

# Agro-industrial by-products to food additives: A review

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**Abstract:** The developments in science and technology have a remarkable impact on life existing on this planet. With increased interest in the food processing industry increases the amount of agro-industrial by-products generated. Most of these by-products are biodegradable organic matters which on processing can be used as a value-added product. These products are inexpensive materials and can curb environmental pollution which is a major concern worldwide if not handled. Food additives are natural or synthetic compounds added intentionally and not consumed as such includes sweeteners, colorants, antioxidants, anticaking agents, thickener, emulsifiers and stabilizers. It has been widely used to maintain stability in nutritional value, season supplies, sensory attributes and to protect from microbial attack. There is an increased demand for natural additives to replace their synthetic counterpart due to their toxic effects when consumed in the longer run. This review gives an insight to use of these by-products as food additives and their potential to replace the synthetic counterpart.

**Keywords:** Food additives, Food processing waste, chitosan, betalains, antioxidants.

## I. INTRODUCTION

Food waste without any doubt is the most challenging delinquent that mankind is currently facing around the world. It is biodegradable with a range of sources, including food industries. By-products and wastage are affecting ecological, monetary, and social areas [1]. The issue of food waste is expanding, including all areas of waste administration from assortment to removal. The best method for waste administration is to decrease the loss at its source. Avoiding food waste along its lifecycle is imperative for every one of those overseeing food production dispersion [2].

Food additives are the substances added to food to improve its sensory attributes. It befits share of the completed food item either directly or by implication, now and again purposefully and accidentally, during some phase of processing. It includes colorants (E100-199), preservative (E200-299); antioxidant, acidity regulators (E300-399); thickener, stabilizer, emulsifier (E400-499); anticaking agents (E500-599); flavour enhancer (E600-699); miscellaneous (E700-999) and additional chemicals (E1000-1999). Contemporary, the customers are opting for natural ingredients and bio-based additives due to antagonistic impacts by certain synthetics [3]. Natural additives are utilized to re-establish supplements lost or debased during production, invigorating or enhancing certain food varieties to address dietary deficiencies, or adding supplements to food substitutes. The use these food additive is regulated by national or international authorities [4].

One of the important sensory attributes is color which attracts the consumers. The natural colorants are susceptible to degradation in the processing of food. In order to overcome this loss synthetic colorants such as yellow (E100-109), orange (E 110-119), red (E120-129), blue (E130-139), green (140-149), brown (E150-159) and gold and other colors (E160-199) are used. Out of these colorants studies have reported that Ponceau 4R (E124), Allura red (E129), sunset yellow (E110), tartrazine (E102), chocolate brown HT (E155), brilliant blue (E133) and green S (E142) are most stable to degradation [5]. Food preservatives are added to delay food degradation by inhibiting the growth of micro-organisms, enzymes and prevents oxidation. The commonly used preservatives in the food industry are sorbate (E200-209), benzoates (E210-219), sulphites (E 220-229), methanoates (E230-239), nitrates (E 240-259), ethanoates (E260-269), lactates (E270-279), propoates (E280-289) and others (E290). These synthetic preservatives can be replaced by the by-products wherein chitosan acts as an antimicrobial agent, the onion by-product as an anti-browning agent and eggshell and peel of citrus and pomegranate as antioxidants can be used [6].

Acidulants as the name suggest are used to adjust pH, imparts sour taste and flavor modification in the food industry. It also prevents deterioration of food as the micro-organisms have criteria of pH to thrive and acidulants decrease the pH (acidification). The commonly used acidulants are acetic acid (E260), lactic acid (E270), malic acid (E296), fumaric acid (E297), citric acid (E330), tartaric acid (E334), phosphoric acid (E338) and succinic acid (E363) [7]. The use of dietary fibre by food industry as nutritional supplement to modify the texture and increase shelf life of a product during production and storage has increased over time. The incorporation of these leads to increase in water absorption and alters rheology of the food [8].

Sweetener is used as a substitute for sugar and is the most commonly used food additive. Based on their origin they are categorised as natural (glucose, sucrose, fructose, sorbitol, maltitol) and synthetic (aspartame, cyclamate) sweeteners. The relative sweetness of sucrose is 1 and is considered as a reference. The consumption of synthetic sweeteners may lead to

dental caries, weight gain and increased prevalence of diabetes which can be curbed by replacing it with natural sweeteners. The fruit peels are usually discarded by food industries which are a potential source of natural sweeteners. Emulsifier ensures the formation of an emulsion between two immiscible phases by reducing the surface tension at the interface, while stabilizer and thickener ensure consistency of food product. Gelatin extracted from fish has better emulsifying and stabilizing properties [9], while studies have reported the use of apple peel as a thickener [10].

The powdered food materials with low moisture content are prone to caking for example coffee creamer, table salt and cake mix. The most commonly used anticaking agent in food industry are silicon dioxide (E551), calcium silicate (E552), magnesium stearate (E572), sodium aluminium silicate (E554) and aluminium silicate/kaolin (E559). These agents increase the flow rate of a powder by reducing its stickiness [11]. Studies have reported that the ash of rice hull has silica which can be used as an anticaking agent [12]. This review provides an overview of the use of agro-industrial by-products as natural food additives and their beneficial effects.

## II. COLORANTS

Natural colorants have a minimal environmental impact, are safe to use and some are plant-derived. On the other hand, synthetic colorants are inexpensive and are stable. With advancements in novel technologies, natural colorants can be extracted with improved stability. Some of the plant by-products that can be used as colorants discussed here are peel of dragon fruit and banana.

**Betalains from dragon fruit peel:** Dragon fruit (*Selenicereus undatus*) is a tropical fruit grown prominently in the south Asia region. 30-45% of the total mass consists of the peel which is discarded as waste in the food industry. It has been reported that the peel of dragon fruit is a rich source of betalains [12]. Betalains are water-soluble, nitrogen-containing secondary metabolites with tyrosine as their precursor molecule. It is an ammonium conjugate of betalamic acid with cyclo-DOPA and amino acids or amines stored in the vacuoles of plant cells i.e. vegetative and reproductive tissues. Betacyanin (red-violet) and betaxanthins (yellow-orange) are the two major structural pigments. The peel of dragon fruit is rich in betacyanin which can be used as natural colorants [13] and functional food [14]. These betalains can be used as an alternative for the red colorants which is the most commonly used in food and beverage industry.

**Carotenoids from banana peel:** Banana is one of the most consumed and cultivated fruit in tropical and subtropical areas. The banana peel constitutes about 38 % of total weight which generates 4 million tons of waste annually [15]. The banana peel is reported to contain 3 to 4 µg/g of carotenoid content ( $\alpha$ -carotene,  $\beta$ -carotene, lutein, isolutein, violaxanthin, auroxanthin, neoxanthin,  $\alpha$ -cryptoxanthin and  $\beta$ -cryptoxanthin), high dietary fibre content, and potassium. The peels are reported to contain a significant amount of carotenoids compared to the fruit.  $\alpha$ -carotene,  $\beta$ -carotene, and  $\beta$ -cryptoxanthin act as precursors for provitamin A. Apart from being a source of carotenoids the antioxidant properties of the peel have been reported [16]. Instead of discarding the peel of banana, it can be used as natural colorants to replace the synthetic yellow colorant with numerous health benefits which also solve environmental issues.

## III. PRESERVATIVES

Preservatives are employed to enhance the keeping quality of a food product. Organic acids and parabens are widely used as anti-microbial agents as they diffuse passively through the membrane. Antioxidants scavenge free radicals and delays the oxidative deterioration of food. Butylated hydroxyanisole (BHA; E320) and butylated hydroxytoluene (BHT; E321) are the most commonly used synthetic antioxidants. Chitosan from shrimp waste as an antimicrobial agent and the use of eggshell and peel of citrus and pomegranate are discussed subsequently.

**Chitosan as antimicrobial agent:** Chitosan, a linear polysaccharide constituting  $\beta$  (1 $\rightarrow$ 4) linked D-glucosamine and N-acetyl-D-glucosamine (GlcNAc) distributed randomly. It is acquired by alkaline deacetylation of chitin found abundantly in the exoskeleton of Crustaceans. Chitosan contains many functional properties as a preservative, it has GRAS status, is biodegradable, and with creditable absorption capacity. It has been used as edible coatings to extend shelf life and maintain product quality [17]. Chitosan being a polycation exhibits its anti-microbial activity by interacting with the cell surface of microbes which consists of negatively charged macromolecule residues ultimately leading to membrane leakage. Water vapor permeability is inhibited by chitosan [18]. Studies report the antimicrobial property of chitosan wherein the sliced mango coated with 1-2% chitosan extended the shelf life up to a week without any sensory and chemical changes [19]. Similarly, its effect on broad-spectrum micro-organisms like algae, some bacteria, and fungi have been explored. The lipid oxidation and putrefaction in meat were reported to be reduced with the addition of 0.01% chitosan. *Bacillus subtilis*, *Escherichia coli*, *Pseudomonas fragi*, and *Staphylococcus aureus* are some of the spoilage bacteria that chitosan inhibited and it reduced the metmyoglobin proportion in the meat stored for a longer duration [20].

**Eggshell as an antioxidant:** Consumption of eggs from chicken, duck and quail is common worldwide. The by-products generated from home, food industry, poultry unit and bakery are calcified eggshell and eggshell membrane (inner and outer membrane). The shell of the egg consists of three layers- Cuticle is the outermost layer, Testa is the calcium carbonate layer and Mammillary layer is the innermost [21]. The major constituent of eggshells is calcium carbonate along with minor components of calcium phosphate and magnesium carbonate. While the eggshell membranes constitute protein majorly along with collagen, glucosamine, chondroitin and hyaluronic acid which in turn are components of cartilage. Calcining eggshells at different temperature ranges produces a powder that is used as a food additive and antimicrobial

agent [22]. The calcium chloride from the eggshell has been used to control the growth of mesophilic, and high-impact microorganisms. It was able to reduce the water reduction of newly cut fruits [23].

**Citrus and pomegranate peel as an antioxidants:** Pomegranate (*Punicagranatum* L.) and citrus (*Citrus aurantium*) fruit peel are some of the by-products generated on a day-to-day basis by the food industry. The peels, as well as the fruit, contain numerous dietary fibre, organic acids, sugars, minerals, and phytochemicals like hydrolyzable tannins, flavonoids, and phenolic acids which contribute to its antioxidant potential [24]. The methanolic extract of citrus peel is reported to be 10 times more effective compared with synthetic antioxidants like BHA and BHA that can be used to reduce rancidity. The total phenolic content and total flavonoid content were reported to be higher in frozen peels compared to fresh citrus peels which is suggested to be due to the inactivation of the enzymes that destroy antioxidants. The addition of 200 ppm of synthetic antioxidant and 2000 ppm of citrus peel extract had a similar effect on the corn oil [25]. Pomegranate peel is rich in ellagic acids and gallic acid which captures the free radicals generated during oxidative damage. Extract from pomegranate peel is used for bio-based edible coating or film alone or in combination with chitosan and alginate. This in turn reduces the respiration rate, enzyme activity, ascorbic acid oxidation and increases the phenolic content [24]. Paneer, an Indian traditional dairy product produced by coagulation of milk on the addition of pomegranate peel unveiled a greater ability to prevent peroxide formation and enhanced its shelf life. But the type of solvent used for extraction plays a pivotal role in the effect of the yield from fruit peel [26].

**Onion by-products as an anti-browning agent:** The enzymatic browning is of major concern in the agro-food industry due to loss of quality and economy. Polyphenol oxidase (PPO, EC 1.14.18.1), is the enzyme responsible for browning which involves phenolic oxidation and alteration of colour in fruits and vegetables prior to or during processing. In most fruits and vegetables, the optimum activity of PPO occurs around pH 6.0–6.5 and the minimum activity below pH 4.5 [27]. Onion (*Allium cepa* L.) is one of the most commonly used vegetables in Indian households and worldwide. The disposal of onion waste has been a major concern due to the swift evolution of phytopathogens and they are not suitable for fodder. Onion consists of flavonoids, phenolic acids and alk(en)yl cysteine sulfoxides (ACSO) as the active components. The sulfhydryl or thiol group present in the onion by-products have been reported to be a potential mitigator of PPO and the PPO activity in avocado fruit was significantly used on treatment with the same [28]. There are reports which also suggests the use of strawberry tree (leaves and branches) and apple pomace for inhibiting PPO and peroxidase in sliced “Rocha” pear (*Pyruscommunis*) [29]. The natural source is always convincing to be used as an anti-browning agent.

#### IV. ACIDULANT

Lactic acid (2-hydroxy propionic acid) is one of the earliest organic acids to be used in the food industry. It is a GRAS additive used as an acidulant and preservative. It is produced in large-scale chemicals via fermentation using the bacteria belonging to the genera *Lactobacillus*, *Lactococcus*, *Streptococcus*, *Bacillus*, and *Enterococcus*. The majority (approximately 75%) production of lactic acid across the world occurs in Cargill Incorporated, PURAC Corporation, Archer Daniels Midland Company, and Galactec fermentation facilities [30]. Maize, wheat, rice, and potato are agricultural products abundant in starch, which is used as raw material for the production of lactic acid. A study reports that food waste consists of 60% carbohydrates as starch which leads to the expression of amylase, a key point for lactic acid production. The potato peel and mango peel when fermented with anaerobic mesophilic produces lactic acid [31]. Milling of canes for sugar gives a by-product molasses which can be converted into lactic acid by using microorganisms. The sulphite pulp obtained from the cane industry was used in lactic acid production by SSF method using *Bacillus* sp. [32]. Some of the other by-products used for the production of lactic acids are corn cob, cassava bagasse, and carrot waste.

#### V. NUTRITIONAL SUPPLEMENTS

Some of the nutrients such as vitamins and minerals are lost during processing and storage. These losses are masked by enrichment or fortification of the product. Dietary fibre is the part of plant food that is resistant to digestion in the upper alimentary tract of humans. They are broadly classified as insoluble and soluble. Cellulose, hemicellulose and lignin belong to the former whereas mucilages, gums and pectin belong to the latter.

**Dietary fibre from mango and potato by-products:** Mango (*Mangifera indica*) is a seasonal tropical fruit and is used for the development of a various product or consumed raw with the generation of huge by-products which includes seed, peel, and kernel. Studies have reported that the by-products contain bioactive components like mangiferin and quercetin 3-O-galactoside (polyphenols); flavanols and xanthenes (antioxidants); lipids possible substitutes of cocoa butter and food-grade pectin [33]. Mango peels and fibrous waste pulp contain approximately 74 % dry matter of dietary fibre. Dietary fibre is a potential functional food and food additive which lowers the blood pressure, improves serum profile, regulate plasma glucose levels and assist in weight loss. Mango peel powder and peel kernel when incorporated into biscuits improve the fibre content from 6.20% to 16.79% [34]. After rice, wheat and maize, potato (*Solanum tuberosum* L.) is the most consumed agricultural crop by humans generating potato peel as a by-product. Potato is a rich source of starch per se the peel also has it as a major constituent along with non-starch polysaccharides, polyphenols lignin, protein, and lipids in minor quantity. These are low-cost products [31]. Potato peel flour when used in food formulations at various concentrations enhanced the nutritional, technical, and stability of foods. The water-binding capacity and fat absorption capability of flours produced from the peels showed promising results [35].

**Fish waste as a protein source:** According to the Food and Agriculture Organization (FAO), 27 % of the total fish catch goes unutilized due to a lack of transportation, storage, or spoilage. These lead to an increase in pollution when dumped in open. The by-products generated are skin, frames, head, fins, and viscera which are abundant sources of collagen, gelatin, protein, and amino acids [36]. Enzymatic hydrolysis of these leads to the generation of fish protein concentrate (FPC) and food hydrolysate with application in the food and pharmaceutical industry. Fish frames consist of lysine, valine, and phenylalanine that can be extracted by enzymatic hydrolysis to produce a protein of superior quality compared to other animal sources. Fish protein hydrolysate is applied in the industry due to its unique property water holding capacity, oil absorption, protein solubility and emulsification ability [37].

## VI. SWEETENERS

Sweeteners are food additives that are used or intended to be used either to impart a sweet taste to food. Natural sweeteners are the most nutritive dietary sweeteners like sucrose, fructose, and glucose. Glucose and fructose are natural sweeteners produced from food waste of pineapple peel waste, apple pomace, date syrup waste, and agro-industrial like cashew industry [38]. Sucrose is considered to be the major sweetener responsible for improving the acceptability of food for centuries. It is abundantly present in the peel of some fruits like prickly pear (*Opuntia ficus-indica*) than in the juice and pulp. These fruits are used to extract natural liquid sweeteners as a healthy alternative to artificial sweeteners. The yellow-skinned fruits are reported to contain 2.21 mg per gram of sucrose while the red skin has 6.36 mg per gram of sucrose [39].

Similarly, invertase is a natural sweetener extracted from prickly pear waste. Invertase ( $\beta$ -D-fructofuranosidase; EC.3.2.1.26), is the enzyme with optimum pH 3-5 and temperature of 40-60°C hydrolyze sucrose to glucose and fructose (invert sugar syrup). There are three isoforms of invertase in plants based on pH and subcellular localization. Cytosolic invertase (alkaline) is active at pH 7.5 while the vacuolar and cell wall invertase (acidic) is active at pH 5 or below. Due to the hygroscopic nature of invert sugar, it finds a place as humectants in the manufacturing of soft-centred candies and fondants [40]. Some of the microorganisms, widely studied for the production of invertase is *Saccharomyces cerevisiae*, *Candida utilis*, *Aspergillus niger*, *Thermomyces lanuginosus* and *Penicillium chrisogenum*. Upon characterization of invertase produced by *Aspergillus flavus* using agro-industrial waste orange, pineapple, and pomegranate fruit peel as the substrate it was reported to be stable at 50°C [41]. Invertase is used widely used in the food industry where fructose is the preferred sugar as it does not crystallize readily and is sweeter compared to sucrose.

## VII. EMULSIFIER, STABILIZER AND THICKENER

The most commonly used synthetic emulsifiers in the food industry to avoid rancidity and ensure mixing of lipid-based food; stabilizer provides texture while thickener increases the viscosity. The most commonly used emulsifiers are polysorbate-80 and carboxymethyl cellulose which has GRAS status. The commonly used stabilizers are sodium alginate and sodium carboxymethyl cellulose. Hydrocolloids are used as a thickener and gelling agent. Long-term use of these additives may have lead to various metabolic syndromes [42].

**Fish skin as emulsifier and stabilizer:** Gelatin is an albumin like-protein derived by hydrolysis of triple helix conformation of collagen. It is a multifunctional ingredient used in food, cosmetics, and pharmaceutical industries that is acquired from cattle hides, pigskin, and bone. There are two types of gelatin based on the method of hydrolysis and the source used. Acid hydrolysis of collagen from porcine skin leads to the generation of type A gelatin with isoelectric pH 9, while alkali hydrolysis of collagen from bovine hide and bone leads to type B gelatin with isoelectric pH 5 [43]. But the major limitation is its source of origin leading to religious concern. Studies have reported that fish skin from various species of fish is used in gelatin production. Due to gelatin foam stability and emulsification, it acts as a stabilizer in pies, bread, and cakes. Researchers have characterized the fish gelatin from Ghol fish/Blackspotted Croaker (*Protonibedia canthus*) and reported it to contain high-quality collagen. The presence of proline and hydroxyproline plays a crucial role in stabilizing collagen structure. A lower level of proline and hydroxyproline leads to a reduction in the melting and gelling temperature of gelatin [44].

**Fruit peel as a thickener:** When the fruit ripens or heating in an acidic medium leads to the conversion of insoluble protopectin present in the middle lamellae of a plant cell to soluble pectin. Pectin is a negatively charged reversible colloid with an average molecular weight between 50-150kDa. It is a linear structure polymer consisting of D-galacturonic acid residues ranging from a few hundred to thousand linked via  $\alpha$ -(1→4)-glycosidic bond. Pectins can contain non-sugar substituents, methanol, phenolic acids, and amide groups. Based on its degree of esterification it is categorised as low methoxy (<50) and high methoxy (>50) pectin [45]. Pectin due to their thickening and gelling properties are used as food additives (E440) in jams and confectionery products. Apple pomace and citrus peel are agro-industrial waste used to produce food-grade pectin. [46]. A comparative study reported the thickening property of peel of passion fruit flour with that of low and high methoxylpectins, xanthan gum, guar gum and carrageenan [47]. Researchers have extracted pectin from the peel of dragon fruit (red powder) and honeydew melon (*Cucumis melo*) which are potential thickeners that can be used in the food industry without any hazardous effect [48].

## VIII. ANTICAKING AGENT

Caking of hygroscopic food material has been a common problem in the food industry. Glass transition temperature and moisture sorption isotherm have a large impact on the caking property. Rice is the staple food in Asia, some parts of

Africa, and Latin America. It belongs to the *Graminae* family which also includes oats, maize, barley, and wheat. Rice hull or husk is the by-product once the grain is removed. These hulls cannot be used as animal feed due to their low cellulose and sugar content. India alone produces 12 million tons of husk annually. The major constituent of rice hull ash is silica (SiO<sub>2</sub>) (85-95%) [49]. The hull contains three-fourths of organic volatile matter and the balance is converted into rice hull ash. Rice hull ash contains nearly 90% of amorphous silica, highly porous and light-weight which can be used as an anticaking agent [50]. The extraction of precipitated silica from rice hulls is economical, cost-effective, and of superior quality compared to silica from quartz. The presence of a high amount of silica makes it a valuable material for use in industrial applications. Silicon dioxide (E551) is the most widely used anticaking agent in the food industry, interestingly the rice hull particles have similar properties like flowability and scavenging moisture similar to silicon dioxide. The addition of rice hull to salt as an anticaking agent during salt manufacturing at a concentration of 1% could replace synthetic anticaking agents [51].

## IX. CONCLUSION

During processing, a lot of by-products are generated which create serious environmental issues. Many renewable products are being derived from fruit, vegetable, meat and fishery which are accommodating in industrial use and are cost-effective. Biomolecules and nutrition from waste organic sources could cut production costs and eliminate the anthropogenic impact on an already-threatened ecosystem. Consumer demands for natural sources, health advantages, and sustainability must be met by any food additives. Food additives are interrelated in shelf life and maintain the product quality. The utilization of novel waste methods to overcome food waste and environmental problems is one of the approaches to overcome this crisis. Efforts maybe are focused on yield increment from processing waste, sustainable food packaging and reduction of chemicals used in food industries.

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