under ambient conditions, it remained microbiologically safe for up to 56 days, and also maintained sensory attributes, antioxidant potency, and synergy compared to the control candy.

Novelty and scientific contribution. It is envisaged that this newly developed semi-hard antioxidant-rich sugar-free candy containing three important antioxidants, namely L-tryptophan, serotonin, and melatonin, could be a food matrix for molecular nutrition and a substitute for commercial candies consumed globally.

Keywords: minimal additives; biomolecules; sensory, physicochemical and biochemical characteristics; flexible packaging; shelf-life

INTRODUCTION

Green plantains (*Musa paradisiac*a) are extensively consumed in Asia, Africa, and South America, particularly for their high contents of carbohydrates (including resistant starch), dietary fibres, and minerals, such as iron (*1*). However, they are less recognized for their abundant antioxidants. Our previous studies on green plantains found them to be a rich source of antioxidants, primarily for the monoamine neurotransmitter serotonin (5-135 µg/g), the neurohormone melatonin (0.5-7.2 µg/g), as well as their biosynthetic precursor molecule- the essential amino acid L-tryptophan (*2*). In some less developed countries with food shortages, plantains serve as a staple food (*3*). The development and consumption of plantain-based food products are limited to wine, beer, pickles, chips, cookies, bread and cakes, *etc*. (*3-7*). However, there is limited knowledge on candy making with plantains.

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Candies are very popular with all age groups worldwide. In America, 26 % of the total population (aged≥2 years) consume an average of 40 g of candy (176 kcal) per day (*8*). However, the added sugars and saturated fats present in candies are specifically responsible for several health-debilitating metabolic and cardiovascular disorders (*8*). In addition, the presence of synthetic antioxidants such as butylated hydroxyanisole (BHA) and butylated hydroxytoluene (BHT) (*9)* in commercially available candies do pose adverse health consequences (*10*). Even in candies produced from natural fruit or vegetable pulps and juices, the contents of natural antioxidants are compromised consequent to heat degradation that occur during thermal processing. Furthermore, the presence of miscellaneous food additives in them in addition to high fat and sugar, could possibly impede utilization of their health-beneficial antioxidants (if they survive post processing). These do not conform to labelling the fruit/vegetable candies to be antioxidant-rich. We envisage that minimal processing encompassing usage of minimal additives with concomitant low sugar content can render a plantain-based candy to be truly antioxidant-rich and thereby minimize adverse health effects.

Some recent studies documented the formulation of candies with substitutes of the commonly used ingredients to circumvent the deleterious effects of candy consumption. Few researchers have successfully incorporated spices such as a mixture of turmeric, ginger, liquorice, white wormwood, and citrus lemon extracts (*11*) to develop antioxidant-rich hard candies. Several studies have reported the formulation of fruit candies utilizing various fresh fruits, including unripe mango (*12*). Acetic, alcoholic, and lacto-fermented apple juice products (*13*), as well as fermented algae such as *Spirulina* (*14*) have been used in the production of chewy candies. The production of soft candies (gummy jellies) by adding pomegranate juice (*15*) and other natural extracts is well documented. A few researchers have also explored preparation of the same using starch or sugar substitutes such as inulin (*16*). Furthermore, use of natural antioxidants such as from stevia-rosemary extract (*17*) along with fructan fibres (chicory inulin and fructo-oligosaccharides), green propolis extract with fructan (*18*), and sage extract with inulin-gelatine-fructooligosaccharides (*19*) have also received recognition. However, information on hard and semi hard candy production in the truest sense is lacking.

Studies on the formulation of hard and semi-hard candies utilizing sugar substitutes are scarce, except for a solitary report by Jeon *et al.* (*20*) on the formulation of a nutraceutical hard candy using isomalt, maltitol syrup, and xylitol as sucrose substitutes. Here extracts of *Cudrania tricuspidata* (manadarin melon berry) fruit, lemon, and ginger as sources of natural antioxidants have been utilized. However, to the best of our knowledge, there is no report on the development of natural antioxidant-

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rich (plantain) sugar-free hard or semi-hard candy emphasizing the molecular identities of important biotherapeutic molecules, particularly L-tryptophan, serotonin, and melatonin.

Therefore, the first objective of the present study was to develop an antioxidant-rich sugar-free candy that could be a potential dietary source of L-tryptophan, serotonin, and melatonin without perturbing the intrinsic natural antioxidant synergy (*21*). The chief impediment in designing antioxidant-rich foods lies in the preservation of the natural food synergy (*22*), which, if altered, would render the consortium of antioxidants present in the finished food product detrimental rather than beneficial *in vivo* (*21*). The current investigation therefore focused on the modification of the candy processing parameters by (i) using minimal food additives to aid unimpeded release of antioxidants; (ii) minimizing the losses of heat-sensitive molecules (chiefly serotonin and melatonin); (iii) employing minimal processing not only to preserve the natural antioxidant synergy of the green plantains but also to avert the destructive side effects of consuming ultra-processed food products (*23*); and (iv) utilizing substitutes for the classical 'Doctors' (*24*) to deliver a nutraceutical yet palatable sugar-free candy.

The second objective of the study was to assess the shelf-life of the newly formulated designer candies after packaging in commercially available flexible laminates (chiefly foil wrappers) to ensure an extended shelf-life without compromising the physicochemical properties of the candies throughout the storage period.

The novelty of this investigation lies in designing a value-added food product using a widely cultivated healthy fruit. This could be a unique confectionary product housing important biotherapeutic antioxidant molecules, besides promoting better utilization of plantains globally.

MATERIALS AND METHODS

Authenticated Indian ('*desi'* variety) green plantains (*Musa paradisiaca)* cultivated in Narendrapur (Kolkata, India) were used for the present study. The details of authentication, procurement, and selection of plantains have been elaborately described in our previous publication (*2*). Individual plantains were meticulously selected for this study based on visual assessments of their colour (only the green unripe ones were chosen); surface morphology, texture (turgid and free from blemishes, bruises, and black spots), and size-weight [~(175±10) g on an average having ~(120±5) mm diameter].

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For candy ingredients, food-grade chemicals such as D-sorbitol, D-mannitol, gum acacia, SiO_2, sorbic acid (all in powder form), and vanilla essence were purchased online from Amazon.in. Specialty chemicals, such as 2,2-diphenylpicrylhydrazyl (DPPH), high performance liquid chromatography (HPLC)-grade acetonitrile, acetic acid, and water; Folin Ciocalteu's reagent; standards such as L-tryptophan, serotonin, and melatonin, gallic acid; and all other AR-grade chemicals and culture media for microbiological analyses were purchased from M/s Merck (Mumbai, India) and M/s HiMedia (Mumbai, India). For solid-phase extraction, Quick, Easy, Cheap, Effective, Rugged, and Safe (QuEChERS) kits were purchased from M/s Agilent Technologies, Wilmington, DE, USA.

The experimental design of the present study has been elaborated in Fig. S1. For the development of a new plantain-based antioxidant-rich sugar-free candy, it was necessary to first assess the antioxidant potential of raw plantains as well as the content of the desired molecular antioxidants, namely L-tryptophan, serotonin, and melatonin. Three plantain samples were randomly selected from the procured sample lot and analysed for their antioxidant potency; chiefly their total phenolic content (TPC), DPPH radical scavenging activity, and ferric ion-reducing antioxidant power (FRAP), as well as their contents of L-tryptophan, serotonin, and melatonin. Detailed methodologies for the same can be accessed from the previous publication of the authors (*2*).

Preparation and characterisation of lyophilized plantain powder

The fully matured green plantains were washed thoroughly, then cut into very small cube-shaped pieces along with the skin (except for the stalk and the black tip) and frozen in an ultra-low temperature freezer (Premium C340, M/s New Brunswick Scientific, Enfield, CT, USA) at −80 °C for 24 h and finally dried to a final moisture content below 6 % using a bench top freeze dryer (FDU-1200, M/s Eyela, Tokyo, Japan) at −45 °C, 14.4 Pa. The freeze-dried plantain pieces were then ground in a mixer grinder (HL 1618, M/s Philips India Limited, Chennai, India) to obtain a powder with a particle size of <300 µm, and the same was subjected to solvent extraction in accordance with the method described by Dal-B´o and Freire (*25*), with few modifications. The lyophilized plantain powder (0.5 g) was initially dissolved in dimethyl sulfoxide (DMSO) (in a 1:10 solid/solvent ratio) in an incubator shaker (IS 02, M/s Incon Instruments Company, New Delhi, India) at 120 rpm for 2 h at 30 °C, following which the mixture was centrifuged (R-8C, M/s Remi, Mumbai, India) at 1,000×*g* for 15 min. The supernatant was analysed for its antioxidant activity (in terms of TPC, DPPH radical scavenging activity and FRAP value) in accordance with the procedures followed for the raw plantains

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(*2*). The lyophilized plantain powder was subsequently used in the preparation of the candy (*vide infra*).

Formulation of a plantain-based, antioxidant-rich, sugar-free candy

The ingredients selected for the formulation of the antioxidant-rich candy, their functions, and the safe limits of usage (*26*) have been presented in Table S1. Several preliminary trials were conducted to determine the exact amount of each ingredient for the candy formulation. Sugar alcohols such as D-sorbitol and D-mannitol were selected as the base ingredients (*27*) for the candy formulation, mainly because they provide fewer calories (3 cal/g and 2 cal/g, respectively) (*28*); have a negligible glycemic index (4 and 2, respectively) (*29*) compared to regular table sugar (4 cal/g; glycemic index: 65); and also serve as plasticizers (*9*). Since D-sorbitol and D-mannitol have a relative sweetness of 0.6 and 0.5-0.6, respectively (*30*), compared to sucrose (relative sweetness: 1), it was opined that the sweetness of the formulated candy should not be compromised. Gum acacia as a thickening agent was varied in a concentration range of 3-35 %. The preliminary trials established that an amount of 30 % (based on the total weight of the candy ingredients) could impart the desired "stand-up'' property to the candy. The amount of lyophilized plantain powder was varied in a concentration range of 55-85 % of the total weight of the candy ingredients. It was found that a concentration of 65 % conferred desirable sensory properties to the candy. Quantities less or more than 65 % imparted improper texture to the candies, i.e., they were either too hard or poorly formed, i.e., lacking structural integrity (data not shown).

To formulate a plantain-based candy sample (PCS), sorbitol, mannitol, gum acacia, SiO_2, sorbic acid, and water were mixed using a spatula, and the mixture was heated (180-210 °C) with constant stirring (to prevent lump formation) until it acquired the desired caramel colour. Then the lyophilized plantain powder and vanilla essence were added to the mixture after it had cooled to 60 °C to preserve the targeted heat-labile antioxidants. The mixture was then 'seeded' with sorbitol and mannitol powder and allowed to set inside candy moulds for 75 min. In this procedure, the use of food additives was kept to a minimum. No synthetic food colours, organic acids (acidulants), or synthetic antioxidants were used in the formulation of the candies, contrary to those used in the production of commercial candies (*9*). In the formulation of the experimental control candy (without the plantain) sample (CCS), all ingredients and procedures were similar to those used for formulating PCS, except that no plantain powder was used. The weights and dimensions of the resulting CCS and PCS

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samples were measured using a weighing balance (BSA 224S-CW, M/s Sartorius AG, Göttingen, Germany) and a vernier calliper (532 series, M/s Mitutoyo, Kawasaki, Japan), respectively.

Assessment of the safety parameters of CCS and PCS

The analyses described below were performed to evaluate the safety profile of the newly formulated candies.

Microbiological assessment

The microbial load of the candies for bacteria and fungi (as CFU/g candy) was evaluated in terms of total plate counts of the formulated candies using the pour plate method (*31*).

Energy dispersive X-ray (EDX) analyses

To ensure the absence of toxic heavy metals such as Pb, Ti, Hg, Ni, As, Si, and Mo, that may be present in the raw material (plantains) or may have been acquired during the production of the candies, EDX analyses of CCS and PCS were performed using INSPECT F50, M/s FEI Company, Hillsboro, OR, USA.

Sensory and physicochemical characterisation of CCS and PCS

The analyses described below were performed to evaluate the eating quality of the newly formulated candy samples primarily in terms of characteristic plantain flavour (assessed by sensory evaluation) and mouthfeel (assessed by both sensory and by textural evaluation).

Sensory evaluation

The sensory evaluation of CCS and PCS was performed using an acceptance test based on an effective method, namely, rating on a 9-point hedonic scale (where 9 denotes ‘like extremely’ and 1 denotes ‘dislike extremely’). The candy samples were served to the participants using the serving protocols used for sensory evaluation of foods and beverages described in E1871-17 ASTM International methodology (*32*). Fifteen men and fifteen women (35-45 years old) were selected from faculty members and research scholars of our university to form a semi-trained sensory panel (*33*). Panellists were selected based on their interest, and their performance was assessed using screening tests conducted with the control sample. Prior to sample evaluation, they were well acquainted with the sensory attributes of the prepared candies and the commercially available hard candies (*Mango*

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bite and Poppins) and asked to evaluate the experimental candies in terms of appearance, texture, colour, taste, flavour, mouthfeel, aftertaste, and overall quality. The sensory evaluation was conducted in the morning from 10 a.m. to 12 noon in a well-ventilated room with white light. Unsalted crackers, and water (to rinse the palate) as well as expectoration cups (covered) were provided to all participants before each evaluation if they did not wish to swallow the samples (*34*). Triplicates of each individual sample were served in each session, and the mean scores (rounded off) were presented graphically in radar plots.

Colour and texture analyses

Instrumental analyses of the colour and texture properties of the newly formulated candy samples were performed to validate the sensory ratings provided by the human judges (as described above) and to remove subjectivity. CIE colour value analysis (L^* for lightness; a^* for red-green; b^* for yellow-blue) of CCS and PCS was conducted using a colour reader (CR-10 Plus, M/s Konica Minolta Inc., Osaka, Japan) (*2*). The texture profiles of CCS and PCS were analysed by the two-bite compression test using a texture analyzer (TA.XT Express, M/s Stable Micro Systems, Godalming, UK) in accordance with the method described by Sarkar *et al.* (*2*) and the data were processed by Exponent Lite Express software version 6.1.4.0 (M/s Stable Micro Systems Ltd., Godalming, UK) (*35*). A cylindrical aluminium probe (P/5) of 5 mm diameter was used for the test, and the sample was oriented so that the compression was symmetrical to its geometric centre.

Microstructure analyses of CCS and PCS

The following analyses were carried out for the detailed structural characterisation of the newly formulated candies.

Field emission scanning electron microscopy (FE-SEM)

The surface morphology of the new designer candies was investigated using FE-SEM (INSPECT F50, M/s FEI Company, Hillsboro, OR, USA) at an operating voltage of 5 kV. First, the samples were dried under vacuum (6-7 Pa) and then coated with gold using a coating device (Q150R ES, M/s Quorum Technologies Ltd., Ashford, England).

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X-ray diffraction (XRD) analysis

The XRD patterns of CCS and PCS were analysed at ambient temperature [(23±2) °C] (D8 Advance, M/s Bruker, Billerica, MA, USA) using Cu-Kα1 radiation at a wavelength of 0.15406 nm. The measurements were performed at a voltage of 40 kV/40 mA. The XRD data were collected to cover an angular range (2 θ) of 30-100 ° at a width of 0.01 ° and a counting time of 0.5 s/step.

Thermal stability assessments of CCS and PCS

It is necessary to study the thermal stability of the candies in order to assess the thermal (and therefore structural) changes that the candies could undergo during storage and transportation, post-packaging under ambient conditions (*vide infra*).

Thermogravimetric analysis (TGA)

To predict the thermal stability of CCS and PCS, a TGA was performed using the TGA 4000 (M/s Perkin Elmer, Hopkinton, MA, USA). In this analysis, an empty platinum crucible containing α-alumina powder was used as a reference. The samples, which were individually placed in a hermetically sealed Al pan, were heated from 51 to 600 °C and scanned at a rate of 10 °C/min under a nitrogen flow of 20 mL/min.

Differential scanning calorimetry (DSC) analysis

For DSC analysis, CCS and PCS were equilibrated at -80 °C for 5 min, then heated to 130 °C at a rate of 5 °C/min and held this temperature for 1 min. The samples were then cooled to −50 °C at a rate of 5 °C/min and held at this temperature for 5 min. The samples were then reheated at a rate of 5 °C/min to 110 °C, using the heat-cool-heat method, which was considered suitable for hard candies (*36*). For this study, the samples were analysed using a DSC Q2000 (M/s TA Instruments, New Castle, DE, USA).

Analysis of the water activities (a_w) *of CCS and PCS*

The water activities of CCS and PCS were determined according to AOAC (*37*) to assess their shelf stability.

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Proximate analyses of CCS and PCS

Proximate analyses of CCS and PCS were conducted in accordance with the standard AOAC methods, including estimation of % moisture (dry mass basis-D. M.), % protein, % crude fat, % ash, % crude fibre, and total carbohydrates by difference (*2*).

Acidity and solid contents of CCS and PCS

For evaluating acidity and solid contents of the CCS and PCS samples, the same were dissoled in DMSO following the protocol described for the lyophilized plantain powder (*vide supra).* The resulting supernatants were stored at −20 °C prior to analyses. These were subjected to analyses of total titratable acidity (% TTA) as percent malic acid equivalent (*33*) using standard NaOH (0.1 N), pH (using pH meter, Cyberscan PC510m, M/s Eutech Instruments Pvt. Ltd., Singapore City, Singapore) (*2*), total insoluble solids (gravimetrically), and total soluble solids as °Brix (*21*) (using OptiDuo Refractometer, M/s Bellingham + Stanley Ltd., Tunbridge Wells, UK).

Antioxidant properties

Since CCS was chiefly composed of sugar alcohols and contained no additional source of natural antioxidants, the evaluation of antioxidant properties was carried out solely for PCS.

Assessment of the antioxidant potency of PCS

The supernatant obtained from the PCS sample (*vide supra*) was also used for the determination of reducing power as μg BHT equivalent/g D.M. (*38*); TPC as mg gallic acid equivalent (mg GAE/100 g D.M.) by the Folin-Ciocalteu method (*39*); DPPH radical scavenging activity as IC_{50} values (mg/mL); and FRAP value as mM $FeSO_4$/100 g D.M. (*2*), by standard spectrophotometric methods using UV-Vis double beam spectrophotometer (Halo DB-20, M/s Dynamica Scientific Ltd., Newport Pagnell, UK).

Determination of L-tryptophan, serotonin, and melatonin by HPLC-photo diode array (PDA) analysis using QuEChERS-solid phase extraction (SPE) extract of PCS

For the extraction of L-tryptophan, serotonin, and melatonin from PCS, the QuEChERS-SPE method was followed according to the procedure elaborated in our previous publication (*2*). Candy (15 g) was crushed using a mixer grinder (HL 1618, M/s Philips India Limited, Chennai, India) and

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placed in a clean centrifuge tube, to which 1 % acetic acid in acetonitrile solution and the contents ($MgSO_4$ and NaCl) of an SPE AOAC packet were added. After thorough mixing using a vortex (iSwix VT, M/s Neuation Technologies Pvt. Ltd, Gujarat, India), the tube was centrifuged at 1500×*g* for 1 min in a centrifuge (R-8C, M/s Remi, Mumbai, India). One mL of supernatant was withdrawn and added to the dispersive-SPE (dSPE) sample cleanup tube [containing primary secondary amine, C18 sorbent (trifunctionally bonded C18 silica), graphitized carbon black, and $MgSO_4$] and thoroughly mixed. The dSPE sample cleanup tubes were again subjected to centrifugation at 1207×*g* for 5 min using a microspin centrifuge (TC- 4815D, M/s Eltek, Haryana, India). The supernatant, *i.e.*, the extracted sample was filtered using a micro syringe filter (0.22 µm nylon) and stored in an amber-coloured glass vial at −20 °C for further analyses. HPLC-PDA detection of L-tryptophan, serotonin, and melatonin was conducted in accordance with our laboratory developed procedure (*40*) employing a C18 reversed phase HPLC system (LC-Net-2/ADC, PU-4180HPLC pump, DG-4000–04 degasser, MD-4015 detector, M/s Jasco, Tokyo, Japan). HPLC-grade methanol and 1 % HPLC-grade acetic acid in HPLC-grade water were used as mobile phase solvents in gradient mode, each at a flow rate of 1 mL/min. A PDA detector with a deuterium (D2) lamp set at 280 nm was employed for continuous monitoring of the eluents. The peaks of L-tryptophan, serotonin, and melatonin were identified based on the retention time of their corresponding sigma standards.

Assessment of antioxidant synergism among L-tryptophan, serotonin, and melatonin in PCS

All naturally occurring antioxidants in any food are always present as a synergistic consortium (*22*). Once isolated, antioxidants can act either synergistically or antagonistically (*41*). Therefore, it is of utmost importance to achieve antioxidant harmony without perturbing the natural synergy of foods if the benefits of antioxidants *in vivo* are to be experienced. Preservation of the naturally occurring synergism among the three antioxidants mentioned in PCS was one of the prime objectives of this study to ensure that the designer candy is an antioxidant-rich product. To evaluate the 'food synergy' in the developed candy, the synergism among L-tryptophan, serotonin, and melatonin was assessed *in vitro* by determining the synergistic effect (SE) value using the DPPH radical scavenging capacity of pure chemical standards of L-tryptophan, serotonin, and melatonin separately in varying concentrations similar to those present in PCS and of a mixture comprising of the above antioxidants in the same concentrations (as found by HPLC-PDA analysis of PCS). The experimental scavenging capacity (% ESC), theoretical scavenging capacity (% TSC), and SE value were calculated in accordance with the method described by Chakraborty and Bhattacharjee (*21*). An SE value greater than 1 indicates the preservation of natural synergism amongst these three biomolecules.

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Packaging and shelf-life study of packaged candies

For the commercialization of the newly developed candy, it is important to know its storage properties under packaged conditions. For this purpose, two flexible packaging laminates, commonly used for commercial candy packaging, were provided and tested by the Indian Institute of Packaging, Kolkata, India as used for the study. The flexible 2 and 3-ply packaging laminates in the form of sheets of uniform dimensions consisted of 12 µ non-heat sealable biaxially oriented polypropylene (NHS BOPP)/8 µ white pigmented extrusion coated polyethylene/15 µ metallized biaxially oriented polypropylene (MET BOPP) (NHS BOPP/MET BOPP); and 12 µ polyethylene terephthalate (PET)/8 µ white pigmented extrusion coated polyethylene/12 µ metallized polyethylene terephthalate (MET PET)/20 µ polyethylene (PE) (PET/MET PET/PE). Their characteristic chemical and mechanical properties [*i.e.*, thickness, grammage, water vapour transmission rate (WVTR), oxygen transmission rate (OTR), and tensile strength] were evaluated using standard methods (*42*) and are presented in Table 1.

Selection of the best-packaging laminate (wrapping material) based on the microbiological safety of the packaged candy

To select the best flexible packaging laminate, the microbial load, and sensory attributes of the candy samples (CCS and PCS) packaged in the said laminates were evaluated over a period of three months. Freshly prepared batches (each batch consisted of 72 CCS samples and 72 PCS samples (batch size decided based on sample amounts needed for regular physicochemical analysis) of candies were immediately packaged inside foil wrappers of composition NHS BOPP/MET BOPP laminate (36 CCS and 36 PCS) and PET/MET PET/PE laminate (36 CCS and 36 PCS), leaving negligible void space inside the package. It was then sealed with a heat sealer (Delta Seal V2, M/s Sevana Traders and Services Pvt. Ltd., Cochin, India). The packaged candies were stored under ambient conditions [(27±2) °C, (80±2) % RH]. Three randomly selected samples were subjected to microbial and sensory evaluation on the first day of each week, following the methods described above.

Shelf-life assessment of candy packaged in the selected wrapper

The best packaging laminate for the newly designed candy was selected in terms of microbial safety and organoleptic acceptability of the packaged candies. A comprehensive shelf-life study was then conducted with CCS and PCS. Each batch comprised of 40 candy samples (reasons for choice

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of the said batch size has been explained *vide supra*) wrapped inside the best packaging laminate. Alterations in microbial load, sensory attributes, and physicochemical properties [moisture content, alterations in CIE colour values, texture profile analysis (TPA)]; antioxidant efficacy (TPC, DPPH radical scavenging activity, FRAP values); L-tryptophan, serotonin, and melatonin content, and antioxidant synergy, were assessed throughout the storage period in accordance with the previously described methods (*vide supra*). Each week at a regular interval, three candies were randomly withdrawn for analyses. For candies approaching the end of their shelf lives, microbiological analysis was performed on all days of storage.

Statistical analysis

All the data reported are the mean value±S.D. of the data obtained from three candy samples. Student's t-test to evaluate the individual and interactive effects of two variables and Duncan's multiple range test to determine significant differences among means were performed using IBM SPSS Statistics software version 26 (M/s IBM, New York, NY, USA) (*43*). A value of $p \leq 0.05$ was considered statistically significant to establish differences in all tests.

RESULTS AND DISCUSSION

Antioxidant properties of raw plantain and its lyophilized powder

The plantains procured for this study had a relatively high TPC value (573.17 mg GAE/100 g D.M.), DPPH radical scavenging activity (IC_{50} value 5.59 mg/mL), FRAP value (3438.12 mM $FeSO_4$/100 g D.M.), and L-tryptophan, serotonin, and melatonin contents (102.45 µg/g, 4.56 µg/g, and 2.08 µg/g, respectively). The lyophilized plantain powder also exhibited a remarkable TPC value (865.48 mg GAE/100 g D.M.), DPPH radical scavenging activity (IC_{50} value 0.56 mg/mL) and FRAP value (5528.95 mM $FeSO_4$/100 g D.M.), an increase of 50.99 %, 89.96 % and 60.81 %, respectively, over the raw plantains, possibly owing to the concentration effect. A similar increase in antioxidant potency was also reported by Dal-B´o and Freire (*25*) in lyophilized avocado pulp powder compared to that of fresh avocado pulp. As a consequence of the enhanced antioxidant activity, homogeneity, and low moisture content (<6 %) of the lyophilized plantain powder vis-à-vis those of the fresh plantains, the powder was utilized in the development of the designer candy instead of the fresh plantain pulp to achieve an acceptable texture as well as low water activity in the candy (determined from preliminary trials).

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Weights and dimensions of CCS and PCS

The weight of the CCS from the aforementioned plantains was 20±2 g, whereas that of the PCS was 24±2 g. The dimensions of either type of candy was as follows: length: 4 cm; width: 2 cm; and height: 2 cm. From 100 g of raw green plantains, 4 pieces of PCS with the above dimensions were obtained (Fig. S1).

Safety aspects of CCS and PCS

Safety is of paramount importance for all consumable food products. Any microbial or heavy metal contamination beyond safe limits can lead to mild to severe health hazards and can even be fatal to the consumers. The total bacterial and yeast/mould counts (no growth was detected on the first day in either sample) of CCS and PCS were within the safe consumption limits as per the guidelines of FSSAI (*44*) which state that the bacterial and fungal counts in thermally processed (except by pasteurization, which is conducted at a temperature less than 100 °C) fruit and vegetable products should not exceed 1000 and 100 CFU/g of the food product, respectively.

The EDX analyses of CCS and PCS (Fig. 1a and 1b, respectively) showed that both candy samples were free of heavy metal contaminants such as Pb, Ti, Hg, Ni, As, Si, and Mo. However, Cu was detected in the PCS sample since plantain is reportedly known to be a source of this micronutrient (*45*).

The findings of the microbiological and EDX analyses assured that the formulated candy was completely safe for human consumption and therefore could be subjected to sensory evaluation.

Sensory and physicochemical characteristics of CCS and PCS

Consumer acceptance is the most important criterion for a new food product. The hedonic scores from the panel responses for CCS and PCS are presented as radar plots (Fig. 1c and 1d, respectively). It was evident that PCS was well accepted by the panellists owing to its uniform dark brown colour (L^*: 48.5; a^*: 5.1; b^*: 11.6; c^*: 12.7; h: 66.5), rich mouthfeel, pleasant aroma, moderately hard texture, and moderate sweetness. The typical characteristic flavour of green plantains in the processed candy was also moderately appreciated. CCS mimicked the dark caramel-like colour (L^*: 51; a^*: 3.9; b^*: 10.8; c^*: 11.5; h: 70.1) (lighter than PCS) and texture of ‘hard boiled candy’, tasted extremely sweet, and was sticky in the mouth, while PCS was less hard (more brittle) and not sticky. The caramel-like appearance of the newly developed candies was in consonance with the findings of

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Ronda *et al.* (*46*) who reported that the use of polyols (sorbitol and mannitol) in the cake preparation darkened the crust compared to the cakes prepared with sucrose, primarily owing to the classic Maillard reaction. Additionally, the presence of brown-coloured lyophilized plantain powder also contributed to the formation of a darker colour of PCS compared to CCS. The findings of the TPA [PCS showed hardness(g): (6803.52±391); adhesiveness (g.s): (−238.80±15.83); springiness: (0.14±0.01); cohesiveness: (0.41±0.02); gumminess: (196.54±32.86); and chewiness: (95.77±2.33)] corroborated well with its sensory attributes. CCS exhibited significantly ($p<0.05$) higher hardness (10643.08±893) g, adhesiveness (−3033.96±147) g.s, springiness (0.97±0.03), cohesiveness (0.18±0.01), gumminess (1899.09±156) and chewiness (1847.51±124) vis-à-vis that of PCS. However, another sugar-free nutraceutical hard candy comprised of isomalt, maltitol syrup, xylitol, and extract of melon berry (*20*) had a harder (in KgF) texture than the candy developed in the present study. Therefore, the newly formulated designer candy was labelled as ‘semi-hard’.

The FE-SEM image of CCS (Fig. 1e) revealed a continuous, uniform, compact, and less porous structure with a smooth surface, which corresponded well with the textural attributes of the sugar-made hard candies (*47*). In contrast, PCS (Fig. 1f), exhibited a grainy, non-homogeneous, and discontinuous (with voids) microstructure, conferred by the complex composition of plantains (especially the soluble and insoluble fibre content). There is a lack of similar data on sugar-free hard or semi-hard candies to compare these findings.

Fig. 1g and 1h present the XRD graphs of CCS and PCS, respectively. Both graphs revealed few nanocrystalline structures on a predominantly amorphous base. Thus, the candies were inherently amorphous in nature since they were sugar-cum-fat-free candies and free of classical defects such as graininess and crystals. However, they were opaque, and owing to the presence of insoluble plantain constituents, little graininess (although the same were not the classical ‘blooms’) was present in PCS. CCS was thus categorized as ‘grained hard candy’ (*24*) and PCS as a ‘grained semi-hard candy’, the same being relatively more ‘brittle’ than CCS. These data could not be compared or validated with literature reports since, to the best of the knowledge of the authors, there is no existing XRD data for sugar-free hard candy.

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Thermal stability of CCS and PCS

The TGA graphs of both CCS and PCS could be classified under 'multi-stage decomposition' type (3-stage decomposition), wherein the first decomposition was observed at 125 °C for either candy (Fig. 1i and 1j), indicating that both candies were thermostable up to very high temperatures.

From the DSC thermograms (Fig. 1k), for CCS, the onset of glass transition temperature (T_g) was found to be −50 °C, the T_g midpoint was at −38 °C, and the T_g endpoint was at −28 °C during both the first and second phases of heating. On the other hand, T_g onset, midpoint, and endpoint for PCS (Fig. 1l) were −35 °C, −30 °C, and −25 °C, respectively, during the first heating phase, and −30 °C, −25 °C, and −20 °C, respectively, during the second heating phase. The higher T_g value (mid-point) for CCS affirmed the hard and brittle texture of the candy during mastication; whereas the lower T_g value of PCS was in consonance with the mouthfeel (as assessed by the sensory panel) of the hard-cum-brittle candy, which gradually softened with mastication. The melting peak of PCS was observed to be in the relatively wide range of 94-105 °C, implying slow melting in the mouth, while that of CCS was in the narrower range of 90-94 °C during the first heating phase. During the second heating, CCS exhibited no change in the T_g values; however, there was a slight shift in the T_g and heat flow (w/g) curve for PCS, possibly due to the presence of polysaccharides, dietary fibres, and other complex compounds therein.

The absence of peaks indicated absence of crystallinity of the candies (*36*) which corroborated well with the findings of the XRD data (*vide supra)*. This was owing to the replacement of sucrose as the doctoring agent by sugar alcohols, resulting in the absence of sugar blooms in the new designer candies. These findings of TGA, DSC analysis, and sensory evaluation were similar to those reported by Wang (*36*) who had conducted extensive research on sugar-boiled hard candy.

The a_w value and proximate composition of CCS and PCS

Although CCS were sugar-free, it had an a_w value of 0.43, which was similar to that of conventional hard sugar candies (0.25-0.40). The a_w value of PCS was 0.57, which was similar to that of soft candies (0.46-0.60) (*48*). These results corroborated well with that of the texture analysis data on hardness, based on which PCS was categorised as 'semi-hard'. PCS had a significantly higher ($p<0.05$) a_w value (Table 2) than CCS, since the lyophilized plantain powder (non-packaged) was hygroscopic (*49*). The higher a_w value of PCS would pose a challenge for its shelf-life extension, which was successfully averted by appropriate packaging of the candies (*vide infra*).

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The proximate analyses of CCS and PCS (Table 2) revealed that PCS had significantly higher % moisture ($p<0.001$), % ash ($p<0.001$), % crude fibre ($p<0.001$), and % protein ($p<0.001$) content and significantly lower % fat ($p<0.001$) and total carbohydrates ($p<0.001$) content, which rendered PCS nutritionally richer than CCS.

Percentage TTA, pH, and soluble and insoluble solids of CCS and PCS

CCS showed an alkaline pH of 8.1, whereas PCS was acidic (pH=4.3). A significantly higher ($p<0.05$) percentage of TTA (0.12 in terms of % malic acid in PCS compared to 0.09 in terms of % malic acid in CCS) and lower pH ($p<0.05$) were observed for PCS, which could be beneficial for the extension of its shelf-life. These findings were in consonance with the findings documented by Supriyanto *et al.* (*50*), who have reported the pH of sucrose-free (with xylitol and glucose syrup) hard candy prepared with Javanese long pepper extract to be 4.3. CCS consisted mostly of soluble solids (82 %), whereas the crude fibre content of the plantains (mostly from their peels) contributed to significantly higher ($p<0.05$) insoluble solids (36.26 %) in PCS. The higher insoluble solids as well as crude fibre contents in PCS justified the increased weight of PCS, although it was dimensionally similar to CCS.

Antioxidant properties of PCS

PCS had a considerable amount of reducing power (11.30 µg BHT/g D.M.), TPC value (679.28 mg GAE/100 g D.M.), DPPH radical scavenging activity (IC_{50} value 6.23 mg/mL), and FRAP value (2565.96 mM $FeSO_4$/100 g D.M.), although the processing losses were 18.5 %, 11.6 %, and 25.4 %, respectively, compared to the values obtained for raw plantains. The same were 21.51 %, 1012.5 % and 53.59 %, respectively, compared to the values obtained for the lyophilized plantain powder. The TPC value, DPPH radical scavenging activity, and FRAP value were significantly ($p<0.05$) reduced in PCS compared to the raw and lyophilized plantains. These findings corroborated well with those reported by Dadwal *et al.* (*51*), who found significant decreases ($p<0.05$) in TPC, DPPH, and FRAP values in candies prepared from bamboo shoots compared to the corresponding values of the same for fresh bamboo shoots. These differences could be attributed to the treatment/processing that the ingredients underwent during candy production. Since phenolic content is strongly related to antioxidant activity, the reduction of TPC value during candy preparation also led to a reduction in DPPH radical scavenging activity and FRAP values in the case of PCS (*51*).

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The contents of L-tryptophan, serotonin, and melatonin in PCS were 4.54 µg/g, 1.83 µg/g, and 1.23 µg/g, respectively, with a SE value>1, indicating that the natural synergism among these three biomolecules is maintained in the processed candies. This finding is in consonance with the findings of Chakraborty and Bhattacharjee (*21*) who reported on the preservation of antioxidant synergism in processed food products in a nutraceutical beverage formulated using an ultrasonication-assisted solvent extract of mustard seeds with lemon and citric acid.

Best packaging laminate for the extension of the shelf-lives of CCS and PCS

During the storage period, the growth of microbes in the candies packaged in the two laminates was restricted for the first three weeks. However, on day 28 of the storage period, NHS BOPP/MET BOPP packaged (both CCS and PCS) candies had considerable bacterial and yeast/mould counts (data not shown) that were beyond the safe limit of consumption as per the guidelines of FSSAI (*44*). The NHS BOPP/MET BOPP packaged candies were therefore discarded on day 28. This study affirmed the suitability of PET/MET PET/PE as a packaging laminate for CCS and PCS.

Based on the properties of the foil wrapper (Table 1), it was found that the values of the mechanical properties (thickness, grammage, and tensile strength) were significantly higher ($p<0.001$) and the OTR value was significantly lower ($p<0.001$) for the PET/MET PET/PE (Fig. S1) laminate, vis-à-vis that of the NHS BOPP/MET BOPP laminate (Fig. S1). The high values for thickness, grammage, and tensile strength values of the three-ply PET/MET PET/PE flexible packaging laminate indicated improved mechanical strength, while a low OTR value indicated its good barrier property against oxygen and thus a possible prevention of oxidative degradation of the antioxidants present in the candy. An insignificant difference between the WVTR values suggested similar permeability of the packaging films to atmospheric water vapour. The low OTR and WVTR values of the PET/MET PET/PE laminates aided in the prevention of moisture loss in candies and thus averted the candies from becoming soft (and therefore sticky). These properties of the wrapper foils proved to be beneficial for the maintenance of the organoleptic wholesomeness of the candy and rendered them safe in terms of microbiological bioburden.

The CCS packaged in PET/MET PET/PE were microbiologically safe for up to 56 days, while the PCS was safe for consumption for up to 63 days (Table S2). Based on these findings, the storage study period for the candy packaged in the best packaging laminate was ascertained to be 63 days post-packaging in PET/MET PET/PE laminate. A similar packaging laminate (PET/MET PET/PE) is

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reportedly used for commercial packaging of powdered spice mixes (for export) imparting it a shelf-life of 9-12 months under normal storage conditions (*52*).

Shelf-life of 3-ply PET/MET PET/PE-packaged CCS and PCS

Alterations in sensory attributes during storage

The response scores of sensory attributes of CCS and PCS during storage are presented as radar plots in Fig. 2a and 2b, respectively. On day 56, PCS was disapprovingly dry and brittle to the sensory panel, and CCS was also sensorially unacceptable owing to its increased stickiness and hardness on day 49 of storage. A reduction in the sensory attributes has been reported for *Basella alba* (Malabar spinach) extract-incorporated hard candies when wrapped in a 2-ply laminate of non-stick paper or in an aluminium foil and stored in airtight containers under ambient conditions (*53*). The authors attributed the decreases in the sensory scores to deteriorating changes in hardness and adhesiveness, in consonance with the findings of the present study. Henceforth, in this study, the shelf-life of PET/MET PET/PE-packaged PCS was considered to be 56 days, although the same was microbiologically safe up to 63 days.

Alterations in physicochemical properties during storage

For PCS and CCS, significant ($p<0.001$) declinations in percent moisture content during storage (Fig. 2c) were observed from day 0 until the end of their respective shelf-lives, possibly owing to the migration of moisture through the wrapper(s) into the immediate environment (*48*). Moreover, significant differences ($p<0.001$) were also observed between the percent moisture contents of CCS and PCS on each assessment day. Significant ($p<0.001$) decreases in texture parameters (such as hardness, adhesiveness, springiness, cohesiveness, gumminess, and chewiness) occurred steadily throughout the storage period for both CCS and PCS; although alterations of the same were greater in PCS vis-à-vis CCS (Table 3). Very little changes ($p<0.001$) occurred for L^*, a^*, and b^* values (Fig. 2d and 2e) for both CCS and PCS during storage.

The decline in moisture content during storage of PCS explained the occurrence of dryness, brittleness, and incoherency in the candy structure of the same on the last day of storage. Candies tend to lose moisture to the environment as moisture migrates out of the candy and through the package, making them harder (*48, 53*). In the present study, PCS became brittle instead of being harder, probably owing to the presence of void spaces within, as has been confirmed from its

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microstructure analysis (Fig. 1f). Loss of % moisture also led to loss of adhesiveness (*48*), thereby causing losses in springiness, cohesiveness, gumminess, and chewiness. Changes in colour parameters were possibly due to the occurrence of non-enzymatic browning (*53*) and perhaps accelerated by arabinose, an abundant glycoside present in one of the constituents of candy, *i.e.,* gum acacia (*54*). The findings of the present study are in consonance with the findings of Yan *et al.* (*53*) described earlier (*vide supra*).

Alterations in antioxidant properties and antioxidant (L-tryptophan, serotonin, and melatonin) synergy during storage

PCS showed appreciable TPC, IC_{50} value of DPPH radical scavenging activity, and FRAP value on day zero, which significantly declined with storage (Fig. 2f, 2g, and 2h) until the end of its shelf life (56 days). A similar trend of decline in TPC and antioxidant activities during storage has been reported by Seremet *et al.* (*55*) in candies containing steviol glycosides, sorbitol, inulin, psyllium, citric acid, and white tea extract. At the molecular level, L-tryptophan, serotonin, and melatonin contents decreased significantly ($p<0.001$) from their respective contents (4.54 µg/g, 1.83 µg/g, and 1.23 µg/g, of L-tryptophan, serotonin, and melatonin, respectively) on day 0 until the last day of shelf-life (3.04 µg/g, 0.21 µg/g, and 0.67 µg/g, respectively of L-tryptophan, serotonin, and melatonin) of PCS (Fig. 2i). The SE value of PCS was found to be greater than unity, signifying that the natural antioxidant synergy was not perturbed during candy processing and also remained unaffected by physicochemical changes in the candy during storage (Table 4). Thus, the plantain-based candy would be a rich source of the antioxidant triad - L-tryptophan, serotonin, and melatonin even after 56 days of storage inside a commercial foil wrapper. It is anticipated that the candy processing and packaging presented in this work would be a sustainable solution for the delivery of plantain-based antioxidant confectionery.

CONCLUSIONS

The newly developed plantain-based candy could be a promising nutraceutical confectionary, rich in three therapeutically important antioxidants, namely L-tryptophan, serotonin, and melatonin, which could be safely stored for 56 days without any physicochemical and considerable antioxidant deterioration when packaged in a 3-ply flexible (PET/MET PET/PE) laminate wrapper. It is envisaged that the same would be a vehicle for *in vivo* delivery of these biomolecules since the formulation involves a sugar-free base (of sorbitol and mannitol) and minimal food additives (except for a

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thickener, a desiccant, and an anti-fungal agent) with appreciable shelf-life. This semi-hard candy could be a novel potential antioxidant-rich food supplement, especially for the geriatric population.

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CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

SUPPLEMENTARY MATERIALS

Supplementary materials are available at: www.ftb.com.hr.

AUTHORS' CONTRIBUTION

P. Sarkar performed the analyses, collected, analysed, and interpreted data, drafted the article, and did critical revision. P. Bhattacharjee conceptualized and designed the work, analysed the data, interpreted the findings, supervised, and validated the work, and finally approved the version to be published. B. Das analysed the data of packaging materials.

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Table 1. Mechanical and chemical properties of different packaging laminates

Test	PET/MET PET/PE	BOPP/MET BOPP
Thickness/μm	(52±0.54)[a]	(40±0.97)[b]
Grammage (g/m^2)	(59±0.42)[a]	(35±0.75)[b]
Water vapour transmission rate [g/m^2/day at (38±1) °C and (90±2) % RH]	(0.127±0.05)[a]	(0.124±0.08)[a]
Oxygen transmission rate (cc/(m^2/24 h/atm) at 25 °C)	(1.08±0.05)[a]	(56.67±1.56)[b]
Tensile strength (kg/cm^2)	Machine direction: (1028.00±2.56)[a]	Machine direction: (725±2.58)[b]
	Transverse direction: (875±1.86)[a]	Transverse direction: (645±1.46)[b]

Data are mean value±S.D. of three samples
Different letters in a row indicate significant differences at $p<0.05$ level

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PET/MET PET/PE=12 micron polyethylene terephthalate (PET)/ 8W. ext. coting/12 micron metalized polyester (MET PET)/20 micron polyethylene (PE)
BOPP/MET BOPP=12 micron NHS biaxially oriented polypropylene (BOPP)/8W. ext. coating/15 micron metallized bi-orientated polypropylene (MET BOPP)

Table 2. Proximate composition and water activity of control candy sample and plantain-candy sample

Analysis parameters	Composition (% on fresh mass basis)	
	Control candy sample	Plantain-candy sample
Moisture	(3.65±0.84)^{a}	(14.74±1.53)^{b}
Fat	(0.66±0.03)^{a}	(0.16±0.06)^{b}
Protein	(0.29±0.02)^{a}	(2.6±0.35)^{b}
Crude fibre	0	(2.16±0.73)^{a}
Ash	(0.90±0.02)^{a}	(4.17±0.88)^{b}
Carbohydrate (by difference)	(94.51±1.43)^{a}	(76.17±2.85)^{b}
Water activity	(0.43±0.01)^{a}	(0.57±0.02)^{b}

Data are mean value±S.D. of three samples of each set
Different letters in a row indicate significant differences at $p<0.05$ level

Table 3. Texture profile of control candy sample and plantain-candy sample during storage

t/day	Hardness (g)	Adhesiveness (g·s)	Springiness	Cohesiveness	Gumminess	Chewiness
			Control candy sample			
0	(10643.08 ±893)^{a}	(-3033.96±147)^{a}	(0.97±0.03)^{a}	(0.18±0.01)^{a}	(1899.09±156)^{a}	(1847.51±124)^{a}
7	(10653.25 ±683)^{b}	(-3075.25±285)^{b}	(0.99±0.03)^{b}	(0.19±0.02)^{b}	(1893.48±135)^{a}	(1839.93±131)^{b}
14	(10514.48 ±844)^{c}	(-3012.23±216)^{c}	(0.97±0.05)^{c}	(0.18±0.01)^{c}	(1886.75±128)^{b}	(1824.03±103)^{c}
21	(10541.88 ±580)^{d}	(-2986.85±194)^{d}	(0.95±0.02)^{d}	(0.17±0.02)^{c}	(1872.56±117)^{b}	(1798.28±116)^{d}
28	(10442.26 ±719)^{e}	(-2964.47±201)^{e}	(0.92±0.01)^{e}	(0.17±0.02)^{d}	(1856.37±123)^{c}	(1782.47±119)^{e}
35	(10475.84 ±904)^{f}	(-2993.26±178)^{f}	(0.93±0.06)^{d}	(0.17±0.02)^{df}	(1839.25±169)^{d}	(1777.24±136)^{f}

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42	(10344.31 ±729)^{g}	(-2955.19±255)^{g}	(0.91±0.06)^{f}	(0.16±0.01)^{c}	(1823.07±105)^{d}	(1763.07±148)^{g}
49	(10243.24 ±812)^{h}	(-2913.43±242)^{h}	(0.90±0.08)^{g}	(0.19±0.02)^{c}	(1815.65±122)^{c}	(1758.25±152)^{h}
56	(10181.10 ±914)^{i}	(-2879.37±268)^{d}	(0.90±0.03)^{g}	(0.16±0.02)^{f}	(1804.36±108)^{d}	(1752.72±150)^{h}
			Plantain candy sample			
0	(6803.52 ±391)^{j}	(-238.80±15.83)^{i}	(0.14±0.01)^{h}	(0.41±0.02)^{g}	(196.54±32.86)^{e}	(95.77±2.33)^{i}
7	(6888.84 ±268)^{k}	(-242.27±17.15)^{j}	(0.18±0.02)^{i}	(0.07±0.03)^{h}	(486.12±25.92)^{e}	(85.22±4.21)^{j}
14	(6743.49 ±342)^{l}	(-175.63±10.52)^{k}	(0.16±0.02)^{j}	(0.06±0.01)^{i}	(389.75±20.86)^{f}	(64.29±1.53)^{k}
21	(6535.92 ±184)^{m}	(-115.09±9.92)^{l}	(0.11±0.01)^{k}	(0.06±0.01)^{i}	(415.98±21.05)^{f}	(46.22±2.65)^{l}
28	(6315.33 ±165)^{n}	(-125.94±11.05)^{m}	(0.15±0.01)^{l}	(0.04±0.01)^{j}	(228.55±18.84)^{g}	(33.30±2.14)^{m}
35	(6440.39 ±228)^{o}	(-130.14±4.86)^{n}	(0.13±0.01)^{k}	(0.05±0.01)^{jk}	(359.95±2.10)^{h}	(45.33±3.16)^{n}
42	(6325.23 ±271)^{p}	(-103.04±7.25)^{o}	(0.09±0.02)^{m}	(0.07±0.01)^{i}	(234.17±16.85)^{h}	(21.39±1.32)^{o}
49	(6057.72 ±193)^{q}	(-120.09±9.92)^{p}	(0.11±0.01)^{n}	(0.05±0.01)^{i}	(314.86±28.44)^{g}	(37.32±2.96)^{p}
56	(4683.31 ±158)^{r}	(-223.50±5.27)^{q}	(0.12±0.01)^{n}	(0.07±0.01)^{k}	(339.75±22.85)^{h}	(40.27±3.13)^{p}

Data are mean value±S.D. of three samples of each set
Different letters in a column indicate significant differences at $p<0.05$ level

Table 4. The *in vitro* synergistic effect of L-tryptophan, serotonin, and melatonin of plantain candy sample

Sample	Serotonin (μg/mL)	L-tryptophan (μg/mL)	Melatonin (μg/mL)	Synergistic effect
S1	1.87	0.00	0.00	-
S2	0.35	0.00	0.00	-
T1	0.00	4.54	0.00	-
T2	0.00	3.04	0.00	-
M1	0.00	0.00	1.23	-
M2	0.00	0.00	0.67	-

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Plantain candy sample (on day 0)	1.87	4.54	1.23	(1.03±0.05)[a]
Plantain candy sample (on day 56)	0.35	3.04	0.67	(1.01±0.07)[b]

Data are mean value±S.D. of three samples of each set.
Different letters in a column indicate significant differences at $p<0.05$ level

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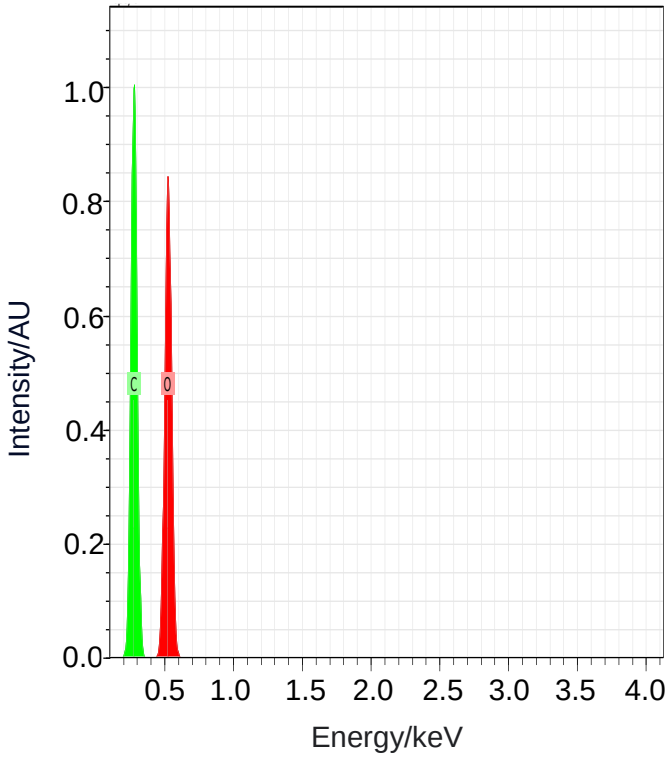

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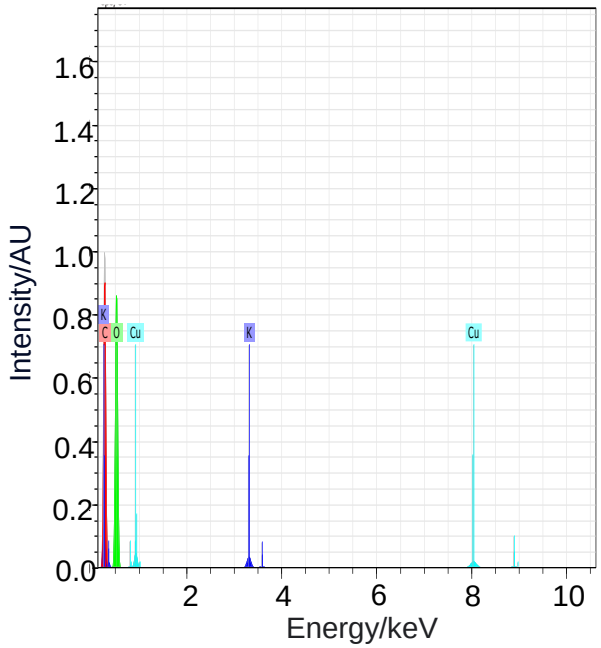

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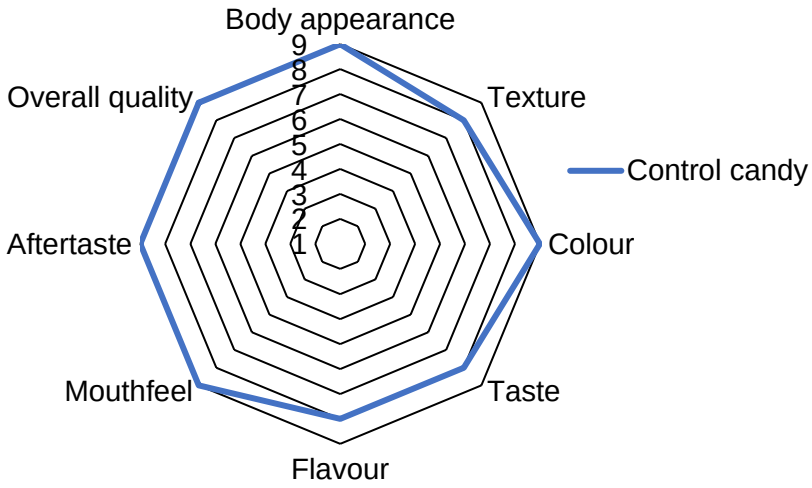

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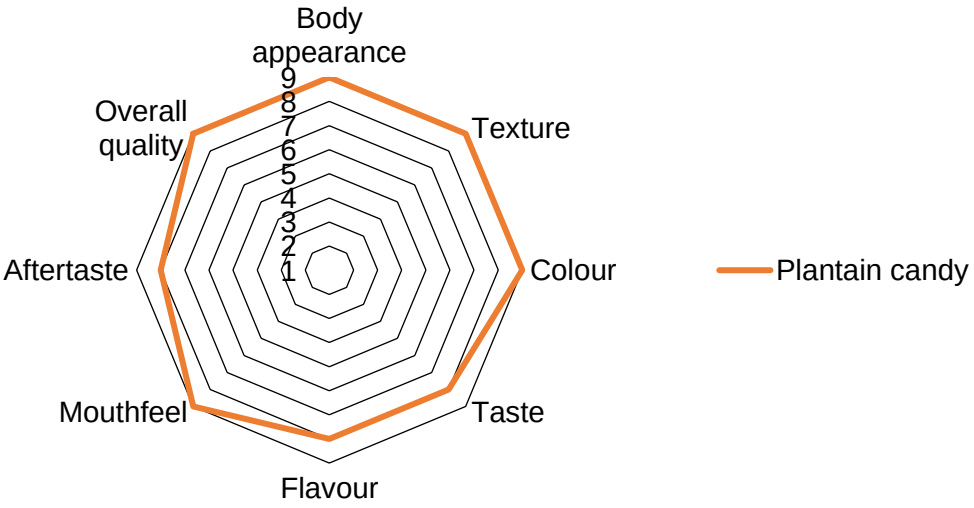

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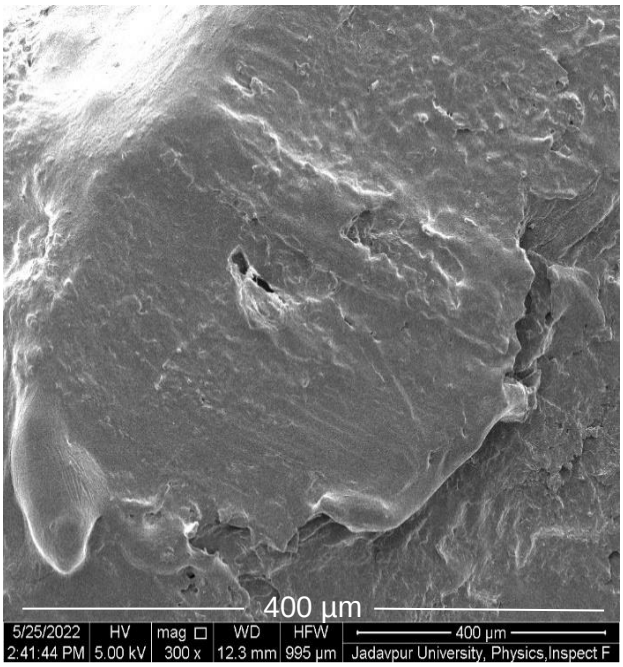

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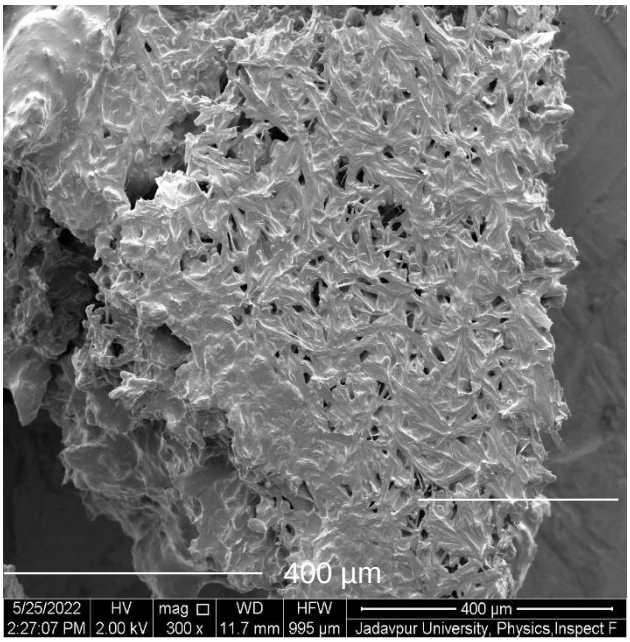

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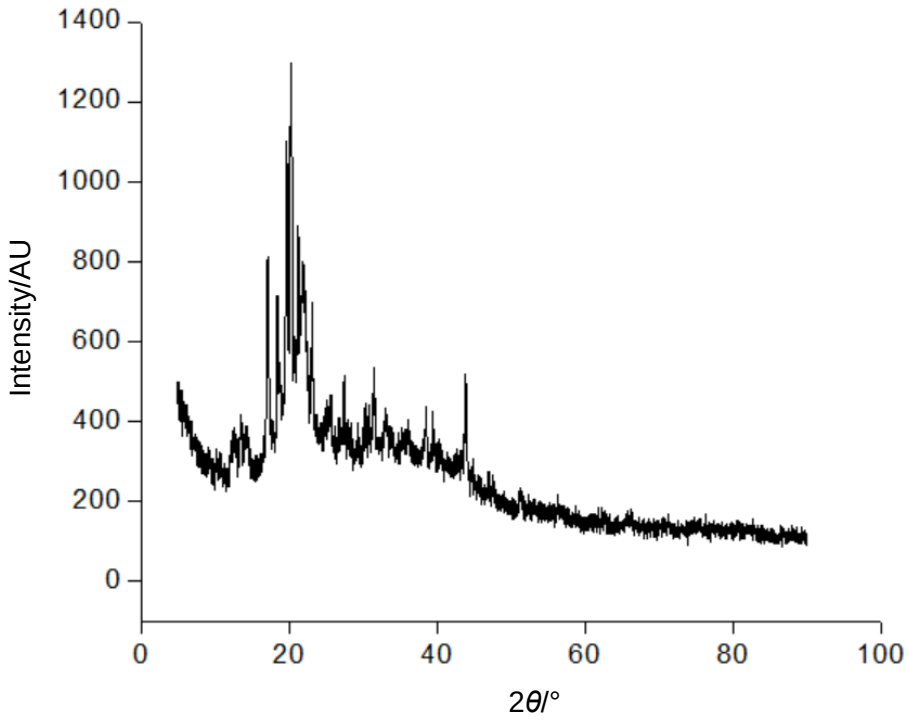

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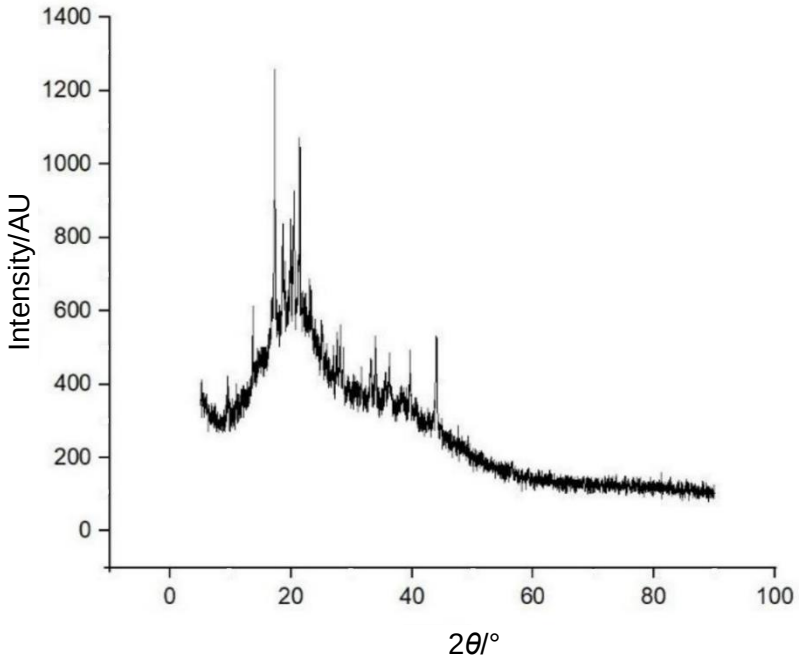

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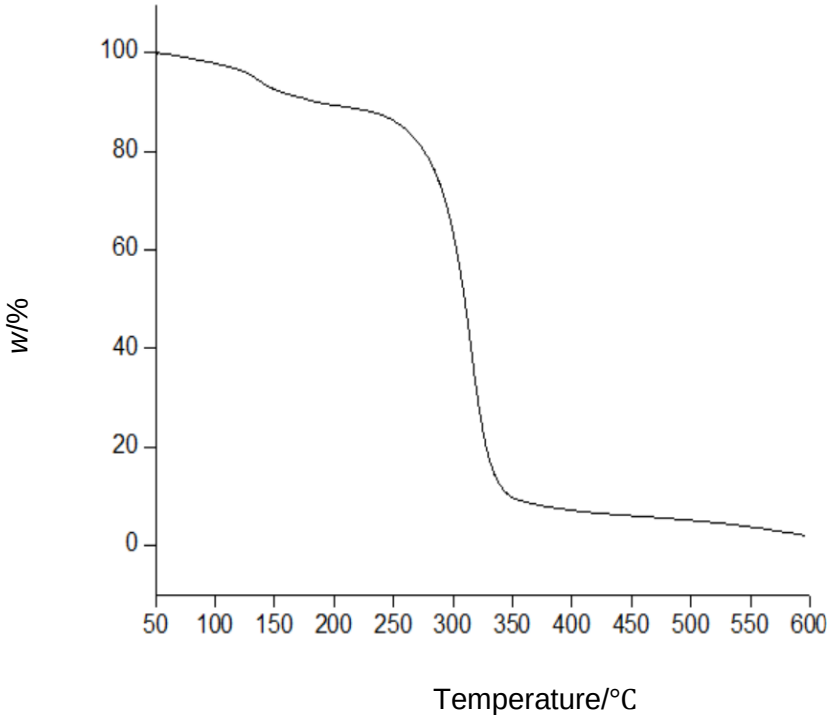

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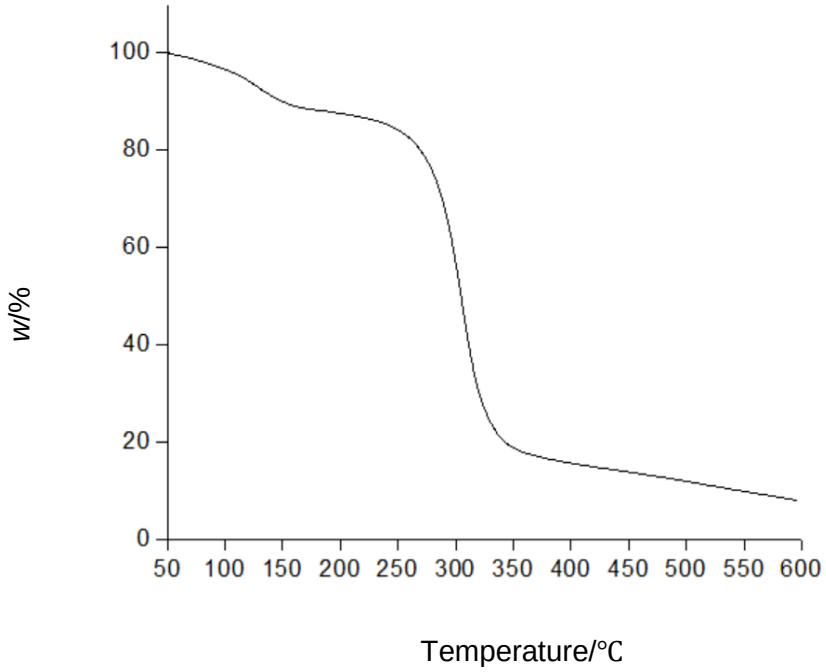

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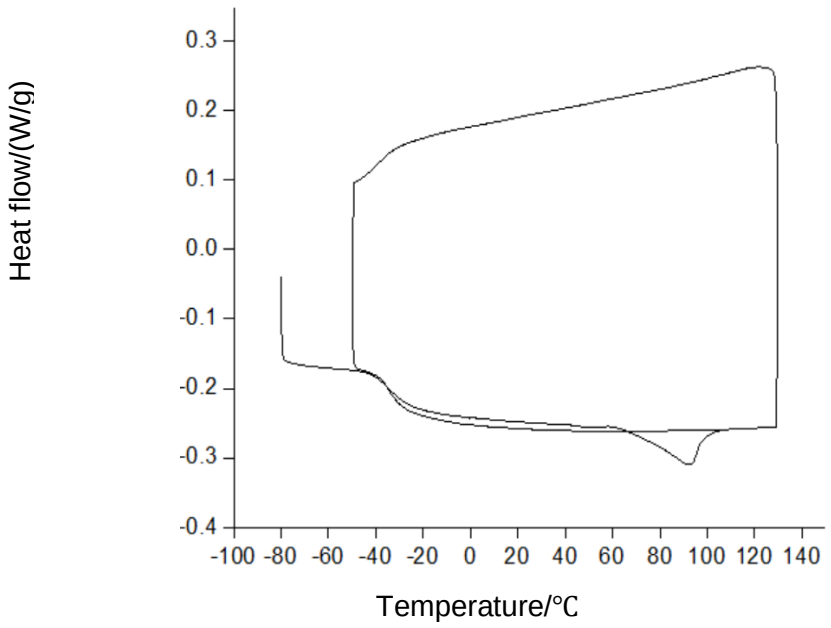

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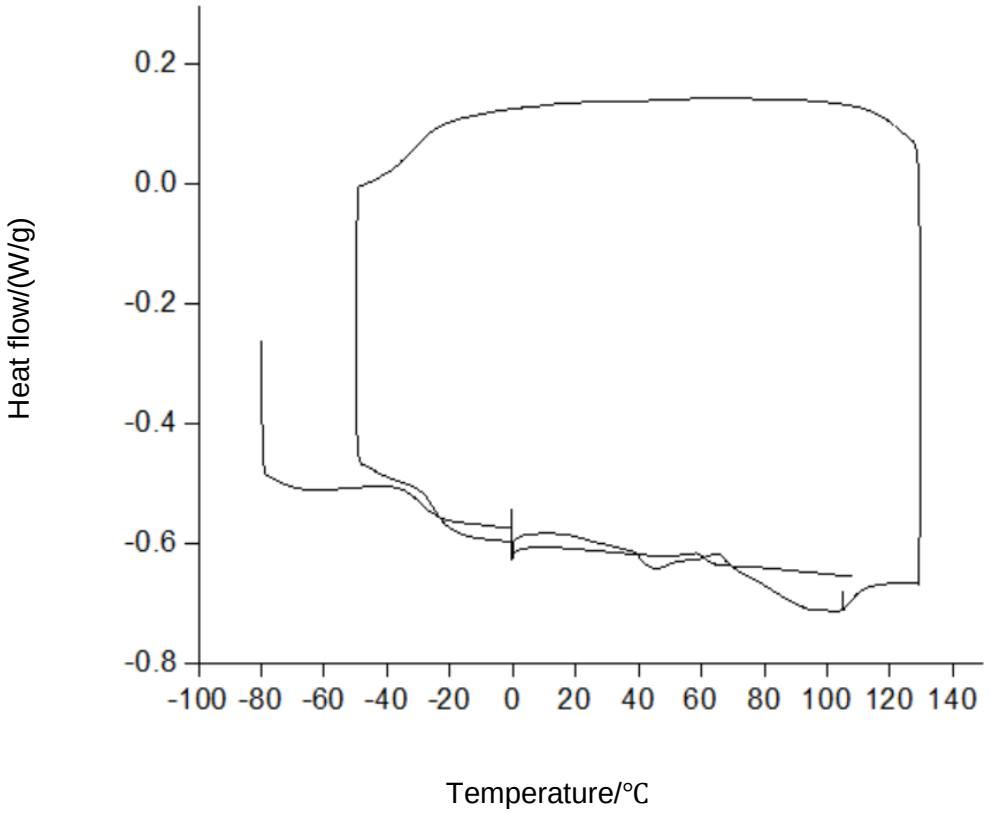

Fig. 1. Physicochemical properties of control candy sample and plantain-candy sample after formulation- energy dispersive X-ray spectra of: a) control candy sample and b) plantain candy sample. Radar plots of hedonic scores obtained by sensory analyses of: c) control candy sample and d) plantain candy sample where each value represents mean value±S.D. of three sets of experimental data. Field emission scanning electron microscopy images of: e) control candy sample and f) plantain candy sample. X-ray diffraction spectra of: g) control candy sample and h) plantain candy sample. Thermogravimetric graphs of: i) control candy sample and j) plantain candy sample. Differential scanning calorimetry thermographs of: k) control candy sample and l) plantain candy sample.

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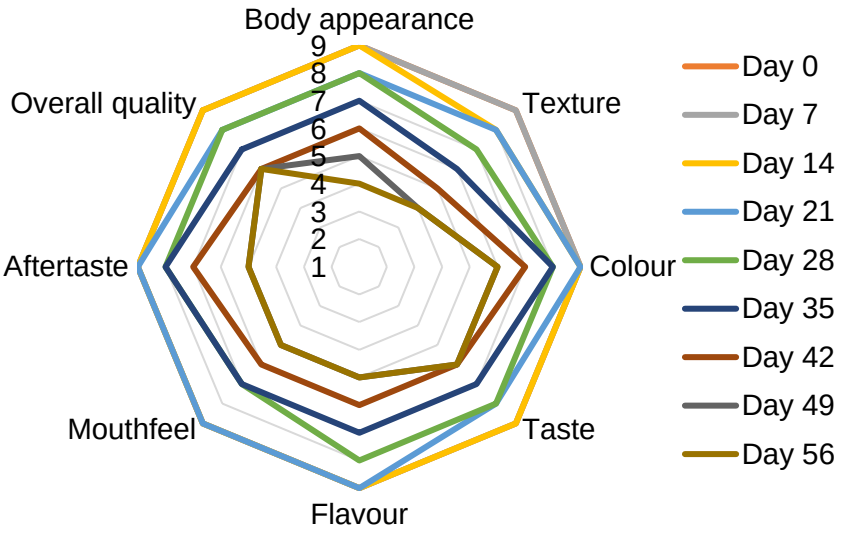

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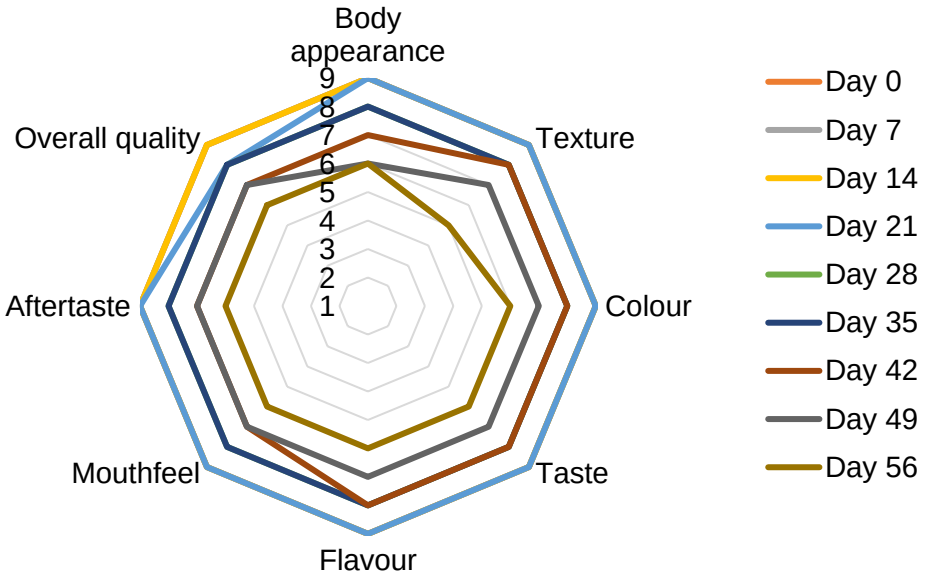

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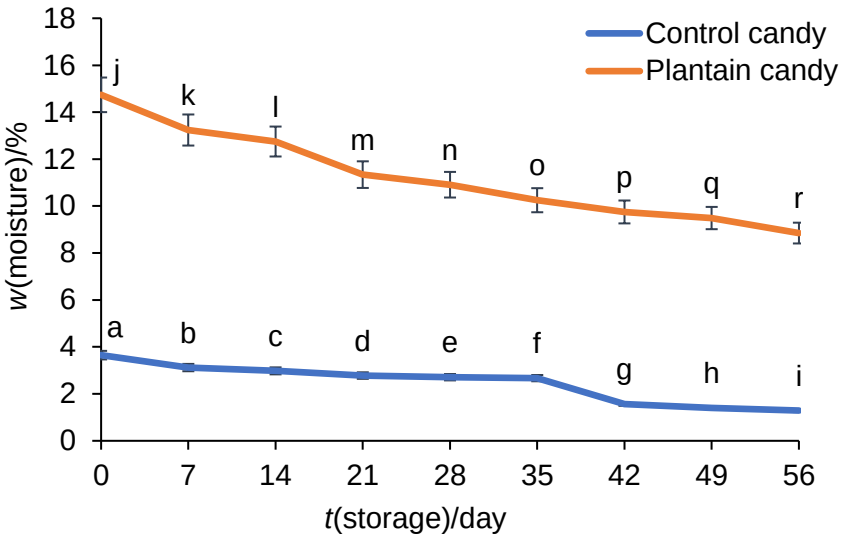

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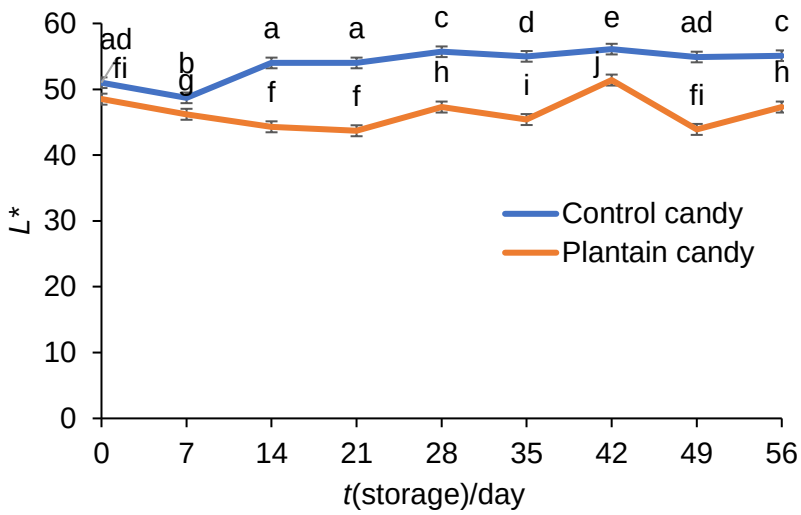

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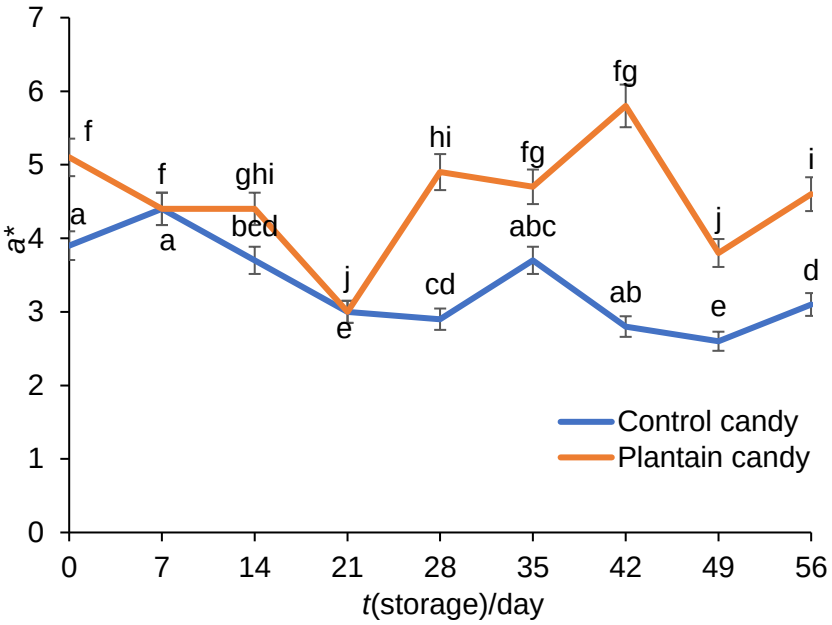

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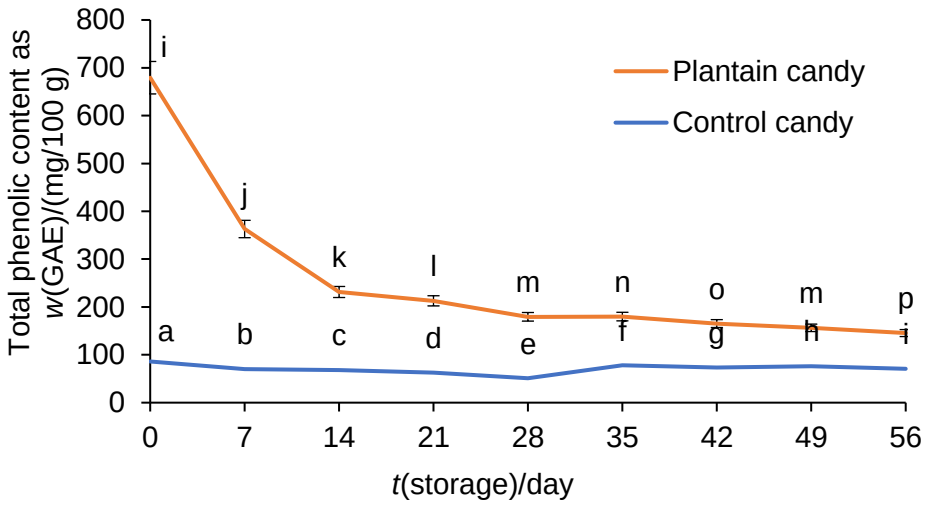

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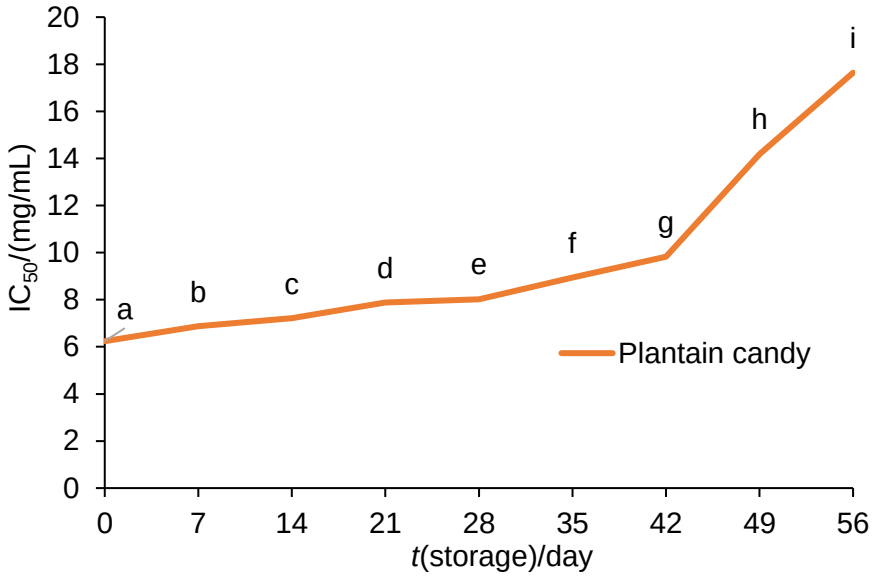
g)



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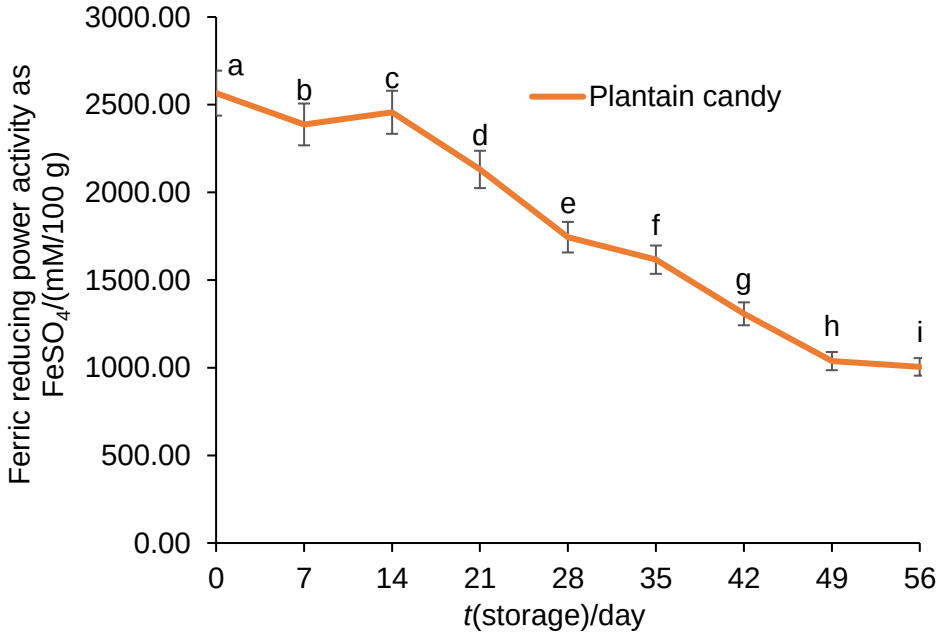

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i)

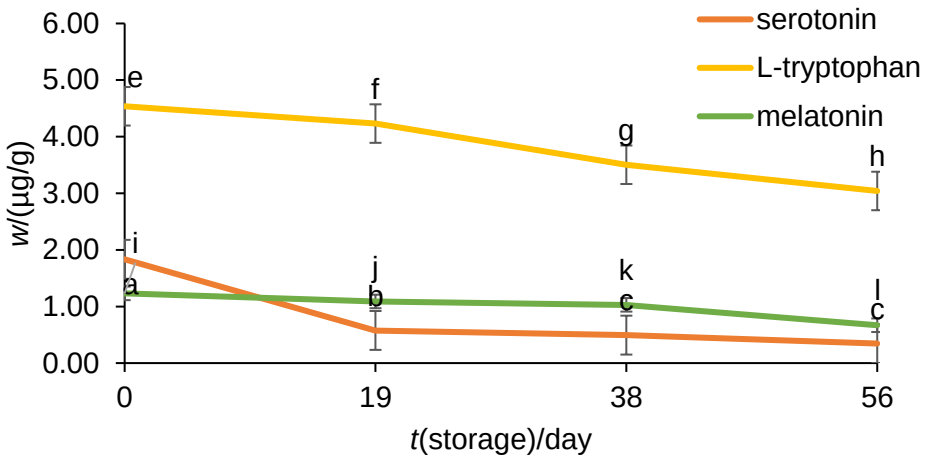

Fig. 2. Radar plots of hedonic scores obtained by sensory analyses of: a) control candy sample and b) plantain candy sample during storage at (27±2) °C, (80±2) % RH. c) alterations in % moisture content (on wet mass basis) during storage of control candy sample and plantain candy sample. Alterations in colour parameters: d) *L** value and e) *a** value of control candy sample and plantain candy sample during storage. Phytochemical analysis of plantain candy sample during storage: f) total phenolic content (mg GAE/100 g D.M.), g) IC_{50} value by DPPH (mg/mL), h) FRAP value (mM $FeSO_4$/100 g D.M.), i) L-tryptophan, serotonin, and melatonin (µg/g D.M.) in plantain candy sample during storage. % RSD of high-performance of liquid chromatography analyses results≤3 %. The limits of quantification for L-tryptophan, serotonin, and melatonin were 1.88, 0.89, and 0.51 µg/L, respectively. Each value is mean value±S.D. of three sets of samples. Different alphabets denote that mean values belong to different subsets at $p<0.05$.

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SUPPLEMENTARY MATERIAL

Table S1. Details of ingredients used in candy formulation

Ingredient	Function	Permitted amount	Used amount	Reference
Sorbitol	Low calorie sweetening agent, forms the body of candy	GMP (FSSAI)	Sorbitol: D-mannitol=3: 1 (% by mass)	(*27*)
D-Mannitol	Low calorie sweetening agent, forms the base of candy	GMP (FSSAI)		
Gum acacia	Thickening agent, stabilizer, emulsifier	-	30 %	(*26*)
SiO_2	Desiccant, anticaking agent	-	0.5 %	(*26*)
Sorbic acid	Antifungal agent	1000 ppm (FSSAI)	0.1 %	(*26*)
Lyophilized green plantain powder	Principle fortifying material rich in antioxidants	-	65 %	-
Potable water	To solubilize the ingredients	-	50 %	-
Vanilla essence	Flavouring agent	-	100 µL	-

FSSAI=Food Safety and Standards Authority of India

Table S2. Microbial counts in PET/MET PET/PE packaged candies during storage

t/day	Control candy sample		Plantain candy sample	
	Bacteria count (CFU/g)	Yeast/mould count (CFU/g)	Bacteria count (CFU/g)	Yeast/mold count (CFU/g)
0	0^{a}	0^{k}	0^{a}	0^{k}
7	0^{a}	0^{k}	0^{a}	0^{k}
14	0^{a}	0^{k}	0^{a}	0^{k}
21	0^{a}	0^{k}	0^{a}	0^{k}
28	187.5^{b}	0^{k}	0^{a}	0^{k}
35	250^{c}	0^{k}	187.5^{b}	0^{k}
42	500^{d}	0^{k}	437.5^{g}	0^{k}
49	562.5^{e}	0^{k}	812.5^{h}	0^{k}
56	1750^{f}	937.5^{l}	687.5^{i}	0^{k}
63	-	-	1062.5^{j}	312.5^{m}

Data are mean value±S.D. of three samples of each set
Different letters indicate significant differences at $p<0.05$ level

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PET=polyethylene terephthalate; MET PET=metalized polyester; PE=polyethylene

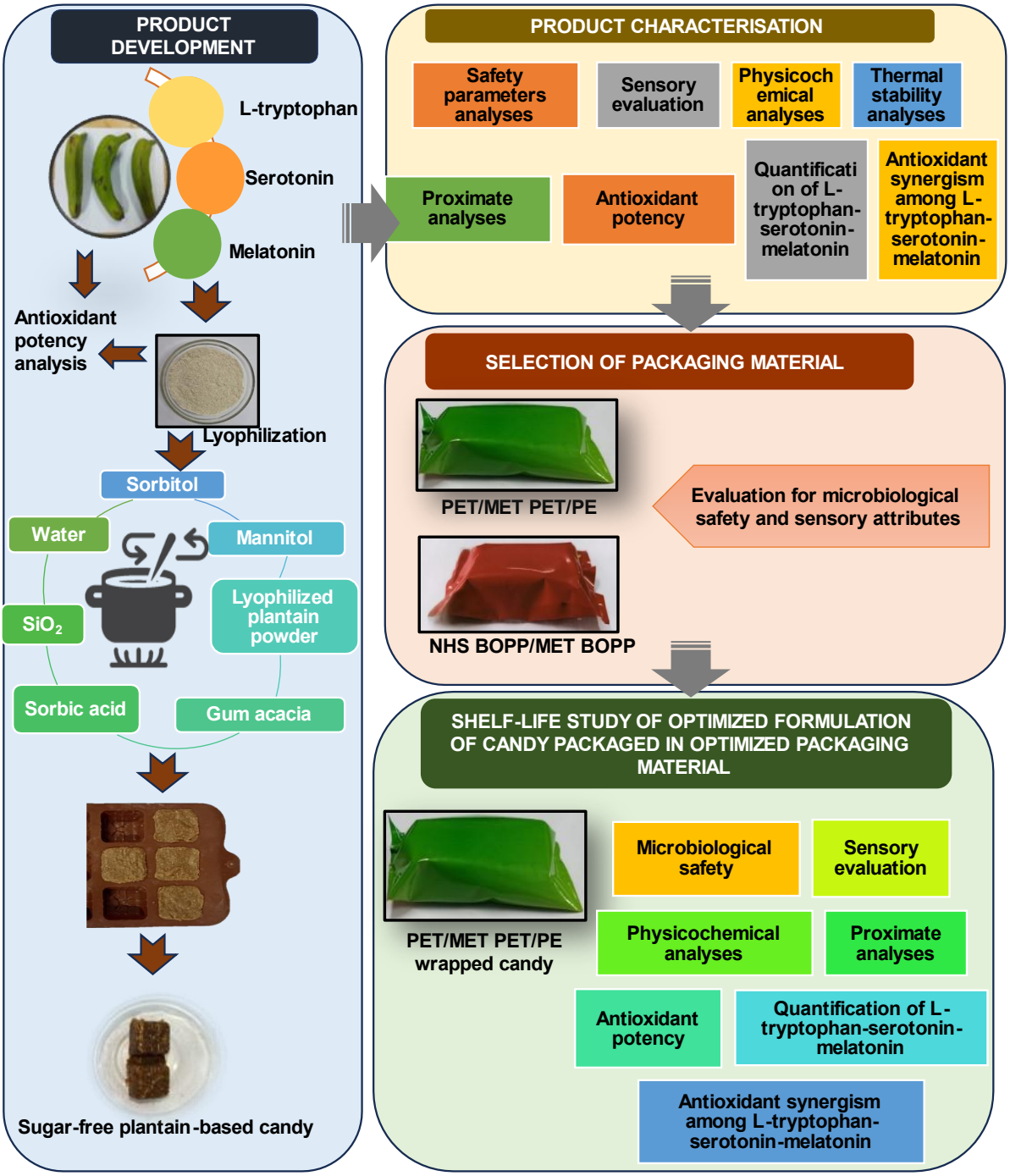

Fig. S1. Experimental design of the present study