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Review Article

Natural Food Colourants Juxtaposed with Synthetic Food Colourant: A Review

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Abstract

Application of synthetic colourants to foods, snacks and beverages has been on the increase within the past 50 years and up to 500% increase has been reported. Consumers of colour foods and beverages have been showing worries on the possible health hazards of such products over time and this has led to a series of reports by researchers on the health implications of synthetic food colourants. This present study took a hard look into variously published reports on a plant-based and commonly used synthetic food colourants. It is revealed that synthetic colourants cause more harm than good to a human and some harmful compounds have been revealed in synthetic colourants. The noticeable side effects of consumption of synthetic colourant include behavioural problems inform of neurobehavioural disorder, aggression, attention-deficit/hyperactivity disorder (ADHD) and inflammatory cascade. In conclusion, synthetic food colourants should be replaced with natural food colourants to avoid further health effect on humans. Also, parents and food producers should be advised on the adverse effect of synthetic food colourants while the government regulates every food production company to ensure proper choice and application of food colourants as well as candid labelling for safety purposes.

Key words: Food colourants, synthetic colourants, natural colourants, health effects, health benefits

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INTRODUCTION

Food additives are defined as substances or mixture of one or two substances that are not a basic component of the food, which is introduced to food to check factors such as spoilage, degradation, nutrient loss, decrease or loss of aesthetic properties and functional properties[1-3]. According to Food and Drug Administration (FDA)[4], a food colourant is defined as any pigment, dye, or substance which when applied or added to a drug, food, cosmetic, or the human body, is capable (through reactions with other substances or alone) of imparting colour. In addition to the definition, food additives do not serve as an agent hiding the poor quality of food and food products[5]. Various food products for example chocolates, chips, dairy products, drinks, fermented products and others attain a high shelf-life level due to the addition of food additives. Food additives such as food colourants and others can either be direct or indirect depending on the purpose for which it is applied and their use is regulated to ensure the safety of that food in which they are added[6]. Addition of food additives in food products at various stages is done for unique purposes which include; colour improvement, nutritional improvement, prevention of microbial invasion, avoids physicochemical changes, prevents oxidation reaction of any kind, to improve the organoleptic properties of the food such as flavour, taste, texture, mouthfeel, aroma, to extend shelf life via inhibition of; ripening, sprouting and browning. Food additives are of two types; it can be natural or synthetic depending on the source from which it is derived. Salt and wood smoke are ancient natural food preservatives since 900BC, while mauvine is the premium synthetic food colourant[7].

Natural colourants are the first food colourants used in the Palaeolithic period before the birth of synthetic food colourants[8]. In China, the premium record on the use of natural dyes is dated back to 2600BC[7]. Also, Saffron, as mentioned in the Holy Bible, is an orange-yellow colour dye made from the saffron plant's stigma and hennas, a reddish plant substance prepared from the dried leaves of *Lawsonia inernis* was used prior 2500 BC. Wrapping of dead bodies in Egypt was done with coloured cloth[5], in the quest for knowledge, a chemical test carried out on the red cloth is seen in king Tutan Khamen's sepulchre confirmed the presence of alizarin. Alizarin is a pigment, extracted from a herbaceous plant, *Rubia tinctorum* cultivated for a red-purple dye obtained from the root; it is also known as madder. In the damage of Menhnanjo-daro and Harappan Civilisation in the 3500 BC, a subcontinent of India, dyes were applied and this was confirmed by a coloured garment of cloth and traces of madder dye found at the scene[8]. Within 1500 BC in Egypt, candy producers do apply natural colour extracts as well as wine to candies in order to improve its aesthetic value. Dyes such as brazil-wood, indigo, madder and dark reddish-purple colour were known in the 4th century which gave the name Brazil to the country because brazil-wood and a blue dye got from the leaves of *Isatis tinctoria* known as woad was first found there. In Japan, the application of natural colourants in food was traced to the 8th century from the Nara moment text, in shosho which has references to adzuki bean cake and soybean containing coloured substances. The people of Aztec and Maya culture moment of Central and North America were the first to use cochineal dye extracted from edible insects. Mauve is the first synthetic dye, manufactured in the year 1856 by Sir, William Henry Perkin[9]. Further production and extraction of synthetic food colours sets-in during the early 19th century. They were produced from petroleum-derived products such as aniline. Coal serves as the raw material, which gave it the name "coal-tar"[7]. The relationship between the art of colouring and civilisation has made scientist dwell in deeper research on food colourants, scrutinising for its safety purposes[10].

Consumer's concern about the toxicity of most synthetic food colourants such as cancer, a hypertensive reaction in children, allergies, asthma, neurobehavioural hazard among others has led this research to critically look into mostly applied synthetic food colourants and natural food colourant as a healthy alternative, to help both the manufacturer and consumer in making a healthy choice always.

Advantages of using food colourants: We eat with our eyes. An adage says; humans feast/eat with their eyes first. This is true in all perspective. The food colour, either natural or synthetic source, is a very crucial factor to the degree of pleasure man derives from food. Colour and flavour are major factors that make food appealing to the consumer. It is proven that we taste with our eyes; this shows the important role played by colour in life. Colour, flavour, shape and other physical properties of food always affect the eating desire of humans[11,12]. Information on a food product is mostly dependent on the colour it has. Whenever the colour of a food deviates from the information it is originally meant to convey; a negative psychological effect is being created in human on the physicochemical properties of the food. Colour of food affects its general acceptability due to people are easily moved by colour[5].

Additionally, the degree of sweetness and palatability of food is also affected by its colour which deems it right to say that colour and appearance play a vital role in the quality attribute of a food. Colour is as old as mother nature herself and has been used since the onset of recorded history to increase the appeal of foodstuffs. It is a major factor for acceptability of products such as food, cosmetics, textiles and others because naturally, man has a keen interest in colour.

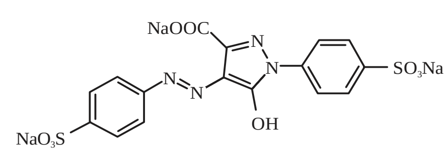
Addition of food colourants to foods, ranging from candy to wine is essential to meet the expectations of consumers. Foods can be coloured due to its effect as in the case of green ketchup by Heinz in 2000. Food colour can affect the flavour of foods because humans relate distinct flavours to distinct colours. Due to variation in colour of foods through the seasons, coupled with the effect of processing and storage, colour addition is employed commercially to retain the preferred and expected colour by consumers-example; adding red colour to glace cherries. Primarily, the reasons for colouring foods include; to bring back the original appearance of the food which may have been affected by the processing operations, to improve the aesthetic attribute of the food, to make the food appealing and irresistible, to enhance the visual characteristics of the food, to apply colour to virtually colourless foods, to ensure uniformity in the colour of a food batch, to enhance the already existing food colour, to provide identity to the food, to protect vitamins and flavours from destruction by light, to mask natural variation in colour, to stimulate appetite, to prevent discouragement and reduction in the desire for the food and beautification/artistic purposes as in cake icing.

Classification of food colourants: Generally, food colourants are divided into two major classes depending on their sources as synthetic and natural.

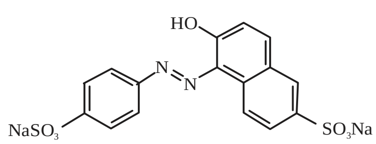
Synthetic food colourants: Synthetic food colours, also referred to as artificial food colours are man-made food colourants, manufactured through the chemical synthesis of organic coal-tar. In food, pharmaceutical and cosmetic industries, synthetic colourants are mostly used because of the following properties; high colouring ability, brightness, stability, application ease, homogeneity, a variety of colour shades and less expensive, although its health safety issues are alarming[13]. Examples of the synthetic colourant are sunset yellow, tartrazine, brilliant blue FCF, ponceau 4R, fast green, Allura red, carmoisine. Synthetic colourants are mainly derived from the synthesis of chemical substances of petroleum family such as coal-tar. They are more economical for large scale and industrial use because they have various shades of colour readily available for use, they are affordable, the colours are bright, homogeneity properties and good colour shade, which makes them more popular[5]. A large number of synthetic colourants, similar to organic aniline dyes with different colours and rainbow shades have been developed and are used in food colouring without knowledge on their safety. Regulators moved into critical research on synthetic food colours due to the significant toxicity of petroleum-based colourants and aniline. The most popular certified food colour is FD and C Red No. 40, followed by FD and C Yellow No. 5. Food colourants with the acronym FD and C show that the Food and Drug Administration (FDA) has approved its application in food, drugs and other products. For application in foods, the certified synthetic colours are; Tartrazine (lemon yellow), Orange B, Brilliant Blue FCF, Fast green FCF, Indigotine, Citrus red AC (orange) with assigned E-numbers[14]. Synthetic food colourants can further be classified into two major classes: primary and secondary food colourants.

Primary food colourants: These are food colourants that exist basically on their own. This class of food colourants when mixed with another primary colourant yields a desired different colour. Examples include; tartrazine, quinolone yellow, erythrosine, sunset yellow FCF, ponceau 4R, carmoisine, Allura red, amaranth, chocolate brown HT, patent indigo carmine, blue v, black PN, fast green, fast red, red 2G, brilliant blue FCF.

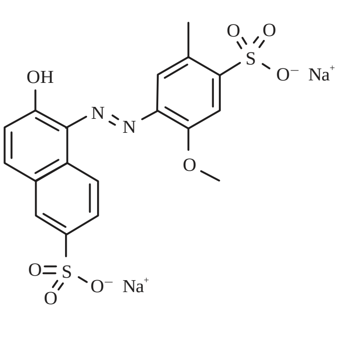
- **Tartrazine:** Tartrazine is a lemon-yellow, water-soluble, azo dye synthetic colourant, with a molecular formula $C_{16}H_9Na_3O_9S_2$, CAS 1934-21-0 and molar mass of 534.3 g mol^{-1}. The International Union of Pure and Applied Chemists (IUPAC) name is trisodium (4E)-5-oxo-1-(4-sulfonatophenyl)-4-((4 sulfonatenyl) hydrazono) 3 pyrazole-carboxylate. Other names for tartrazine includes E number E102 or C.I 19140, FD and C yellow 5. Tartrazine has 427 = 2nm maximum absorbance when in a liquid solution. According to the European Food Safety Authority (EFSA), tartrazine is mostly applied in cereals, honey and lemon products, sauce, sports drinks, chewing gum, yoghurt, noodles, pickles, fruit squash, potato chips, energy drinks, flavoured corn chips, cake mixes, ice cream, candy and others[15,16]. Tartrazine is the most commercially used synthetic food colourant due to its yellow colour and also can be used in the manufacturing of green shades when in combination with other synthetic food colourants such as Green S (E142) and brilliant blue FCF, FD, E133 and C blue)[15].



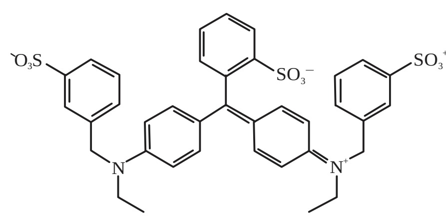
- **Sunset yellow:** Sunset yellow FCF is a yellow, water-soluble synthetic coal tar azo dye with E number E110, IUPAC name disodium 6-hydroxy-5-[(4-sulfophenyl) azo]-2-naphthalene sulfonate, a molecular formula of $C_{16}H_{10}NaO_7S_2N_2$, the molar mass of 452.37 g mol^{-1}, CAS No. 2783-94-0 and a melting point of 300° C. It is also known as orange-yellow 5, C.I. 15985; FD and C yellow 6 and E110. It is very common in fermented foods. When applied in fermented foods, the food product should be properly heat-treated[16,17]. When dissolved in water, sunset yellow FCF passes through a change in phase that moves from anisotropic liquid to a nematic liquid crystal. It is mainly used in food like packet soups, lemon curd, sweets, soft drinks, sweets, apricot jam, orange jelly and squash, bread crumbs, Swiss roll, snacks and other food products that have red, orange and yellow colour[15,18]. Sunset yellow has been linked with a lot detrimental effect on human such as cancer, hyperactivity in children, allergic reactions, diarrhoea, vomiting, skin irritation and has been repeatedly called to be banned from products.



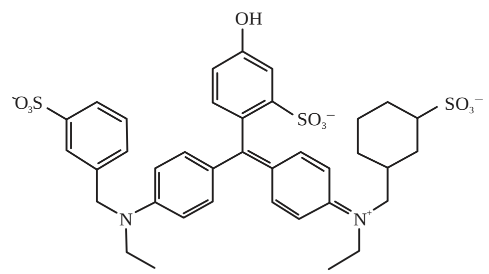
- **Allura red AC:** Allura Red AC as the name implies is a red azo dye, with the IUPAC name; Disodium 6-hydroxy-5-(2-methoxy-5-methyl-4 sulfophenyl) azo] -2-naphthalene sulfonate; molecular formula: $C_{18}H_{14}N_2O_8S_2$, Molar mass of 496.42 g mol^{-1}, CAS No. 25956-17-6, melting point of >300°C and E number E129. Other names of Allura Red AC are Food Red 17, Allura Red, FD, C.I. 16035 and E129, C Red 40 2 naphthalene-sulfonate acid, disodium salt; 6-hydroxy-5- [(2-methoxy-5-methyl 4sulfophenyl) azo]-2-naphthalene-sulfornate. It has a maximum absorbance of 504 nm in water and is gotten from petroleum or coal tar and is applied in products like children' drugs, soft drinks, cotton candy as Food and Drug Administrator approved[3].



- **Brilliant blue FCF:** Brilliant Blue is a water-soluble coal tar derived synthetic food colourant with the E number E133 and 42090 as a colour index. Brilliant Blue FCF has a maximum absorption of 628 Nanometer in water solution. It has a molecular formula of $C_{37}H_{34}N_2Na_2O_9S_3$ and CAS No. 3844-45-9. Its other names include; patent blue AR, D and C Blue No 4, Antacid Blue FG, Xylene Blue VSG, FD and C Blue No. 1, Erioglaucine Eriosky blue, Acid Blue 9 and Alzen Food Blue No. 1. It appears in a reddish-blue powder form and can give various shades of green colouration when mixed with tartrazine (E102). Mostly, Brilliant Blue FCF is a disodium salt but can also exist as calcium and potassium salts, with 2650-18-2 as its CAS number. Brilliant Blue FCF is commonly present in sweets, canned, processed pear, dairy products and ice-cream[17]. It has been banned in most countries such as Switzerland, Germany, Italy, Greece, Norway and Austria among others due to its power to stimulate an allergic reaction in asthma patients.

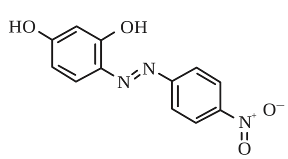


- **Fast green FCF:** Fast green FCF is a synthetic sea-green triaryl methane food colourant with IUPAC name;ethyl-[4-[[4-[ethyl-[3-sulfohenyl]methyl]amino]phenyl]-[4-hydroxy-2-sulfophenyl]methylidene]-1-cyclohexa-2,5-dienylidene]-[[3sulfophenyl]methyl] azanium, molecular formula of $C_{37}H_{37}N_2O_{10}S_3$ and the E number E143. Its maximum absorption is 625 nm. Other names for fast green FCF includes; green 1724, food green 3 and C.I. 42053. Fast green FCF has poor absorption by the intestine. This food colourant has been confirmed to cause the growth of tumour and mutagenic effect both in humans and animals. Also, in concentrated form, fast green FCF can lead to skin, eye, respiratory and digestive tract irritation. Its application is found in jellies, fish, sauce, desserts, vegetables, green peas and dry bakery mixes at 100 mg kg^{-1} and has been banned in some countries as well as European Union[1].

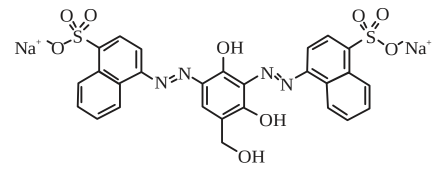


Secondary food colourants: These are synthetic colourants produced by blending of two primary colours. It is done by the manufacturer to get a desired colour shade and strength to satisfy the consumer. Example chocolate brown, dark chocolate, blackcurrant, coffee brown, pea green, lime green, apple green, egg yellow, yolk yellow, strawberry red, rose pink, raspberry red, grape and violet.

- **Azo violet:** Azo violet is a dark reddish-brown organic chemical substance with the molecular formula $C_{12}H_9N_3N_3O_4$; IUPAC name 4-[4-ni trophenyl] azobenzene-1,3-dol, the density of 1.45 g/cm^3 and a vapour temperature of 261.7°C. Azos can be described as compounds derived from diphenyl diazene, azobenzene or diazene which are nitrogen-based[13]. At times, it is applied as a pH indicator or a dye; it gives different colour shades based on the salts, the amount used and the medium. In a slightly alkaline medium with magnesium, it gives blue colour and in a salt solution of Magnesium will form a violet colour. It is still used for magnesium test.



- **Chocolate brown:** Chocolate brown is a synthetic coal tar diazo dye that is brown with molecular formula $C_{27}H_{18}N_4Na_2O_9S_2$, Molar mass of 652.56 g mol^{-1} and E-number E155. It is applied mostly in chocolate cakes, yoghurts, jams, fish, milk, cheese, fruit products and other products. Its application is banned in some countries such as France, Denmark, Australia, Norway, Sweden, United States and Switzerland due to its ability to trigger allergic reactions in asthmatics people, skin sensitivity and other individuals sensitive to substances such as aspirin[2].

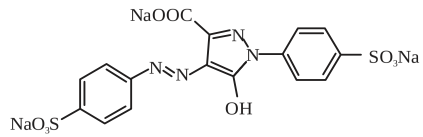
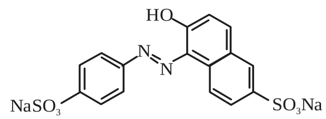
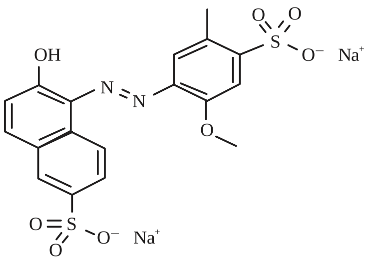
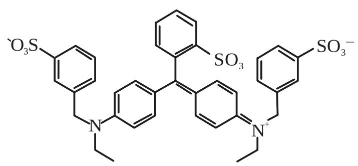
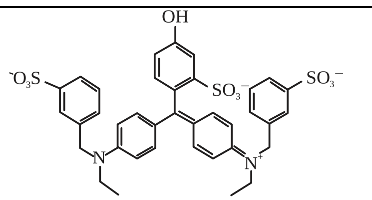
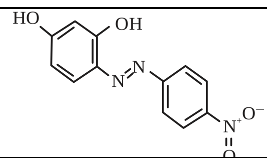
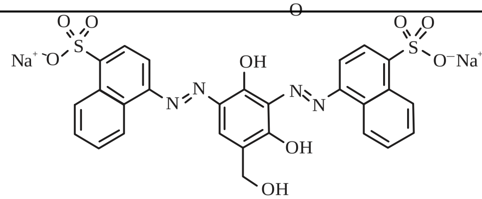


Chemistry of synthetic food colourants: In food products, colour additives are available as either dye or lake pigments.

- **Dye:** Dyes are synthetic colourants, soluble in water but insoluble in oil. They are produced in granules, liquid, powders and another form in which they are needed. Dyes are applied in food products like dairy products, baked products, dry mixes, beverages and pet foods. An example includes azo dyes[13]. Dyes are capable of colouring stool when ingested in a higher amount, which is one of the adverse effects of dyes, unlike lakes.
- **Lakes:** This group of synthetic colourants are manufactured by the mixture of dyes with salts to yield both water and oil insoluble compound but are oil dispersible. Lake colourants are a finely powdered precipitate of aluminium hydrate substrate and are stable and ideal for fats, oils or moisture lacking products[3]. Dye's particle size and content determine the tone of the lake's colour. It is applied in cakes, hard sweet, powdered drinks, confectionery, chewing gum and biscuit fillings[18]. An example includes; lake Amaranth (E123), lake Sunset yellow FCF(E110), lake Tartrazine (E102), lake Carmoisine (E102), lake Ponceau 4R (E124), Brilliant lake blue (E133), lake Allura red (E129). Table 1 below shows the structure, property, application and safety of synthetic food colourant.

Natural colourants: Natural colourants are the group of colourants derived from nature, either from plant source (fruits and vegetables), animal source (insects, shellfish, cow urine), mineral source (iron, archers, umbers) or microbial source (algae, yeast, fungi and bacteria)[5]. Natural colours are present in our daily diets. Through our daily meals that we eat, we consume chlorophylls, anthocyanins and carotenoids. For food production purposes, they are extracted from their natural source and then applied to foods and food products to improve its aesthetic value and to ensure that identification properties are not defeated. Nowadays, consumers are seriously concerned with their health and what they eat, which makes them move towards natural food products. Natural colours are moderately bright, have an antioxidant effect and also are bioactive. Natural colours have relatively no

Table 1: Structure, property, application and safety of synthetic food colorant

Colourant	Structure	Property	Application	Safety
Tartrazine		A lemon-yellow, azo dye, water-soluble synthetic colourant, with a molecular formula $C_{16}H_9Na_3O_9S_2$ and molar mass of 534.3g/mol and E number E102	Cereals, honey and lemon products, sauce, chewing gum, yoghurt, noodles, pickles, fruit squash, chips, energy drinks and dairy products.	Not safe
Sunset yellow		Water-Soluble synthetic coal tar and an azo yellow dye which is denoted by the E number E110, a molecular formula of $C_{16}H_{10}NaO_7S_2N_2$ and the molar mass of 452.37 g mol^{-1}	Packet soups, lemon curd, sweets, soft drinks, sweets, apricot jam, orange jelly and squash, bread crumbs, Swiss roll, snacks	Not safe
Allura Red AC		A red azo dye, with the IUPAC name; Disodium 6-hydroxy-5-(2-methoxy-5-methyl-4 sulfophenyl) azo] -2- naphthalene sulfonate; molecular formula: $C_{18}H_{14}N_2O_8S_2$, Molar mass of 496.42 g/mol, the melting point of >300°C and E number E129	Children's medications, soft drinks, cotton candy	Not safe above approved level
Brilliant Blue FCF		Water-Soluble coal tar derived synthetic food colourant with the E number E133 and 42090 as a colour index. Brilliant Blue FCF has a maximum absorption of 628 Nanometer in water solution. It has a molecular formula of $C_{37}H_{34}N_2Na_2O_9S_3$	Sweets, canned, processed pear, dairy products and ice-cream	Not safe
Fast green FCF		A synthetic sea-green triarylmethane food colourant, the molecular formula of $C_{37}H_{37}N_2O_{10}S_3$ and the E number E143. Its maximum absorption is 625 nm.	Jellies, fish, sauce, desserts, vegetables, green peas and dry bakery	Not safe
Azo violet		A dark reddish-brown organic chemical substance with the molecular formula $C_{12}H_9N_3N_3O_4$; density of 1.45 g/cm^3 and a vapour temperature of 261.7°C	pH indicator, the colouring of salts and magnesium test	Not safe
Chocolate brown		A synthetic coal tar diazo dye that is brown with molecular formula $C_{27}H_{18}N_4Na_2O_9S_2$, Molar mass of 652.56g/mol and E-number E155	chocolate cakes, yoghurts, jams, fish, milk, cheese, fruit products	Not safe

health effect on human unlike the synthetic colourant, therefore are recently used in food industries to maintain and satisfy their customers[19].

Natural food colourants are those substances derived from mineral, plant, animal or any other natural sources that can serve as a colourant for food, pharmaceutical and cosmetic purposes. Natural colourants remain the safest for business purposes due to consumers' consciousness of health implications caused by synthetic food colourants[19,20]. A highly purified form of these natural colourants is usually made available to attain good yield, enhance its stability, meet consumers demand and for convenience purposes[21]. Some factors as pH, temperature, light, storage time and presence of other ingredients are considered in the application of natural food colours. Examples are; Annatto, Paprika, Saffron, caramel, cochineal, saffron, chlorophyll, Anthocyanin, Betanin, betalain, lycopene, carmine.

Classification of natural colourant: Natural colourants can be classified based on; origin, extraction methods and chemical constituents.

- **Classification based on origin:**

Plant origin: This group of natural colourants are derived from trunks of plants, the bark of trees, fruits, flowers, leaves, dried or wet vegetables of plants. All these mentioned parts of plants produce colour in their unique way which includes purple and blue colour form cornflower which is mainly applied as sugar colouring, tea ingredients and also for confectioneries[22], red colour fruit juice from bilberry that can turn blue in basic medium. Brown colour from *Camelia tea*, the curcumin which is the primary colour pigment that yields yellow-orange colour from *Curcuma longa* L. (turmeric) mainly used in beverages, cereal, baked and dairy products, green colour from chlorophyll which the green pigment found in plants. Blueberries also yield blue colour which is applied in foods and medicine[18] Red colour from Paprika as a result of capsanthin and capsorubin which are carotenoids[23]. Other examples are madder roots; red, turmeric; yellow, waterlily-blue. Most of these colourants from plants also have other application such as medical purposes[22], the colouring of papers and others. Over 500 species of vegetables and plants are found suitable to serve as colourants in India[7].

Animal/insect origin: These colourants produced from animal sources is gotten from dried animal/insect bodies and their secretion. The oldest animal dye and most expensive is known as murex. Examples are; sea mollusc secretes a deep violet colour, shellfish, lac insect secretion and cow urine[7]. Cochineal (*Dactylopius coccus*) is an insect. The red dye extracted from this insect and its egg exists n form of ceramic acid and is mostly applied in foods such as yoghurt, candy, juices and ice cream. It can lead to an allergic reaction as well as anaphylactic shock in some humans. Sepia Officinalis L (female cuttlefish) gives rich concentrates of an orange-red pigment known as sepiaxnthine[22].

Mineral origin: These group of natural colourants unlike that of plant and animal origin is derived from earth or minerals such as umbers, iron, hydrated oxides and oxides of iron and manganese, archers, titanium dioxide[24-26]. This group of natural food colourant can be applied in food pantries, paints and cosmetics.

Microbial origin: As the name implies, these natural colourants are gotten from microbiological action of algae, fungi, yeast and bacteria and are mostly applied in energy drinks, baby foods, sauces and dairy products[27]. These natural colourants include yellow from ash by a gossip, brown from bacillus, red from Dunaliella sali. These colourants have both anticancer properties and antioxidant. Monascus purpureus is a fungus that produces a red pigment traditionally used in candy production in some oriental countries[28]. They are affected by pH, moisture content, temperature and fermentation during production. Several food colourants have been isolated from microbes and applied as food-grade colours, they include; Astaxanthin, Canthaxanthin, Prodigiosin, Violacein, Phycocyanin, Ribo avin, Beta-carotene, Lycopene, Melanin among others[27].

- **Classification based on extraction methods:** Natural colourant under this group is manufactured by natural sources such as plant, fungus, microbes, minerals and animals. In the preparation processes which are a very difficult process, the ink is manufactured from the colourant extracted from the natural source using appropriate constituents. There are various methods of extraction of natural colourant and they include; aqueous extraction, ultrasonic and microwave aided extraction, Solvent extraction, Acid/alkali extraction, enzymatic extraction, Fermentation extraction and Supercritical fluid extraction[29].

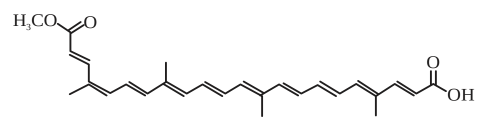
Classification based on chemical constituents:

Class	Composition
Anthocyanidins	Under this group falls carajurin which is produced from Bignonia chica
Anthraquinone	In this group are the red colour dyes, examples are cochineal, lac, Indian madder, Alizarin and morinda
Carotenoids	In this group, the presence of the double bond determines the colour. The major natural colourants under this group are Bixin and norbixin from annatto seed and crocin from saffron stigma
Dihydropyran	This group are replaced dihydropyrans with a flavonoid-like structure
Flavonoids	Under this class are the yellow pigments, they include rutin, quercitrin, luteolin and others
Naphthoquinone	This group is responsible for the production of reddish-brown, orange and red shades. Examples are henna and walnut shell
Tannin	This group is made up of polyphenolic components of tannins. In textile manufacturing, tannin needs mordant and the colour of tannin varies as the mordant varies too. Example; Cutch and Babool

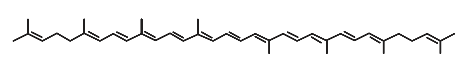
As shown in Table 2 below, the classification of food colourants of natural origin can be extremely complex because of their various unique properties and functional groups they pose. Therefore, distributing various roles at different capacities due to colour shades each provides.

Chemistry of natural colourants

- **Annatto:** Annatto is a natural food colourant with E number E160b, made up of fat and water-soluble part. The bixin is known as the fat-soluble part while norbixin is the water-soluble part. A 4.5- 5.5% of the pigment is contained in the orange-reddish pulp that surrounds annatto seed and bixin content is within 70-80%[30]. Bixin has the carboxylic acid end and the methyl ester acid end which is liposoluble. Norbixinis derived from the de-esterification of methyl ester end which makes norbixin soluble in aqueous alkaline solution[31]. The chemical substances bixin and norbixin give rise to the yellowish-orange colour a type of carotenoid known as Xanthophyll that lacks provitamin A activity. If a pink shade is produced, then an acid-proof version is employed at low pH, the higher the level of bixin in an annatto colour, the more the shade is red and the higher the norbixin level in an annatto colour, the yellow colouration dominates. Bixin or norbixin can be applied in the absence of saffron[32]. Another name for annatto is Roucou; it has a slightly peppery scent, a nutmeg hint and a slightly peppery sweet flavour[32]. It is mostly used to manufacture food colourants that are red and also serves as a flavouring. It is derived from the Achiote tree in the tropical areas of America. Foods applied include; custard powder, rice but ter, cheese, margarine[33]. Its application is banned in the EU, while it is termed exempt in the United States.

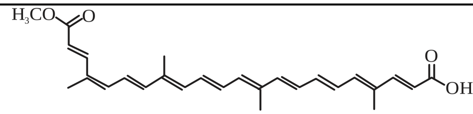
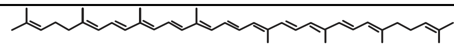
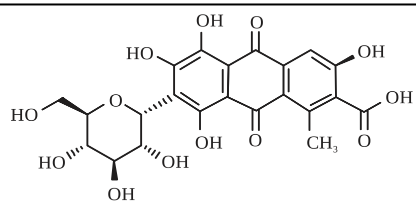
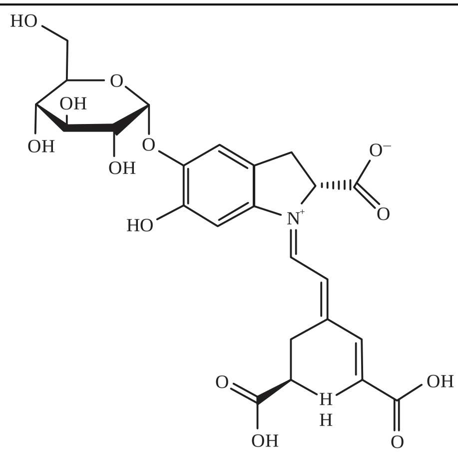


- **Lycopene:** Lycopene is a natural food colourant, extracted from tomato whose colour is determined by the concentration; when in a diluted solvent, it gives orange colour but when together with the tomato fruit peel, it gives a bright red colour which is an attribute of carotenoids[31]. Water-dispersible, oleoresins and powders of lycopene preparations are stable at the anaerobic condition in the presence of wide pH level, temperature and absence of light. Tomato lycopene extract and concentrate are approved as a colourant[34]. Lycopene can also be derived from fungus Bacillus Tripura and has been approved in the EU.

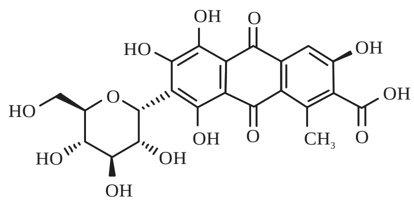


- **Carmine:** Carmine is a natural food pigment of bright red colour extracted from dry insects scale known as cochineal[35], with E number E120[36]. This pigment is chemically classified as anthraquinones. The major pigment contained in cochineal is C-glycoside (>95%) and carminic acid. Other names for carmine include cochineal, due to its source (cochineal insect), Natural Red 4, crimson lake, E120 and C.I. 75470. It is prepared by boiling of the dried insects (cochineal) in water to get a clear solution, extraction of the carmine acid via treatment with alum, potassium hydrogen oxalate, cream of tartar or stannous chloride and precipitation of the animal matters present in the liquid. It has a deep red colour appearance, aside from food application like juice,

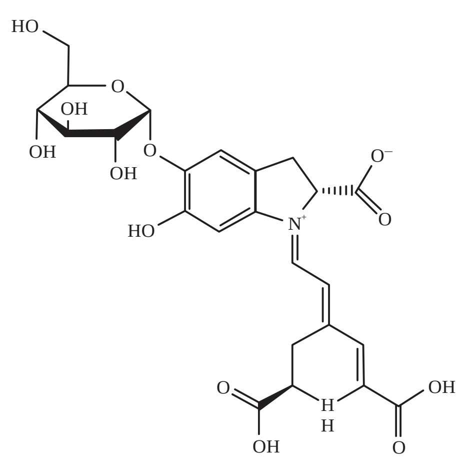
Table 2: Structure, property, application and safety of natural food colorant

Colourants	Structure	Property	Application	Safety
Annatto		Annatto is a natural food colourant with E number E160b, made up of fat and water-soluble part. Its powder is stable at anaerobic condition at a wide pH level	It is mostly used to manufacture food colourants that are red and also serves as flavourant in custard powder, rice but ter, cheese, margarine	Banned in some countries like EU and US
Lycopene		Lycopene is a water soluble natural food colourant, extracted from tomato	It is mostly applied in rice, sauce and soups as an extract in food products	Approved and safe
Carmine		Carmine is a bright red colour extracted from dry insects scale known as cochineal. It contains majorly C-glycoside (>95%) and carminic acid. Its E number is E120. This pigment is chemically classified as anthraquinones. Mostly affected by degree of light that passes through it	Its application is mostly seen in juice, yoghurt. Also, can be used as staining agent, industrially for the production of paints, range, cosmetics, crimson ink	Approved and safe but allergic individuals should stear clear.
Betalain		This is a yellow and red-indole natural colourant derived from plants known as the Caryophyllales. It is classifeid into Betaxanthins (yellow to orange at 480nm adsorption) and Betacyanins (red to violet at 538nm adsorption)	Mostly applied in fruit juices, dairy products among others	Safe and approved
Caramel		Caramel is a natural colourant derived from a conditioned heating of sugar with a food-grade salt, acid or alkali. Its E number is E150, has bitter taste and dark in colour	Mostly applied in soft drinks, beverages and dairy products	Safe and approved

yoghurt, it is also applied industrially for the production of paints, range, cosmetics, crimson ink. Also, it can be applied in the microbiologically with a mordant as a staining agent. People who are allergic to carmine or have decided not to ingest any food of insect origin such as vegetarians suffer allergic reactions and anaphylactic shock[37]. Carmine is mostly affected by; the degree of light passage during its precipitation, temperature, sunlight for the yielding of brilliant blue colour.



- **Betalain:** This group of natural colourants mostly derived from plants known as the Caryophyllales. They are yellow and red-indole colourant, mostly replaced by fungi pigments of higher-order[38] and anthocyanin. The name "betalain" is a Latin word for common beet known as "*Beta vulgaris*" which is the first source for betalain extract. The presence of betalain pigments gives amaranth, bougainvillaea and cacti it is deep root colour. Betalains, according to plant physiologist may possess some fungicidal functions in plants in which it is contained. It is mostly found in some part of the plant such as flowers petals, stems, leaves, roots and fruits. Each group of betalain is made up of a coloured part and sugar part. They are all glycoside which sunlight promotes its synthesis, as well plays a role in the cardiovascular health of humans[39]. Its unique shade of purple to red is also available in plants which attract animals to encourage pollination and seed dispersion.



Betalains are classified into two groups:

Betaxanthins: This class of betalain are mostly present in the plant. Their colour shade varies from yellow to orange with maximum absorption at 480nm. Examples are; portulaxanthin, vulgaxanthin, miraxanthin and indicaxanthin.

Betacyanins: These are betalain pigments with the colour shades that vary from red to violet with maximum absorption at 538nm[40].

- **Caramel:** Caramel is a natural food colourant, derived from properly controlled heating of carbohydrates mostly in the presence of food-grade acids, salts or alkali. It has E number E150, a dark appearance and a bitter taste. Caramels are the major component of all food colourant[41] and are classified into four. The classification is based on the reactants applied during the production process. The reactants include sulphite compounds and ammonium compounds. The first class (E150a) is obtained by heating of carbohydrate with either acid or an alkali only and it is the lightest shade of caramel. The second class (E150b), third class (E150c) and fourth class (E150d) are derived through the addition of ammonium, sulphite and both group of compounds[42]. In soft drinks, the fourth class of caramel is mostly applied because of its darkest shade[41]. Caramel without 5-hydroxymethyl-2-furfural has been proposed to prevent health-related concerns on caramel (E150c and E150d)[31].

Analysis in light of the structure of synthetic food colourants: Due to the negative impact of most synthetic food colourants on humans, sensitive and very efficient analytical methods have been devised to assist in detected in foods for human safety. The most prominent method for the analysis and detection of their presence in foods includes; thin-layer chromatography, paper chromatography, HPLC with diode-array detection (DAD) system and capillary zone electrophoresis[43]. Synthetic colours in food products mainly are triarylmethane and azo dyes which are either acidic or anionic comprising of sulfonic acid, a hydroxyl group or carboxylic acid[44,45]. At basic pH, these compounds form negatively charged ions. In the azo group are; blue-black, red, orange, yellow and brown colour shades while the triarylmethane colour group are those synthetic colours which in the presence of light has high colour brilliance, tinctorial capacity and poor stability. Of all the aforementioned analytical methods, capillary zone electrophoresis (CZE) serves as an ideal tool for the analysis of synthetic colours due to its ability to separate compounds within a short run into different functional groups depending on their wavelength, charge and size[45]. Therefore, to ensure that these harmful synthetic colourants are not applied in food products and that the ones used conform to the standard, these analytical methods should be applied for detection of the nature and concentration of a given colourant in a food product for safety purposes.

Health implications of synthetic food colourants: Assessment of food colourants in terms of health risk comes with different views because not all synthetic food colourants are harmful to man. However, it has been observed that synthetic food colourants harm humans including children's behaviour as hyperactivity, behavioural disorders, attention deficits among others which could be attributed to the azo-benzene compound present in most synthetic food colourants as revealed by their structures above which affects most important organs, genes and alter metabolic activities. The relationship between the structure of the discussed synthetic colourants and their risk to human health lies in the presence of diazotized sulfanilic acid and phenolic sulfonic acid moiety that contains undesired by-products from corresponding impurities. A study performed by Sahar and Manal[46] on the effect of usage of food colourant (Colour fruit juice for 6-12 h) on the serum biochemical, as well as the kidney and liver of rats for 13 weeks. Tomato ketchup potato chips, TKPC (30%) confirmed a tremendous boom in normal LDL cholesterol (TC) and Triacyl-Glycerol (TG). The degree of Alanine Transaminase (ALT) and Aspartate transaminase (AST) changed the growth of rat's administration of colour fruit juice (for 12 h) and TKPC largely at 30%. There was additionally a tremendous growth in albumen and serum creatinine. Both low and excessive consumption of coloured foods exhibited a massive reduction in liver glutathione (GSH). The study additionally showed that excessive concentration of colour ingredients led to white blood cell increase as the outcome to the reaction of the immune to inflammation. Their findings confirmed that fruit juice containing sundown yellow, tartrazine and carmoisine increase ALT of serum rats. Therefore, they concluded that the artificial colours used in their studies have detrimental effects on a number of the serum biochemical, liver and kidney. These effects had been nicely supported[30], who indicated that rats which ate up an excessive dose of artificial colourant (Tartrazine, Carmoisine, Sunset Yellow and Fast Green) confirmed a massive growth in serum ALT and AST compared to control rats. The boom in serum ALT and AST may also characteristic those changes in liver function to be hepatocellular impairment degree of intracellular enzymes into the blood[10]. It was obvious in the histopathological studies. At low dose artificial colour, the liver showed a disruption of hepatic cells close to the vital vein and hepatocellular damage. A report by Okafor *et al.*[30], stated that artificial colours have a hazardous impact on critical organs. The launch of a normally excessive stage of precise tissue enzymes into the bloodstream is depending on both the level and kind of harm exerted by the poisonous compound administration[47,48]. Furthermore, there has been a considerable rise in serum creatinine. It is believed that the extremely good elevation in creatinine stage is closely related to the impairment of renal interest. There was a great increase in serum creatinine and urea level of rats doses with azo dye (fast green) orally for thirty-five days. The class of rat administrated with chocolate and candy coloured at low and high concentrations witnessed a vast boom in triglycerides. These consequences once more are by the results received with the aid of Floriano *et al.*[16], who located extensive increase in serum triglycerides of rats treated with artificial colour (tartrazine) and chocolate colour A and B that contains tartrazine and Carmoisine. The cytotoxicity of 11 dyes, used as meals dyes in Japan, on cultured fetal rat hepatocytes was evaluated[30]. Xanthene dyes containing halogen atoms of their molecules which incorporates phloxin, rose bengal and erythrosine has been more toxic than every other sort of meals dyes. The influence of food dyes on the cellular increase of hepatocytes changed into also was tested. The high hepatotoxicity of phloxin to the cellular increase, which was dose-established, become determined while the dye was added three days after plating. A double-blind placebo-controlled high dose azo dye task in a chosen group of kids with behaviour disturbance suggested a small damaging impact on the kid's behaviour based totally on ratings on the Connor scale[16]. They concluded that children might gain more if synthetic food colourings are eliminated from their eating meal. The research performed at the Center for Science in the Public Interest (CSPI), on meals dyes discovered that some of the most frequently used meals dyes could be connected to numerous types of cancer. CSPI suggested: "The three maximum widely used dyes, Red 40, Yellow 5 and Yellow 6, are contaminated with identified cancer-causing agents. Another dye, Red 3, has been mentioned for years by the Food and Drug Administration to be a carcinogen, yet continues to be within the food supply." In their fifty-eight-page report, "Food Dyes: A Rainbow of Risks" CSPI discovered that nine of the meals dyes whose application is presently authorized in the United States are connected to health issues starting from cancer and hyperactivity to hypersensitive reaction-like reactions. For example, Red no. 40, which is the extensively used dye, might also boost the growth of immune system tumour in mice and triggering hyperactivity in children. Blue no. 2, applied in sweets, beverage, animal feeds and more is related to brain tumours. Yellow 5, utilised in baked products, sweets, cereal and extra, will not best be contaminated with numerous cancers-inflicting chemicals[13] but it is also linked to hyperactivity, hypersensitivity and other behavioural effects in youngsters. As in placebo-managed research performed in 2007 and published in The Lancet journal[49], they critically evaluated the outcomes of usual meal dyes determined in lots of soft drinks, fruit juices and salad dressings. The results showed that dyes studied made some children more hyperactive and distractible. As a support to the findings in Lancet, a study in the Annals of Allergy[22] discovered that 73% youngsters who were suffering from Attention-Deficit Hyperactivity Disorder (ADHD) reacted favourably to food without synthetic colour. The Lancet observed that E-numbered meals dyes cause as lots of damage to the children's brains as the lead in petrol, which causes a reduction in their IQ. At the wake of these findings, the British Food Standards Agency (FSA) issued advised parents to limit their youngster's intake of food additives. FSA additionally suggested the food industry to voluntarily exempt the six food dyes named in the research back in 2009 and replace them with natural alternatives if possible. UK food dyes on which the Food Standards Agency has called for a voluntary ban are Tartrazine, Quinoline Yellow, Sunset Yellow, Carmoisine, Ponceau 4R and Allura Red[13].

The mechanism for the negative health effect of synthetic food colourant: Looking at the toxicogenic tendencies of these colourants, the ability of a food colourant to cause harm to the body depends on the absorption of the colourant into the human system through the gastrointestinal tract; which is affected by factors such as the size of the molecules[50]. The larger the molecular size of the particles of the food colourant, the lesser its ability to pass through the mucosal wall, hence has fewer chances of causing harm, while the molecules with lower molecular size easily pass through a mucosal wall and thus has a high rate of absorption compared to the high molecular-sized ones[50]. The higher the absorption rate, the higher the toxicity of the food colourant, the smaller molecules should be bonded with carrier molecules. Therefore, the smaller molecules are bonded with carrier molecules which makes the smaller molecules (chromophores) large and difficult to be absorbed into the body, with the colouring properties of the smaller molecules of the food colourant not affected. Colourants such as tartrazine (yellow 5), sunset yellow (yellow 6) have a by-product after being broken down by gastrointestinal bacteria, hence cause a negative health effect on the consumer[51]. This detrimental effect depends on the level or degree of exposure which implies the degree or level of colourant ingested into the body[50]. Due to high stability of azo dyes when exposed to external factors like sunlight (light), pH, O_2 among others, over the natural ones, a check on the level of its inclusion in meals should be properly done. The negative health effect of azo dyes on humans could be attributed to its water solubility and rate of metabolism at the liver and kidney carried out by azoreductase enzyme that breaks the azo linkage, thus makes them easily secreted out of the body via urine[52].

Health benefits of natural food colourants: Natural food colourants play a crucial function in human fitness as they contain a few biologically active compounds, which has some pharmacological functions like anti-inflammatory, antiarthritic, antimutagenic and antioxidant effects[19,20]. Carotenoids are also used as diet supplements, given that β-carotene is the precursor of vitamin A. The everyday consumption of carotene can help prevent nighttime blindness as a consequence of an inadequate supply of vitamin A. Carotenoids also act as organic antioxidants, defensive cells and tissues from the damaging outcomes of loose radicals and singlet oxygen and additionally as a great source of the antitumor agent[31].

Lycopene is mainly powerful at quenching the damaging ability of singlet oxygen[30]. Lutein, zeaxanthin and xanthophylls are believed to feature as protecting antioxidants within the macular vicinity of the human retina, which additionally acts in opposition to growing old, macular degeneration and senile cataract, antiplatelet, antihypertensive, anti-atherosclerotic and anti-inflammatory effects[34]. Canthaxanthin also exhibits antioxidant property, while Astaxanthin is taking the place of xanthophyll with amazing antioxidant properties[53].

Food phenolic compounds, particularly flavonoids, are thought to play crucial roles in human health. A range of research, each in vivo and in vitro studies have validated that flavonoids have antioxidant and antimutagenic activities[30] and could be very useful within the reduction of the risk of growing cardiovascular disorder and stroke[31]. Flavonoids may additionally act as antioxidants to inhibit unfastened-radical mediated cytotoxicity and lipid peroxidation, as antiproli ferative retailers to inhibit tumour, growth or as weak estrogen agonists or antagonists to modulate endogenous hormone activity. Flavonoids had been categorised as excessive level natural antioxidants on the premise in their abilities to scavenge free radicals and energetic oxygen species[31] as result of the hydroxyl groups and the conjugated ring gadget via halogenation or complexing with these oxidising species. In those ways, flavonoids may confer protection towards persistent illnesses including atherosclerosis and cancer and assist the control of menopausal signs. Thus, flavonoids have been called semi-critical food components[30]. Consumption of quercetin can also protect in opposition to cardiovascular disease, by way of reducing capillary fragility and inhibiting platelet aggregation[31]. Several flavonoids including catechin, apigenin, quercetin, naringenin, rutin and Norton are suggested for their hepatoprotective activities[19]. Anthocyanins have drawn growing attention because of their preventive impact in opposition to various diseases[31]. Zhu *et al.*[54] validated that anthocyanin cyanidin-3-O-β-glucoside (C3G) increases hepatic glutamate-cysteine ligase catalytic subunit (GCLC) expression with the aid of growing cyclic adenosine monophosphate (CAMP) tiers to set off protein kinase A (PKA), which in flip upregulates cAMP response element-binding protein (CREB) phosphorylation to enhance CREB-DNA binding and growth GCLC transcription-increased GCLC expression results in a decrease in hepatic ROS degrees and proapoptotic signalling. It was also shown that the C3G remedy reduces hepatic lipid peroxidation, inhibits the release of proinflammatory cytokines and protects against the development of hepatic steatosis[54,55].

Other health benefits of natural colourants encompass increment of the immune system, safety from sunburn and inhibition of the development of certain forms of cancers[31]. Epidemiological research has discovered a correlation between the intake of chlorophylls and decreased hazard of colon cancer[56]. They have also been observed to own antineoplastic, radiation-shielding, vasotocin, vasoprotective, anti-inflammatory and hepatoprotective activities. A flavoured and brightly coloured seed of *Manikara obovate* has been confirmed to have each antioxidant and hepatoprotective activities[57].

Stability of natural colourants: Natural food colourants abound with potentials outside its ability to yield the desired colour to foods but with minor limitations which includes; cost, quality, sensitivity to pH, oxygen, temperature, UV and light, method of extraction, complex handling, application, process and production technique as well as technical know-how, accessibility of raw material and shelf instability[58]. Degradation of natural colourants during storage such as, photobleaching of curcumin, heat sensitivity of beetroot and anthocyanin which undergoes degradation by forming an undesirable brown pigment leads to loss of food products. Extraction of colour components from raw materials are labour intensive, capital intensive, high risk of microbial contamination and time consumption. There is always need for facilitation usually by the addition of a metallic compound known as mordant which reacts with the colourant by chelation due to the elusive nature of natural colourants, although some of the metallic mordants are detrimental to health[58]. Therefore, Good stability in natural food colourants can be achieved through the lowering of water activity, addition of an acyl group of sugars with cinnamic acids, the addition of maltodextrins extract from Roselle and dextrins extract from tart cherries[9]. Selection of genetically modified plants, high-yield strains of micro-organism and insects and use of fermentation as a unit operation during natural colour production will reduce cost, time of production and increase the yield of colourants with good stability, easy and uniform dispersion in the food matrix. Colours such as *D. saline*, *B. trispora*, spirulina and meniscus which are in use today and are produced by fermentation method[58]. Additionally, application of micro-encapsulation, nano-emulsions or nano-encapsulation and nano-formulations technologies can improve the stability, solubility and safety of natural pigment in foods during production in food industries.

Applications of natural food colourant in the food industry: Curcumin is used in dairy products, frozen desserts, ice-creams, confectionery, mustards, meal concentrates. It is also added to margarine and seasoning and mayonnaise sauces as a mixture with annatto[9]. Plant coal is derived from the wood charing process. It is used as a black pigment in the food industry. It possesses neither smell nor taste. It is highly resistant to light activity, as well as Sulphur-dioxide (SO_2), heating, chemical and physical agents. It is stable in the wide range of pH: from 2-10. The coal can be dissolved neither in organic solvents nor in water. The pigment is used to either giving colouring or shading. It is also used in wine, vinegar and juices purification process[9]. Heated sugar of a tawny colour and specific taste and smell is called caramel. It is used in food as a natural pigment, despite the fact it does not occur in plants or animals but is obtained by heating of sugars. Glucose, fructose, saccharose, polymers of the substances and starch syrup are used. Adequate pressure and temperature conditions must be provided in the production process of caramel. The whole process may be accelerated by the application of phosphates, carbonates and sulfites. Symbols E 150a-d marks caramels. Various types of caramel are used for dying in Poland [59]. Chlorophyll is found, in the food industry, in vegetable and fruit preserves, alcoholic drinks, or cheese spreads[9,59]. Carotenoids are used in the production of; butter, margarine, oils and fats, cheese spreads, non-alcoholic drinks, fruit juices, confectionery baked goods, ice-creams, yoghurts, desserts, jams, creams, pastries, jellies. Anthocyanin application of the group of flavonoids by the food industry is a common practice and they can be seen in; fruit, alcoholic and non-alcoholic drinks, sauces, cheeses, milk desserts, jellies, jams, candies[9,59]. Cochineal is used to colour milk desserts, alcoholic and soft drinks, jams, tomato preserves and also to various confectionery and bakery products[9,59]. Betalains are used to colour, e.g. frozen food, ice-creams, flavoured milk drinks, yoghurts, powdered desserts, gels, sauces, jams, jellies, candies[59-61].

Advances: Nowadays, other recent technological approaches have been designed to yield food colourants. Presently, Carotenoids are not just extracted from vegetable sources (plant roots, leaves, flowers among others) alone but from aquatic animals and micro-organisms (algae, fungus, yeast). For example, astaxanthin (E161j) is isolated from animal sources, while β-carotene (E160a) maybe both extracted from the roots of *Daucus carota* L. and even from fungus (Blakeslea trispora). The most common colour attributes of carotenoids are yellow to orange and even red colour. As previously highlighted, lutein (E161b) and astaxanthin (E161j) are the carotenoids most commonly used for pharmaceutical and nutraceutical purposes, being used not only to confer bioactive and functional properties but also colourant attributes[19].

Additionally, to enhance the production of natural pigments form micro-organisms, the following methods has been designed; Newly Developed Smarter Screening and identification Methods through the use of condensed handheld Raman spectrometer, mass spectrometry with electrospray ionization, HPLC, UV-VIS spectra and nuclear magnetic resonance (NMR) among others can be applied for rapid identification and purification of individual compounds. Use of Strain Development through random mutagenesis and multiple selections, application of Fermentation tanks for increased production of colourants. Cost-Effective Downstreaming, Metabolic Engineering which involves the manipulation of those genes responsible for biosynthesis and increased production of colours by cloning, Metabolic engineering using the Clustered, regularly interspaced short palindromic repeats (CRISPR) -CAS9 SYSTEM. These colourants derived, outside their colouring capacity has so many health beneficial effect as thus; anti-diabetic, anti-cancer, anti-cholesterol, anti-inflammatory as reported by Wang *et al.*[62] and Patakova[63] although no E code has been assigned to them.

CONCLUSION

Application of food colourants is very vital to food industries in the production of food products such as; soft drinks, canned products, vegetable products, confectionery, baked products, fish and meat products and others in other to win their customers. Due to the change in consumers nutrition habits, since the health of the consumers are at risk due to the application of synthetic food colourants, there has been increasing demand and popularity of natural food colourants among the consumers all over the world because of the health benefits and safety derived from the natural colourant, unlike the synthetic colourant that has detrimental effects as hypersensitivity reactions, carcinogenicity, toxicological effect and behavioural effect on man and can't be considered safe. Food companies should embrace the use of natural food colourants which is of benefit to man as well as safe to be consumed while the consumers adhere strictly to the consumption of naturally produced foods only. Also, synthetic food colourants and synthetically made food products should be banned by food and drug regulatory agencies of various countries as obtains in Japan and all European countries.

Furthermore, since consumers demand is rapidly tending towards naturally made food products, researchers should look critically into making natural food colourants readily available and improving its consistency, to enable its maximum application in foods.

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