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Natural colorants from vegetable food waste: Recovery, regulatory aspects, and stability—A review

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Abstract: Worldwide, approaches inspired by the Circular Economy model have been increasing steadily, generating new business opportunities such as the recovery of high-added value molecules (e.g., pigments) from vegetable food waste that may be applied as food additives (e.g., colorants). Indeed, food waste is a global problem that does not seem to be decreasing, leading to economic, environmental, and social issues. Moreover, synthetic dyes have been associated with adverse effects on human health, encouraging research to explore much safer, natural, and eco-friendly pigments. This state-of-the-art review gives a brief overview of the regulatory aspects concerning food waste, Circular Economy, and natural versus synthetic colorants. We have critically reviewed the recent advances in pigment recovery from vegetable food waste bringing back the green/unconventional extraction methods. Among them, enzyme-assisted extraction as a depth feature technique is highlighted, given that it allows the recovery of pigments in a mild, selective, efficient, and sustainable way. Furthermore, the stability issue of the different natural colorants has been critically discussed in relation to the extraction and application conditions. Several and tailored stabilization methods have been described and reported for each pigment although additional research is necessary on their long-lasting stabilization and utilization in food matrices.

KEYWORDS
Circular Economy, enzyme-assisted extraction, food waste, pigment from vegetables, sustainable routes

Practical Application

This review focuses on the main types of natural pigments in vegetable food waste, their legislative framework, extraction technologies and strategies to improve the stability , as well as their possible applications.

1 | INTRODUCTION

Food waste (FW) is a global issue that does not appear to decrease, leading to economic, environmental, and social questions (Teigiserova et al., 2020). This problem has long been neglected, in such degree more and more waste is recorded at all levels of the food chain (Peira et al., 2018). The European Union defines FW as: "all the foodstuffs discarded from the food supply chain for economic or aesthetic reasons or owing to the nearness of the 'use by' date, but which are still perfectly edible and fit for human consumption and, in the absence of any alternative use, are ultimately eliminated and disposed of, generating negative externalities from an environmental point of view, economic costs and a loss of revenue for businesses" (European Parliament Resolution, 2012).

FW represents a loss of edible food that is discarded from the supply chain by free choice and/or negligence by its actors (Peira et al., 2018) throughout distribution and consumption phase (wholesaling, logistic, retailing, market, food service, and household) (Parfitt et al., 2010). Recent studies have reported that global FW is 1.6 billion tons/year, of which 1.3–1.6 billion tons/year is edible, for a business value of US $750 million. A common family produces most of the FW (about 47 million tons/year), succeeded by the processing phase (17 million tons), catering (10.5 million tons), primary production (9.1 million tons), and distribution (4.6 million tons). FW mainly consists of meat and fish (30%), vegetables (19%), and dairy products (17%), amounting to US $27 billion (Peira et al., 2018). It has been assumed that the lack of initiatives to reduce FW could increase it by about 40%, corresponding to 126 million tons/year (Peira et al., 2018). An effective waste management may be achieved only throughout a valuable food chain control and a better use of the limited resources available (Battilani, 2015). Keeping in mind Circular Economy concepts, recent developments have been focused on the utilization of vegetal wastes as source of natural pigments.

2 | CIRCULAR ECONOMY APPROACH ON FOOD WASTE

One of the pillars of sustainable development is the Circular Economy model. The transition from a linear economy model to a circular one means that resources remain in the economy even after the end of life of materials and products. In detail, Circular Economy may be defined as a model of production/consumption, which involves sharing, reusing, and recycling products as long as possible, thus minimizing waste. The Circular Economy can define the background requirements for the management of FW and other kinds of waste, generating new business opportunities. FW may be applied to produce several biomaterials (Mirabella et al., 2014), bioenergy (Dahiya et al., 2018), and high-value products (Galanakis, 2012; Sagar et al., 2018). Taking into account the definition of waste by Circular Economy (waste = secondary resource), EU has approved an action plan to incorporate it into Member States legislation (Directive EU, 2018/851). The FW legislation in Europe has always been unclear. The key EU directive, Waste Framework Directive (European Parliament. (2008), Directive EU, 2008/98/EC), does not distinguish FW from other kinds of bio-waste (such as garden waste) (Teigiserova et al., 2020). In this scenario, a Circular Economy program was drawn up to manage food waste in order to reduce it as much as possible. This model is explained by means of a pyramid chart (Sanchez et al., 2020) which shows actions that should be implemented (Figure 1).

This pyramid describes the optimal strategies, focusing first on prevention actions, followed by the paths of reusing of surplus food suitable for human consumption, the reuse of food no longer intended for human consumption as animal feed, recycling/recovery of high-added value molecules (e.g., natural colorant), nutrient recycling, energy recovery and, as the last option, food waste disposal (Figure 1) (Sanchez et al., 2020; Teigiserova et al., 2020). In particular, regarding the recovery of high-added value molecules, the scheme in Figure 2 shows five distinct phases. The method (general and flexible based on the matrix and the molecule to be extracted) proceeds from the macroscopic to the macromolecular level, followed by purification of the target compounds and subsequently to the extraction of specific micromolecules (Figure 2) (Galanakis, 2012).

Aiming to minimize the environmental impact of waste supporting the Circular Economy concepts, the great challenge for the future is the exploitation of natural additives from FW. The industrial use of such additives (including natural pigments) has been increasing in the last decades due to the consumers' demand for healthy food (Martins et al., 2016).

3 | FOOD ADDITIVES AND COLORANTS

Food additives present a long history of use and more specifically food colorants are a good example. The former are currently defined as "any substance not normally consumed as a food by itself and not normally used as a typical ingredient of the food, whether or not it has nutritive value, the intentional addition of which to food for a technological (including organoleptic) purpose in the manufacture, processing, preparation, treatment, packing, packaging, transport or holding of such food results, or may be reasonably expected to result (directly or indirectly), in it or its

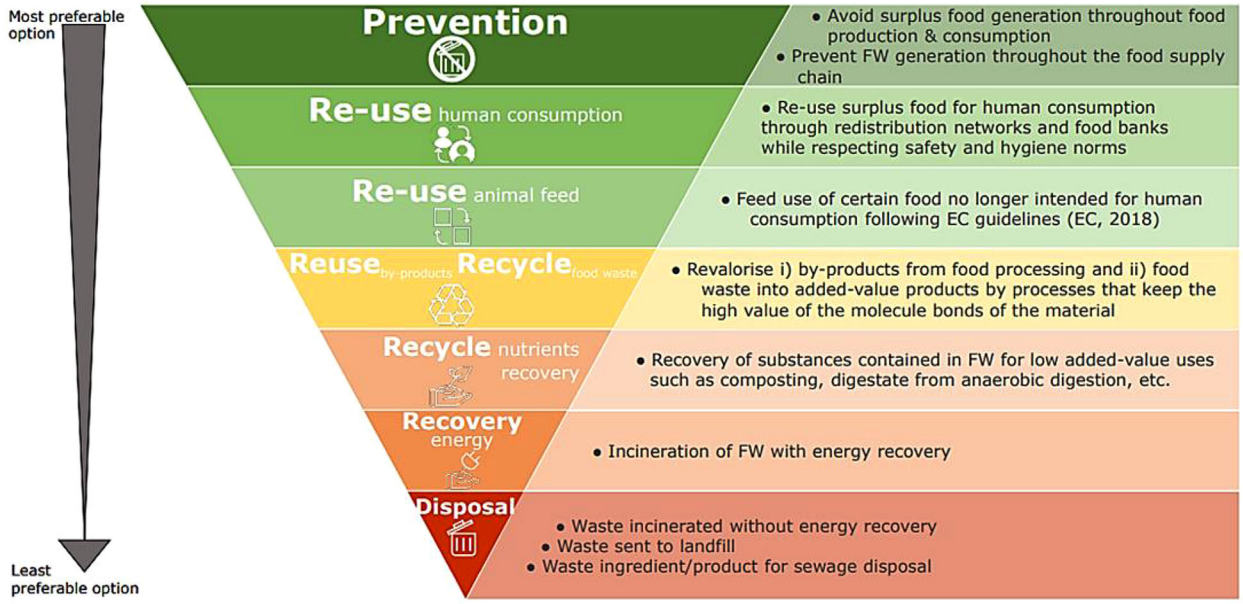

FIGURE 1 Practical application of the waste hierarchy for food. *Source*: Sanchez et al. (2020)

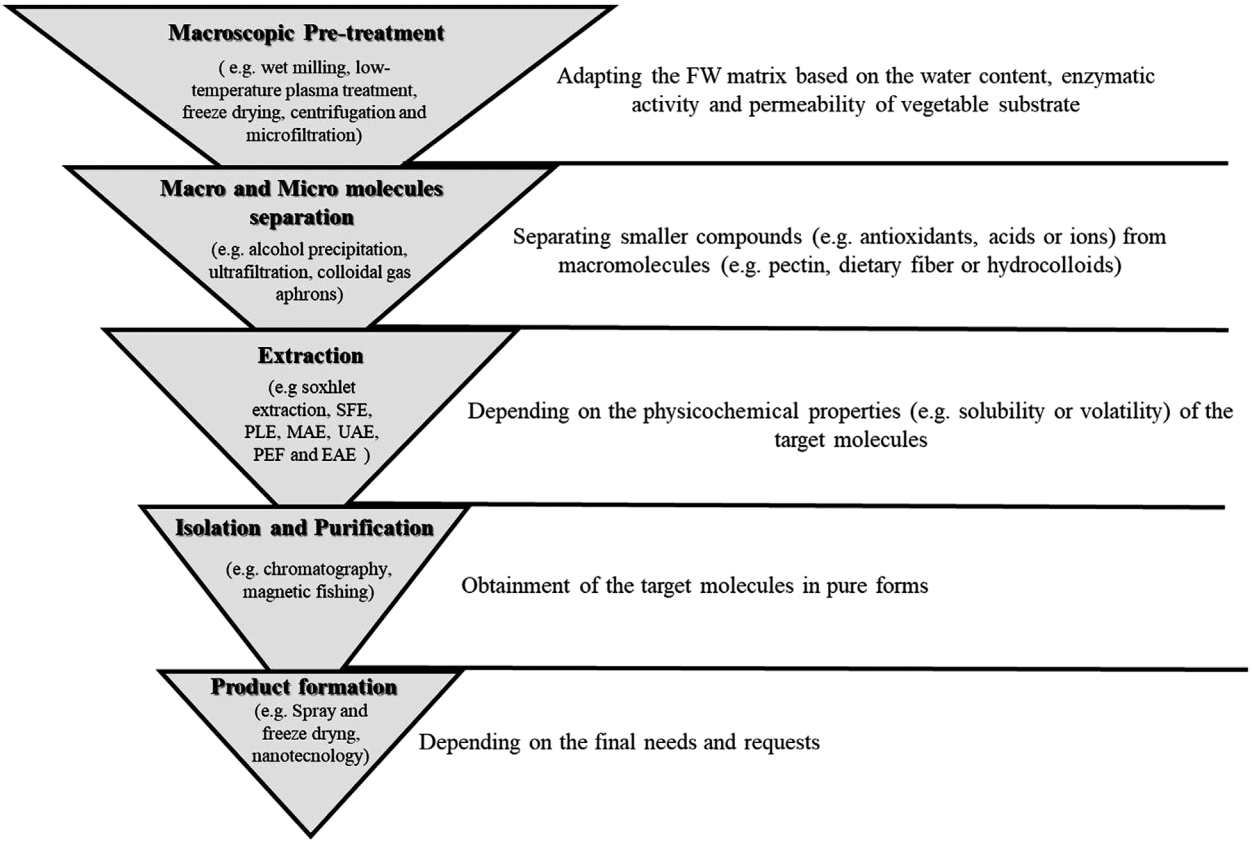

FIGURE 2 Recovery phase of high-added value compounds from food wastes (FW). Abbreviations: EAE, enzyme-assisted extraction; MAE, microwave-assisted extraction; PEF, pulsed electric field; PLE, pressurized liquid extraction; SFE, supercritical fluid extraction; UAE, ultrasound-assisted extraction

by-products becoming a component of or otherwise affecting the characteristics of such foods. The term does not include contaminants or substances added to food for maintaining or improving nutritional qualities" (Codex Alimentarius Commission, 2017). Worldwide, the two main regulatory bodies for food additives are the European Food Safety Authority (EFSA) and the United States Food and Drug Administration (FDA) (Carocho et al., 2014). A huge effort has been made to assemble knowledge for the creation of a unique database of legal additives for use within the European Union. In Regulation 1129 of 2011, all approved additives and their acceptable daily intake (ADI) have been listed (Council Regulation (EC) 1333/2008; Council Regulation [EC] 1129/2011). In the United States, starting from 1961, the FDA ruled that all food components should be labeled as generally recognized as safe (GRAS). In the European Union, food additives are classified into 26 functional groups, according to their function in food (Council Regulation, 1333/2008), whereas in the United States they are ranked in six classes (Carocho et al., 2014). In addition to these classifications, additives, including food colorants, may be categorized according to origin and manufacture, dividing into four groups: natural additives (from animal or plant sources), similar to natural additives (mimicking natural ones), modified from natural (natural additives which are then chemically modified), and artificial additives (synthetic compounds) (Carocho et al., 2014).

In details, food colorant is defined as "any dye, pigment or substance which when added or applied to a food, drug or cosmetic, or to the human body, is capable (alone or through reactions with other substances) of imparting color" (Martins et al., 2016). Legislation regarding food colors differs from country to country.

3.1 | Synthetic food colorants

Colorants or dyes are applied as food additives to change or give color to food, in order to improve its appeal to consumers (Sigurdson et al., 2017). Synthetic colorants may be produced by full chemical synthesis or modification of several natural precursor compounds (E160 carotenoids, E161 xanthophylles, E162 betanine, E163 anthocyans, E140-E141 chlorophylls). Although dyes have long been used by the food industry, there are a number of controversies and disagreements over their potential effects on human health (Branen et al., 2001; Msagati, 2013). Legislation regarding food colors differs from country to country. For example, amaranth (E123), carmosine (E122), and others are banned in the United States but not in the European Union, while fast green (FD&C Green No. 3) is banned in the European Union and lawfully added to food in the United States. Food colorants may be categorized into five groups, according to their chemical structure: azo-compounds, quinophthalone derivatives of quinoline yellow, triarylmethane group, xanthenes, and indigo dyes (Sarikaya et al., 2012).

Among azo compounds, some of the most used are tartrazine, known in the United States as FD&C Yellow No. 5 and in the European Union as E102, sunset yellow (USA: FD&C Yellow No. 6; EU: E110), allura red (USA: FD&C Red No. 40; EU: E129), amaranth (E123), and carmosine (E122) (Carocho et al., 2014). The European Union has revised its opinion on allura red, concluding that there is a possibility that it may be genotoxic; however, some existing studies indicate the opposite (Carocho et al., 2014). Amaranth, another petroleum-derived dye, has been banned in the United States as possibly carcinogenic, but it is approved in the European Union and some other countries. Azo compounds are commonly added in blancmange, marzipan, Swiss roll, jam, yoghurt, jelly, breadcrumb, cotton candy, soft drinks, flavored chips, cereals (corn flakes, muesli, etc.), cake mixes, soups, sauces, some rice, ice cream, candy, and chewing gum (Amin et al., 2010). Quinoline Yellow (E104) is a synthetic quinophthalone dye chemically obtained by mixing sodium disulfonates, monosulfonates, and trisulfonates and is usually used in soft drinks, cakes, cosmetics, and medicines (Al-Shabib et al., 2020). Triarylmethane group includes the following: bright blue (USA: FD&C Blue No. 1; EU: E133), fast green (E143), patent blue (E131), and bright black (E151). Bright blue and patented blue are the most common colorants, although the former is banned in the United States. No authorization has been required for bright black, while fast green is banned in the European Union. These compounds are generally added in yogurt, ice cream, drinks, and sweets (Unsal et al., 2015). Xanthene group is composed of erythrosine (USA: FD&C Red No. 3; EU: E127), fluorescein, eosin, and rhodamine. Erythrosine is permitted in the United States and the European Union, while fluorescein is prohibited in the European Union. They may be found in cocktails, tinned cherries and fruits, biscuits, chocolates, garlic sausages, salmon spreads, scotch eggs, stuffed olives, sweets, bakery items, snack foods, chewing gums, jellies, wines, and ice cream (Tabara et al., 2011). Finally, indigo dyes (FD&C Blue No. 2) are synthetic additives, recovered in the past from the *Indigofera tinctoria* shrub. Nowadays, they are chemically obtained and used in gelatins, ice creams, jelly beans, bubble gums, and candies (Deroco et al., 2018).

3.2 | Food colorants from natural sources

As for ever-growing trends in the food coloring industry, the use of natural pigments (natural colorants) has increased in foods and beverages as substitutes for

their synthetic counterparts. This is mainly due to the growing awareness of environmental risks and potential side effects of chemicals used in the synthesis of food colors (Carocho et al., 2014). Additionally, synthetic dyes tend to be replaced in the food industry in order to be in line with current consumer needs. This marketing strategy is in line with the so-called "clean-label" trend.

The use of natural colorants in the European Union is regulated by the Council Regulation (EC) 1333/2008 (ex 94/36/EC), which includes pigments selectively extracted from a source and intentionally added for the purpose of coloring in the preparation of the final food (Reinhart, 2014).

For example, it is possible to talk about the curcumin dye (E100) only when, after selective extraction from the turmeric root, cellulose, waxes, vegetable lipids, and volatile oily compounds are eliminated. However, some natural pigments, in addition to having coloring properties, may have further physiological (e.g., antioxidant) or flavoring characteristics. For this reason, their classification with respect to food law is not always easy; mainly because the requirements for premarketing approval depend on the classification of the substance (Reinhart, 2014). To solve this problem, the EU Council Regulation excludes from the legislation on food additives those pigments that are explicitly used for their aromatic, savory, or nutritional purpose, and when their coloring effect in special food is only secondary (e.g., butter, which may be colored exclusively with carotenes).

In the United States, dyes are regulated through the Federal Food, Drug and Cosmetic Act (FFDCA). Unlike the European legislation, American legislation does not distinguish between colors produced by synthesis and pigments extracted or isolated with or without modifications from vegetable, animal, mineral, or other sources. However, the approved colors may be divided into two groups: certified colors (which are all artificial) and those exempt from certification (which also contain pigments obtained from natural sources, some of which may also be produced by synthesis) (Lehto et al., 2017).

Commonly used natural food pigments include anthocyanins, carotenoids, betalaines, chlorophylls (Figure 3), among others (Miranda et al., 2021). In addition to their dietary application, the consumption of these naturally colored compounds has been associated with a reduction in noncommunicable diseases such as cancer, diabetes, and obesity (Cortez et al., 2017).

Another possible classification criterion of natural colorants is based on their source (e.g., plants, animals, microorganisms, and minerals) (Sigurdson et al., 2017).

Plant-based pigments are the result of biochemical pathways that take place within the plant, resulting in a large number of organic compounds with unique physicochemical properties (Miranda et al., 2021). These colorful compounds are widespread in nature and perform different functions, participating in photosynthetic processes, attracting pollinators and providing protection from predators and solar energy. Porphyrins, carotenoids, anthocyanins, and betalains are the main chemical classes of plant pigments, which selectively absorb some wavelengths of light and reflect others (Sigurdson et al., 2017). As the size of the natural pigment market is growing rapidly, a more sustainable production is needed. For this reason, research has moved toward the enhancement of vegetable waste (fruit and vegetable) and their byproducts (e.g., skins, seeds, or pomace) for the recovery of natural pigments that find industrial application in food, pharmaceutical, and cosmeceutical sectors (Sharma et al., 2021).

Likewise, a wide range of pigments are obtained from different animals, where they perform various functions (e.g., transporting oxygen in the blood, protecting from predators or UV rays and mating) (Aberoumand, 2011).

Microorganisms (bacteria, fungi, and microalgae) produce a variety of different pigments, able to cover almost any shade of the color. For example, blue pigment (phycocyanins) may be recovered by microalga spirulina (*Arthrospira platensis*) and red colorant (carotenoids) by the fungus *Monascus sp*. and bacterium (*Paracoccus carotinifaciens*). Obtaining pigments from a microbial source is commercially and economically promising, due to the relative simplicity in controlling growth conditions and the source availability (Sigurdson et al., 2017).

Minerals have long been used as dyes in food, cosmetics, and art (e.g., viridian). Their hue depends on the chemicophysical properties (CFR, 2016; Sigurdson et al., 2017).

Moreover, natural colorants may also be classified according to their chemical structure (more thoroughly discussed below).

3.2.1 | Anthocyanins

Among flavonoids (secondary plant metabolites characterized by a carbonaceous skeleton formed by the repetition of the C6-C3-C6 unit), anthocyanins (Figure 3) are important water-soluble subgroup pigments that give plants color ranging from bright red to blue (Obón et al., 2009). Their aglycon form, anthocyanidin, is commonly found in fruits and vegetables and differs in the degree of hydroxylation and methoxylation. Anthocyanidins can be bound to sugars and undergo further acylation processes (with aromatic or aliphatic acids).

The stability of anthocyanins considerably increases both with the degree of glycosylation and with acylation, which explains why simple anthocyanins are rarely found

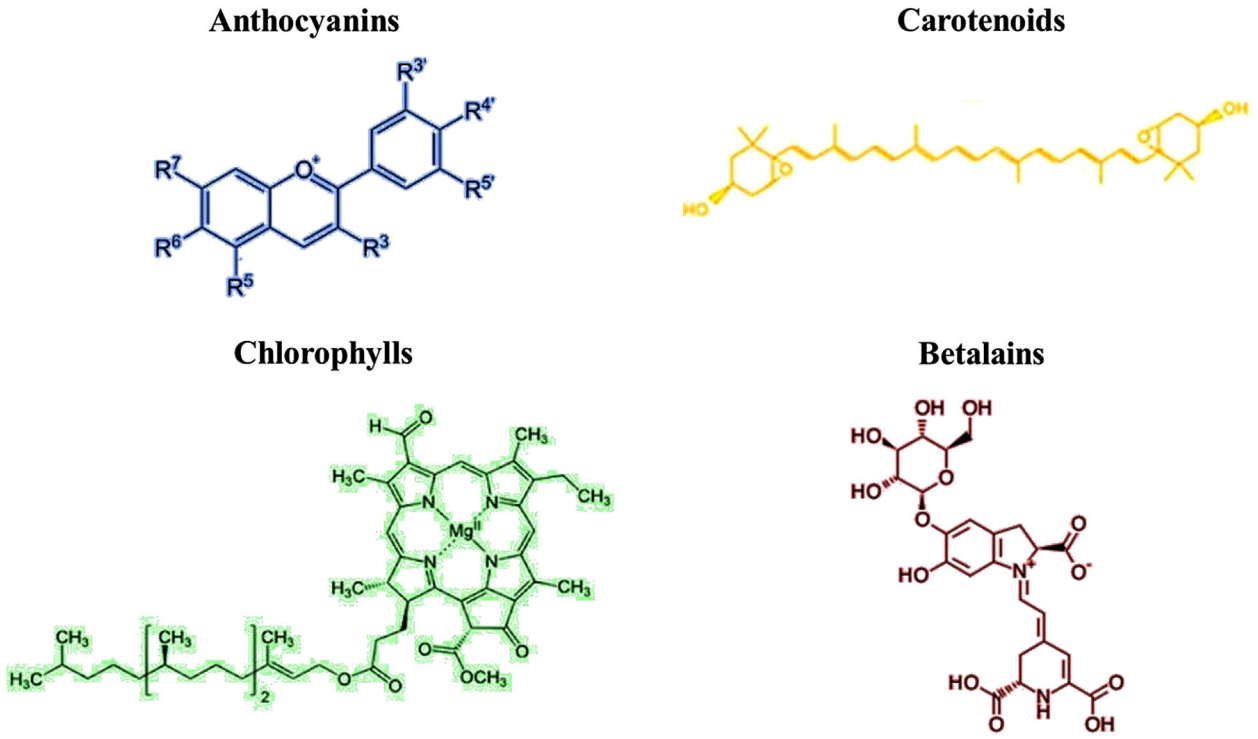

FIGURE 3 Chemical structure of selected natural pigments from vegetable waste

in nature (He & Giusti, 2010). Acylated anthocyanins are more frequent in plants and flowers, while the nonacylated ones are prevalent in fruits (Wallace & Giusti, 2008). Anthocyanins constitute the largest group of natural pigments, and more than 700 different structures have been categorized (Andersen & Jordheim, 2014). Their poor stability makes their application as food colorant challenging. Since these pigments are very sensitive to light, heat, oxygen, and pH (Dangles & Fenger, 2018), the food industry is looking for new ways to improve their stability (e.g., copigmentation, acylation).

Nowadays, anthocyanins as food colorant have been applied to beverages, yogurts, and dry mixes. In the European Union, they are labeled as E163, while in the United States they are exempted from certification (Sigurdson et al., 2017). The FW and byproducts deriving from industrial processes (e.g., wine and juice industry) may be considered excellent sources for the recovery of anthocyanins. The most common FW sources from which they may be extracted are blackberries and grape pomace (Sharma et al., 2021). In this sense, and having the Circular Economy concepts in mind, the use of FW as alternative source of food colorants has gained increased attention in the last few years. Recently, a natural colorant extract from grape skin has been obtained by solvent extraction exhibiting the common properties of polyphenols (Gordillo et al., 2018).

3.2.2 | Carotenoids

Carotenoids are a fat-soluble group of pigments (Figure 3), which vary from yellow to orange to red and they are widespread in nature (e.g., higher plants, bacteria, fungi, yeasts, birds, and insects) (Tanaka et al., 2008). Chemically, carotenoids are tetraterpenoids with 40 carbon atoms made up of isoprenoid units joined in a typical head-to-tail pattern. They can be organized into two classes: carotenes and xanthophylls. The former contains only polyunsaturated hydrocarbons, while the latter has polyunsaturated hydrocarbons with some functional groups containing oxygen. Carotenoids may be classified as acyclic (e.g., lycopene), monocyclic (e.g., γ-carotene), or dicyclic (e.g., lutein). Conjugated carotenoid systems, having delocalized π electrons along the entire chain, are accountable for their absorbance in the visible spectrum (Rivera & Canela-Garayoa, 2012). Depending on the level of conjugation, carotenoids may have more or less intense colors, for example, the larger conjugate systems correspond to the reddest shades, while short chains with a minimum of five double bonds show poor chromaticity. The color is also affected by the degree of cyclization, which explains why lycopene, β-carotene, and γ-carotene, despite having the same number of double bonds, show red, orange, and red-orange colors, respectively (Sigurdson et al., 2017).

Due to their electron-rich and highly unsaturated chemical structure, carotenoids are very sensitive to oxidation and isomerization, especially during food processing and storage (Rodriguez-Amaya, 2016).

These pigments are mainly used as food colors in cheddar cheese, beverages, and flavored milk drinks (Sigurdson et al., 2017). Numerous studies described the recovery of carotenoids from vegetable waste to be applied as food colorants. Many researchers fine-tune green extraction methods, using carrots, pomegranate peel, paprika leaves, orange peel, pumpkin (Sharma et al., 2021), and unsold tomatoes (Lombardelli et al., 2020) as a source. Moreover, carotenoids from tomato peel have been successfully stabilized by means of encapsulation into gelatin nanofibers, retaining antioxidant, antimutagenic, antiproliferative and anti-atherogenic activities (Horuz & Belibağlı, 2018).

3.2.3 | Chlorophylls

Clorophylls (symmetrical cyclic tetrapyrrole, occurring in nature with a centralized attack of phytol and magnesium ion) are able to cover almost the entire chromatic scale of the visible spectrum (Newsome et al., 2014). These two components are an essential part of the molecule to perform its photosynthetic and coloring functions (Figure 3) (Sigurdson et al., 2017). Treatments with weak acids cause centralized displacement of the Mg^{2+} ion, giving rise to phaeophytin, which results in an olive green/gray color shift. Even if this process is permanent, robust complexes may be produced using Cu^{2+} or Zn^{2+} to replace Mg^{2+}, thus restoring chromophore functions and properties. It has been proved that these ions are able to increase the pigment stability, also causing the appearance of more attractive green colors. These complexes are approved for a wide range of application in foods in the European Union, meanwhile in the United States they are allowed only in citrus-based dry beverage mixes (CFR, 2016).

Chlorophylls extracted from various plant waste are commonly used as antioxidants, but their application as food colors is still limited. The only example of chlorophyll used as dye is from avocado peel (Sharma et al., 2021). Furthermore, chlorophyll extracts from spinach byproducts may be used in the formulation of nutraceutical or functional foods due to their antioxidant, anti-inflammatory, and antimutagenic properties (Derrien et al., 2018).

3.2.4 | Betalains

Among nitrogen-heterocyclic compounds, betalains are of particular interest (Figure 3). They are a group of water-soluble pigments, deriving from betalamic acid, that give colors to plants of the order Caryophyllales, such as red beets and prickly pears (Strack et al., 2003). They are composed of two main groups: the red-purple betacyanins and the yellow-orange betaxanthines (Lombardelli et al., 2021a). The former are the products of the condensation of betalamic acid with cyclo-dopa, while the latter are derived from the condensation of betalamic acid and amines (Stintzing et al., 2002). Betacyanins can also be linked with mono or disaccharides, and the resulting glycosides may be acylated, forming several beta-cyanide structures.

Being stable over a wide pH range between 3 and 7, betalains may be used in low-acid food such as dairy products (yogurt and ice cream) (Stintzing & Carle, 2004) and also in syrups, sausages, and confectionaries. In the European Union, they are labeled as E162, and in the United States they are exempted from certification (Sigurdson et al., 2017). Furthermore, the color of betalains is absolutely independent from the pH values compared to other natural colorants, such as anthocyanins. The stability of betalains is affected by several factors, for example, temperature, light, oxygen, and other food components. Sharma et al. (2021) described in depth the betalains extraction from vegetable waste, such as red dragon fruit peels and prickly pear. Recently, Lombardelli et al. (2021a) developed an eco-friendly approach for the recovery of betalain from unsold beetroot. Vulić et al. (2014) showed the antioxidant, antiproliferative, and free radical scavenging activities of betalains recovered from beetroot peel and pomace by ultrasound-assisted extraction (UAE). Likewise, the extracts have proved to be useful in preventing active oxygen-induced and free radical-mediated oxidation of biological molecules.

4 | PIGMENTS EXTRACTION FROM VEGETABLE FOOD WASTE

Vegetable food wastes or byproducts are widely available and inexpensive and may be used as a cost-effective alternative source of natural pigments. Hence, in the last decades, numerous technologies for the utilization of vegetable FW as a resource in the framework of Circular Economy have been implemented.

The main conventional technique applied for pigments recovery from vegetable FW is the solvent extraction (SE). This process consists of a first step of solid–liquid extraction or maceration (Manzoor et al., 2021), and a second step for solvent removal, usually carried out by means of Soxhlet method (Sharma et al., 2021). The selection of the most appropriate organic solvent is crucial, and it depends on its compatibility with respect to target pigment, toxicity, cost, and recovery (de Souza Mesquita et al., 2021). The

most used solvents are hexane (carotenoids from Gac fruit peel, Chuyen et al., 2017), acetone (lycopene from tomato processing waste, Poojary & Passamonti, 2015), methanol (anthocyanins from eggplant peel, Boulekbache-Makhlouf et al., 2013), and trifluoroacetic acid (betalains from pitaya fruits peel, García-Cruz et al., 2017) (Table 1).

Although the conventional extraction techniques with organic solvents are easy to use, economical, and do not require sophisticated equipment, they have numerous disadvantages including the need of a large amount of solvent, a long time, and possible degradation of the extracted pigment. To overcome these limitations, green/unconventional extraction methods have been developed (Figure 4) (Ngamwonglumlert et al., 2017). In order to improve the yield of natural colorants, as well as their chemical stability, the selection of a suitable extraction procedure for each type of pigment is essential. Various innovative techniques of extraction based on supercritical fluids, pressurized liquids, microwaves, ultrasounds, pulsed-electric fields, and enzymes have been explored for the extraction of new potential food colorants from vegetal wastes (Figure 4).

4.1 | Supercritical fluid extraction

It has the benefits of supercritical fluids (which have properties similar to gases) and may lead to an improvement of extraction (Macıas-Sánchez et al., 2005) as these fluids can more easily penetrate the matrix. As shown in Table 1, this method is suitable for the recovery of nonpolar pigments (e.g., carotenoids, chlorophylls), rather than polar ones (e.g., betalains, anthocyanins), since most of the solvents used for supercritical fluid extraction (SFE; e.g., carbon dioxide, organic solvent) are nonpolar (Ngamwonglumlert et al., 2017). Derrien et al. (2018) applied supercritical CO_2 using ethanol for the recovery of chlorophyll and lutein from spinach wastes confirming higher recovery of phytopigments (70% for lutein and 96% for chlorophylls) compared with conventional extraction technique (e.g., acetone) (Table 1).

Despite the need of small quantities of solvent, this approach requires great capital and operating costs due to the high pressures used (Veggi et al., 2013).

4.2 | Pressurized liquid extraction

It uses a liquid solvent at high pressure (10.3–13.8 MPa) at 40–200°C (Miranda et al., 2021). The high temperature facilitates the diffusion of solvent in the matrix and accelerates the degradation of the plant cells, enabling the pigments recovery, thus increasing the extraction yield. Pressurized liquid extraction (PLE) may be applied for the extraction of polar and apolar pigments, depending on the solvent chosen.

Strati et al. (2015) utilized PLE (700 MPa/10 min) assisted with ethanol on tomato wastes, recovering a higher content of total carotenoids (9.30 mg/kg dw) and lycopene (7.04 mg/kg dw) compared to conventional extraction (total carotenoids −3.63 mg/kg dw, lycopene −2.47 mg/kg dw) (Table 1).

Although PLE needs short times and a low number of solvents, it cannot be effectively applied for the recovery of heat sensitive pigments (e.g., betalains) due to the high temperature used (Miranda et al., 2021) and the high capital/operating costs (Ngamwonglumlert et al., 2017).

4.3 | Microwave-assisted extraction

It is a very promising innovative method for the recovery of polar and nonpolar plant pigments (Table 1) due to its high extraction rate and reduced solvent consumption compared to conventional techniques (Dahmoune et al., 2014). This approach is based on fast heating by microwave radiation causing the breakdown of the plant cell walls, due to the expansion of the cell structure. As a result, pigments migrate more easily outside the cells, resulting in an increase of extraction speed and yield (Ngamwonglumlert et al., 2017). Microwave-assisted extraction (MAE) could reduce the time and solvent volume required during the extraction of pigments, but it is not very selective as analytes are extracted in a wide spectrum of polarity (Miranda et al., 2021). The combination of MAE with vacuum (vacuum microwave-assisted extraction) has been suggested for the recovery of heat sensitive bioactive compounds and pigments (Hiranvarachat et al., 2015).

Elik et al. (2020) recovered carotenoids (77.48%) from carrot juice processing waste using MAE at the optimal process conditions (165 W of microwave power, 9.39 min of extraction time and 8.06:1 g/g of oil to waste ratio) (Table 1).

4.4 | Ultrasound-assisted extraction

It uses ultrasonic waves to generate cavitation bubbles that improve extraction efficiency (Rastogi, 2011). Ultrasonic waves passing through a medium create alternating cycles of compression and decompression, which in turn cause the formed cavitation bubbles to compress and expand. As the bubbles grow larger, they can no longer be contained by the surface tension force and therefore collapse. In this way, shear forces are generated and they are capable of breaking the vegetable cell walls, facilitating the release of intracellular compounds (Pitt et al., 2004). As reported in Table 1, UAE is a technique suitable for the

TABLE 1 Natural pigments extracted from vegetal wastes

Pigment	Sources	Method of extraction	Extraction yield	Reference
Carotenoids	Carrot peels	SFE (SC-CO_2)	5.31% $_{\text{dry basis}}$	(de Andrade Lima et al., 2018)
	Apricot pomace	SFE (SC-CO_2 + ethanol)	100.4 μg/g $_{\text{dry pomace}}$	(Şanal et al., 2005)
	Pumpkin	SFE (SC-CO_2)	109.6 μg/g	(Shi et al., 2010)
	Red pepper byproducts	SFE (SC-CO_2)	68.1% $_{\text{dry basis}}$	(Romo-Hualde et al., 2012)
	Tomato skin	SFE (SC-CO_2)	33.0% $_{\text{dry basis}}$	(Kassama et al., 2008)
	Tomato peels	SFE (SC-CO_2)	60.85% (Lyc, $_{\text{dry basis}}$)	(Kehili et al., 2017)
	Pumpkin rind	SFE (SC-CO_2)	20.50 mg/100 g	(Wang et al., 2017)
	Tomato skin and seeds	PLE	9.30 mg/kg $_{\text{dry basis}}$	(Strati et al., 2015)
	Lemon waste	MAE	3.2–6.2% w/w	(Martínez-Abad et al., 2020)
	Gac peel	MAE	262 mg/100 g $_{\text{dry weight}}$	(Chuyen et al., 2018
	Carrot juice waste	MAE	77.48%	(Elik et al., 2020)
	Pomegranate peels	UAE	0.61 to 0.67 mg $_{\text{carotenoids}}$/100 g $_{\text{dry peels}}$	(Goula et al., 2017)
	Mandarin epicarp	UAE	140.70 mg $_{\beta\text{-carotene}}$/100 g $_{\text{dry sample}}$	(Ordóñez-Santos et al., 2021)
	Carrot waste	UAE	83.32%	(Purohit & Gogate, 2015)
	Guava pulp	UAE	135.0 mg$_{\text{Lyc}}$/100 g	(da Silva Lima et al., 2020)
	Gac peel	SE	271 mg/100 g $_{\text{dry weight}}$	(Chuyen et al., 2017)
	Tomato pulp waste	SE	94.7%	(Poojary & Passamonti, 2015)
Anthocyanins	Blackberry residues	PLE	1.02 mg $_{\text{C3GE}}$/g $_{\text{fresh residue}}$	(Machado et al., 2015)
	Cranberry pomace	SFE + PLE	11.10%	(Tamkutė et al., 2020)
	Black currant marc	MAE	20.4 mg/g	(Pap et al., 2013)
	Acerola pulp	UAE + ethanol	2.00–11.16 mg $_{\text{TA}}$/100 g	(Rezende et al., 2018)
	Purple potato peel	UAE	6.84 mg/100 g	(Albishi et al., 2013)
	Sweet cherries skin	UAE + ethanol	12.2 mg/g	(Milea et al., 2019)
	Black carrot pomace	UAE (thermal)	60.85–74.22 mg/L $_{\text{C3XGGF}}$	(Agcam et al., 2017)
	Blueberry byproducts	PEF	55%	(Pataro et al., 2017)
	Blueberry waste	PEF	223.13 mg/L	(Zhou et al., 2015)
	Elderberry pomace	SFE + SE	24.2%	(Seabra et al., 2010)
	Blackcurrant waste	SE	2%–3% w/w	(Rose et al., 2018)
	Eggplant peel	SE	82.83 mg$_{\text{DGE}}$/100 g $_{\text{dry powder}}$	(Boulekbache-(Makhlouf et al., 2013)
	Red grape pomace	MAE, UAE, SE	34188 ppm$_{\text{GAE dry extract}}$	(Drosou et al., 2015)
	Grape byproducts	UAE, PEF	+10% and +17%	(Corrales et al., 2008)
Betalains	Beetroot pomace	UAE	37.22 mg/100 g $_{\text{dry weight}}$	(Vulić et al., 2014)
	Pitaya peel	UAE	101.04 mg $_{\text{betanin}}$/ 100 g	(Mello et al., 2014)
	Pitaya peel	MAE	–	(Cunha et al., 2018)
	Red beetroots	PEF	90%	(López et al., 2009)
	Red beetroot Peels	UAE	9.81 mg $_{\text{betanin}}$/g $_{\text{dry basis}}$	(Šeremet et al., 2020)
	Beetroot peels	MAE	472.11 mg $_{\text{betanin}}$/L	(Singh et al., 2017)
	Pitaya peel	SE	22,053.6 mg/g $_{\text{dry sample}}$	(García-Cruz et al., 2017)
	Beetroot peel	SE	952.5 mg L^{-1} (Bx) and 1361 mg L^{-1} (Bc)	(Zin et al., 2020)
Chlorophylls	Spinach byproducts	SFE	71.0% w/w	(Derrien et al., 2018)

(Continues)

TABLE 1 (Continued)

Pigment	Sources	Method of extraction	Extraction yield	Reference
	Kiwi pomace	MAE	5.9 mg TCC/100 g_{pulp}	(Carbone et al., 2020)
	Green beans	SE	13.3 mg/100 $g_{green\ beans}$	(Cubas et al., 2008)

Abbreviations: Bc, betacyanins; Bx, betaxanthin; C3GE, cyanidin-3-O-glucoside equivalent; C3XGGF, cyanidin-3-xyloside-galactoside-glucoside-ferrulic acid; DGE, delphinidin-3-glucoside equivalent; GAE, gallic acid equivalents; MAE, microwave-assisted extraction; PEF, pulsed electric field; PLE, pressurized liquid extraction; SE, solvent extraction; SFE, supercritical fluid extraction; TA, total anthocyanins; TCC, total chlorophyll content; UAE, ultrasound-assisted extraction.

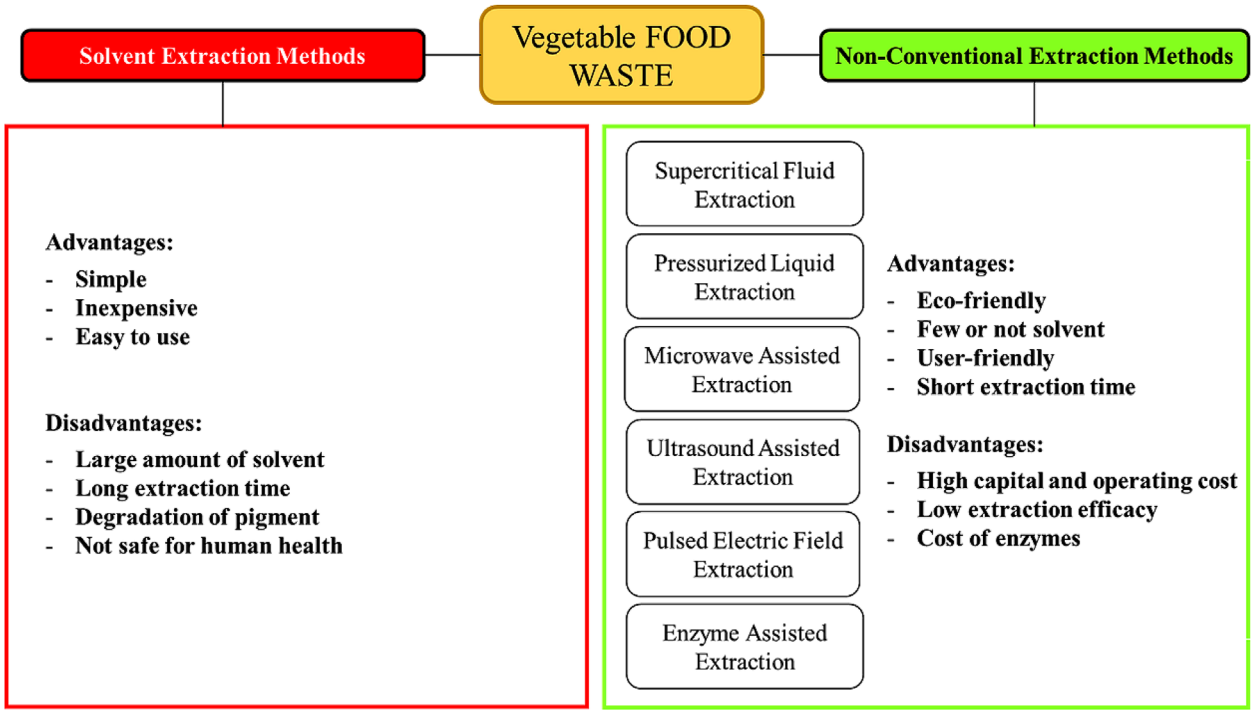

FIGURE 4 Different methods for extraction of pigments from food wastes including advantages and disadvantages

extraction of both polar and nonpolar compounds and also for those sensitive to heat as it is an athermic process, although known for its lower extraction efficiency compared to other nonconventional processes (e.g., SFE and PLE) (Ngamwonglumlert et al., 2017). Drosou et al. (2015) tested three different extraction methods (Soxhlet, MAE, and UAE) for the recovery of anthocyanins from red grapes pomace, revealing the highest procyanidin (43469 ppm) and anthocyanin contents (34188 ppm) in UAE-assisted treatment combined with solvents (Table 1).

4.5 | Pulsed electric field

Pulsed electric field (PEF), using high voltage electrical pulses for short periods, leads to the formation of pores in the plant cell walls allowing more effective extraction (Luzardo-Ocampo et al., 2021). To recover polar pigments (e.g., betalain and anthocyanins), mainly polar solvents with high electrical conductivity are used. On the other hand, PEF cannot be used for the extraction of nonpolar pigments (e.g., carotenoids) since the electric field cannot pass through the nonpolar solvents required for this type of molecules (Ngamwonglumlert et al., 2017). Nevertheless, PEF may be used as a pretreatment to degrade membranes, also enhancing the recovery of apolar molecules (Table 1).

Corrales et al. (2008) compared PEF with other techniques for the recovery of anthocyanins from grape byproducts, observing an improvement in the extraction efficiency (+10% and +17%) with respect to high hydrostatic pressure and conventional solvent extraction (Table 1).

4.6 | Enzyme-assisted extraction

Enzyme-assisted extraction (EAE) of pigments from plants is gaining more attention as it is a useful, mild, green, and

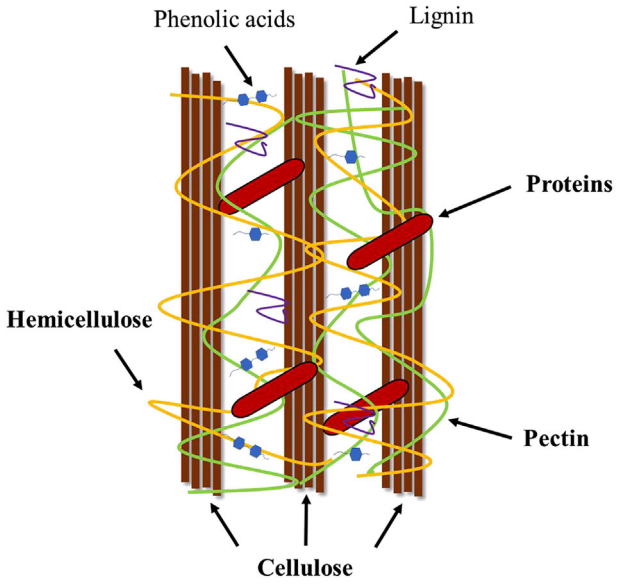

FIGURE 5 Representation of primary vegetable cell wall components

environmental friendly extraction approach, which may be considered a viable alternative to conventional methods (Figure 4) (Nadar et al., 2018). EAE exploits the intrinsic characteristic of enzymes to catalyze reactions with high specificity, region-selectivity, and the ability to conduct such reactions under mild conditions, ensuring the stability of the recovered pigments (Puri et al., 2012). This approach is based on the breakdown of the plant cell wall (Figure 5) by enzymatic hydrolysis. Most of the targeted compounds are trapped inside the cell, and the main obstacle is represented by cell wall consisting of polysaccharides and their derivatives such as cellulose, hemicellulose, and pectin which are linked by hydrophobic interactions and hydrogen bonds. Other compounds, such as phenolic acids, form ether bonds with lignin through their hydroxyl groups in the aromatic ring and ester bonds with structural carbohydrates and proteins through their carboxyl groups (Nadar et al., 2018) (Figure 5).

Hence, enzymes such as hydrolases are employed to solubilize the plant cell wall thereby accelerating the release of intracellular biomolecules. These biocatalysts randomly attack the internal sites of the amorphous region of the polysaccharide chains. This leads to the generation of small oligosaccharides with varied lengths which facilitate easy release of entrapped molecules (Nadar et al., 2018).

Enzymes (e.g., cellulase, hemicellulase, protease, pectinase and xylanase) may be applied alone or in combination as a tailored mix to enhance the extraction yield (Lombardelli et al., 2020).

Among these enzymes, all of them have the ability to hydrolyze some of the main constituents of plant cell walls. In detail, cellulases hydrolyze cellulose releasing cellobiose and glucose molecules when the hydrolysis is complete. Hemicellulases degrade hemicelluloses producing simple sugars or oligosaccharides. Pectinases are able to hydrolyze different types of pectins, releasing uronic acids. Finally, proteases break down proteins in peptides, amino acids, and so forth (Chemat & Vian, 2014).

In order to maximize the enzymatic activities applied, the optimization of all extraction conditions (reaction temperature, extraction time, pH, enzyme concentration, and substrate particle size) is crucial (M'hiri et al., 2014).

The extraction efficiency depends on temperature, pH, treatment duration, enzymatic dosage, and availability of the substrate. As explained above, EAE is very promising in pigment isolation, but the selection of enzymes and the optimization of extraction conditions are very important.

Temperature is a fundamental parameter to consider during extraction since high temperature causes a gradual loss of enzymatic activity and the inactivation of proteins and other bioactive compounds (Peterson et al., 2007). On the contrary, a lower temperature does not accelerate the action of enzymes, leading to a decreased pigment extraction.

The optimal pH for enzymatic hydrolysis differs for each enzyme and it falls within the range of the isoelectric pH of the different proteins (Talley & Alexov, 2010). Since proteins are highly insoluble in this pH range, the release of the biomolecule can be hindered. Therefore, the choice of the optimal extraction pH must be made in such a way as not to hinder the functioning of the enzymes (Nadar et al., 2017).

Considering the treatment time, in general, by extending the contact duration between the enzyme and the extract, it is possible to enhance the degradation of plant cell wall components. However, this is hardly applicable on a large scale as a long incubation time would result in lower product quality and higher energy consumption (Babbar et al., 2016). Furthermore, it has been shown that an extension of the extraction time may lead to a reduction in yield due to the exhaustion of substrates and/or the formation of compounds that inhibit enzymes (Nadar et al., 2018).

Another parameter to be optimized is the overall dosage of the enzyme. High concentrations can ensure a better interaction between enzyme and substrate, facilitating the degradation of the cell wall, but can also lead to the extraction of undesirable substances. The optimal dosage should be a compromise between the cost of the enzyme and the quality of the pigment to be extracted (Jiang et al., 2010).

EAE has been widely applied by using commercially available enzymes (e.g., cellulase, amylase, and pectinase) (Table 2). Prokopov et al. (2017) proved an enhanced recovery yield of carotenoids from tomato peels by an enzymatic mix (cellulase 100 U g^{-1} and hemicellulase 400

TABLE 2 Enzyme-assisted extraction of pigments from food waste

Pigment	Sources	Enzyme	Extraction yield	Reference
Carotenoids	Tomato peel	Cellulase, pectinase, protease, endo-xylanase	55.15 mg/100 g $_{\text{dry basis}}$	(Prokopov et al., 2017)
	Tomato paste	Pectinex Ultra SP-L, Celluclast, Viscozyme L	11.5 mg lycopene/g	(Catalkaya & Kahveci., 2019)
	Red capsicum	Viscozyme L, pectinase, cellulase	41.72–279.83 mg/100 g	(Nath et al., 2016)
	Orange peel	Cellulase, Pectinase	15.0%	(Kumar et al., 2016)
	Pumpkin	Pectinex Ultra SP	2 mg/100 g	(Dinkova et al., 2014)
	Unsold tomato	Polygalacturonase, Cellulase, Xylanase	4.30 mg_{Lyc}/Kg_{tomato})/U	(Lombardelli et al., 2020)
	Tomato peel	Peclyve LI, Cellulyve 50LC, Prolyve 1000	8%−10% w/w $_{\text{dry basis}}$	(Cuccolini et al., 2013)
	Tomato paste	Citrozym CEO and Ultra L, Peclyve EP and L	32.6 mg_{Lyc}/100 g_{tomato}	(Zuorro & Lavecchia, 2010)
	Tomato waste	Peclyve PR, Cellulyve 50LC	72.3%	(Zuorro et al., 2011
	Blueberries	Cellulase, Pectinase	23.5%	(Xu et al., 2016)
	Sohiong fruit	α-Amylase, cellulase, pectinase, protease	984.4 mg	(Swer et al., 2016)
	Blackberries	Klerzyme-150	639 g/L	(Gong et al., 2014)
	Bilberries	Pectinex, Panzym Pro Color, Panzym BE XXL	622, 705, 811 mg $_{\text{MGE}}$/L	(Dinkova et al., 2014)
	Grape skins	Pectinase	3.01 mg/g	(Tan et al., 2020)
Anthocyanins	Bilberry pomace	Viscozyme L	56.15 g/100 g $_{\text{dry basis}}$	(Syrpas et al., 2021)
	Sweet cherry pomace	Depol 740L, Promod 439L, Pectinase 62L	1.75 mg_{GAE}/g	(Domínguez-Rodríguez et al., 2021)
	Raspberry pomace	Pectinex Ultra SP-L, Pectinex Yield Mash, Ultrazym AFP-L	0.32 mg/g	(Szymanowska & Baraniak, 2019)
	Purple corn	Multifect neutral enzyme, Enzeco fungal acid protease	0.94 g/100 g $_{\text{dry basis}}$	(Jing & Giusti, 2007)
Betalains	Unsold beetroot	Polygalacturonase, Cellulase, Xylanase	11.37–14.67 mg/mL U	(Lombardelli et al., 2021a)
	Dragon fruit	Pectinex Ultra SP–L	550 mg/L	(Naderi et al., 2010)
	Red beetroot waste	Celluclast 1.5 L, Pectinex Ultra Mash	3.06 mg/g $_{\text{dry basis}}$	(Fernando et al., 2021)
Chlorophylls	Spinach	Pectinex Ultra SP-L	50.7 mg TCC/100 g_{pulp}	(Özkan & Bilek, 2015)
	Pandan leaf	Pectinex Ultra SP-L	17.82 mg/g $_{\text{fresh weight}}$	(Senklang & Anprung, 2010)

Abbreviations: GAE, gallic acid equivalents; MGE, malvidin glucoside equivalent; TCC total chlorophyll content.

U g^{-1}). In detail, series of experiments were carried out varying enzyme concentrations (100–500 U g^{-1}) in order to establish the influence of enzymatic treatment on the carotenoid extraction yield at a constant temperature (20 or 50°C) and optimum pH. The extraction time was changed from 1 to 5 h. The EAE of carotenoids using mixed cellulase and hemicellulose for 4 h at 50°C resulted in an enhanced recovery yield (1.6 times greater).

Catalkaya and Kahveci (2019) described that lycopene-rich oleoresins may be produced through a treatment of waste by a combination of cellulolytic and pectinolytic enzymes, resulting in a deeper red color extract (Table 2). In depth, central composite design (CCD) was used for the optimization study and response variable was chosen as the lycopene content of the oleoresins (mg/g oleoresin). Enzymatic reaction temperature (40−60 °C) and time (1−5 h), enzyme:substrate ratio (0.2−2 v/w), solvent:substrate ratio (5−30 v/w), extraction time (1−4 h), and enzyme:enzyme ratio (1−3 v/v) were the factors investigated. Optimized conditions were chosen to be as follows: enzymatic reaction temperature = 40°C, enzymatic reaction time = 5 h, enzyme:substrate ratio = 0.2 ml/g, solvent:substrate ratio = 5 ml/g, extraction time = 1 h, enzyme:enzyme ratio = 1. Oleoresin with a concentration of 11.5 mg lycopene/g was obtained at the optimum conditions.

The EAE (cellulase and pectinase) applied to tomato waste increased the carotenoid (sixfold) and lycopene yield (10-fold) compared with nonenzyme-treated sample (Strati et al., 2015). In this study, in the first series of experiments, incubation time varied from 30 to 240 min, while the enzyme concentration was kept constant and equal to 70 and 122.5 U/g for pectinase and cellulase, respectively. The highest carotenoid yield for both enzymes was observed after 180 min of incubation. In the second series of experiments, the incubation time was kept constant (180 min) and the concentration of pectinase and cellulase enzymes varied from 35 to 140 U/g of tomato waste, and 61.25 to 350 U/g of tomato waste, respectively. The highest carotenoid yield was obtained at pectinase and cellulase concentrations of 70 and 122.5 U/g, respectively.

Recently, a tailored EAE protocol for the recovery of carotenoid-containing chromoplasts from unsold tomatoes has been developed. In this study, the specific enzymatic mix (polygalacturonase, pectin lyase, cellulase, and xylanase) was selected taking into account the polysaccharides composition of tomatoes cell wall (Table 2) (Lombardelli et al., 2020). The optimal process conditions for enhancing the recovery of carotenoids from tomatoes (temperature [20–70°C], pH [2.50–8.0], total dosage of enzymatic mix [10–250 U/g] and processing time [20–300 min]) have been investigated. The treatment carried out using the total dosage of 25 U/g for 180 min (45°C, pH 5.5) was the most convenient for maximizing the carotenoids recovery yield.

Dinkova et al. (2014) extracted anthocyanins from bilberry peel by using different commercial enzymes (Table 2), revealing an enhanced yield as well as an extended microbiological juice shelf-life. The extraction was carried out at 50°C for 2 h. Swer et al. (2016) treated *Prunus nepalensis L.* to extract anthocyanins with different enzymes (cellulase, pectinase, protease, and α-amylase), and the highest anthocyanin amount was obtained using cellulase compared to other enzymes (Table 2). In detail, the effect of enzyme concentration, reaction time (30−480 min), and temperature (20−60°C) were evaluated subsequently in several experiments. Cellulase treatment (10% E/S) for 180 min at 4°C exhibited highest anthocyanin yield (Table 2) compared to conventional method.

Lombardelli et al. (2021a) recovered betalain from unsold red-beets applying a tailored enzymatic mix based on the polysaccharide composition of the red-beet cell wall. The process was conducted at 25 and 45°C. Although the pigment extraction at higher temperatures was faster, a comparable recovery yield was achieved at room temperature, also better preserving the color attributes (Table 2). The EAE protocol was optimized, and the most suitable conditions (in terms of pigment yield and color attributes) for the recovery of betalains from unsold beets appeared to be 25 U/g total dose of enzymatic mix, temperature 25°C, and processing time 240 min.

In Özkan and Bilek (2015), Pectinex Ultra SP-L was selected for EAE of zinc–chlorophyll derivatives from spinach pulp, resulting in 39% increase of the extraction yield (Table 2). In particular, the effect of enzyme concentration (1%−9%), treatment temperature (30−60°C), and time (30−210 min) on total chlorophyll content (TCC) was optimized using response surface methodology. Optimum treatment conditions were as follows: 8% enzyme concentration, 45°C, and 30 min, leading to a yield of 50.747 mg TCC/100 $g_{\text{spinach pulp}}$.

Overall, EAE has huge advantages including lower energy consumption, faster extraction rate, higher extraction yield, easier recovery with reduced solvent usage (Puri et al., 2012), reduced risk of their residual in the extract, and higher retention of targeted compounds (K. Ozkan et al., 2021). Furthermore, since this extraction is carried out under mild-controlled temperature conditions, it is very useful for the extraction of heat-sensitive molecules such as flavors and pigments.

Although their use in industrial processes is still limited, several reports in the scientific literature suggest that enzymes have huge potential to be implemented in the advanced large-scale recovery of high-added molecules from agri-food wastes.

Other authors (Sagar et al., 2018) pointed out that the main disadvantages linked to the EAE technique are as follows: (i) high enzyme cost for large volumes of samples, (ii) hardly feasible at industrial level, and (iii) the need to optimize process parameters (e.g., particle size, hydrolysis time, enzyme concentration, and composition).

Despite the price of enzymes represents the bottleneck of this technology, it is certainly important to emphasize that Chemat et al. (2014) have shown, through an economic analysis, that EAE can lead to reduced production costs in terms of savings in time, equipment, and waste disposal compared with other extraction technique. In addition, it has been shown that most commercial enzyme preparations require relatively low application temperatures (<50°C) resulting in a reduction of energy requirements (Chemat et al., 2014).

5 | STABILITY OF NATURAL FOOD COLORANTS

As mentioned above, it has been shown that vegetable FW has the potential to produce very valuable products for further application in the food industries, such as natural colorants.

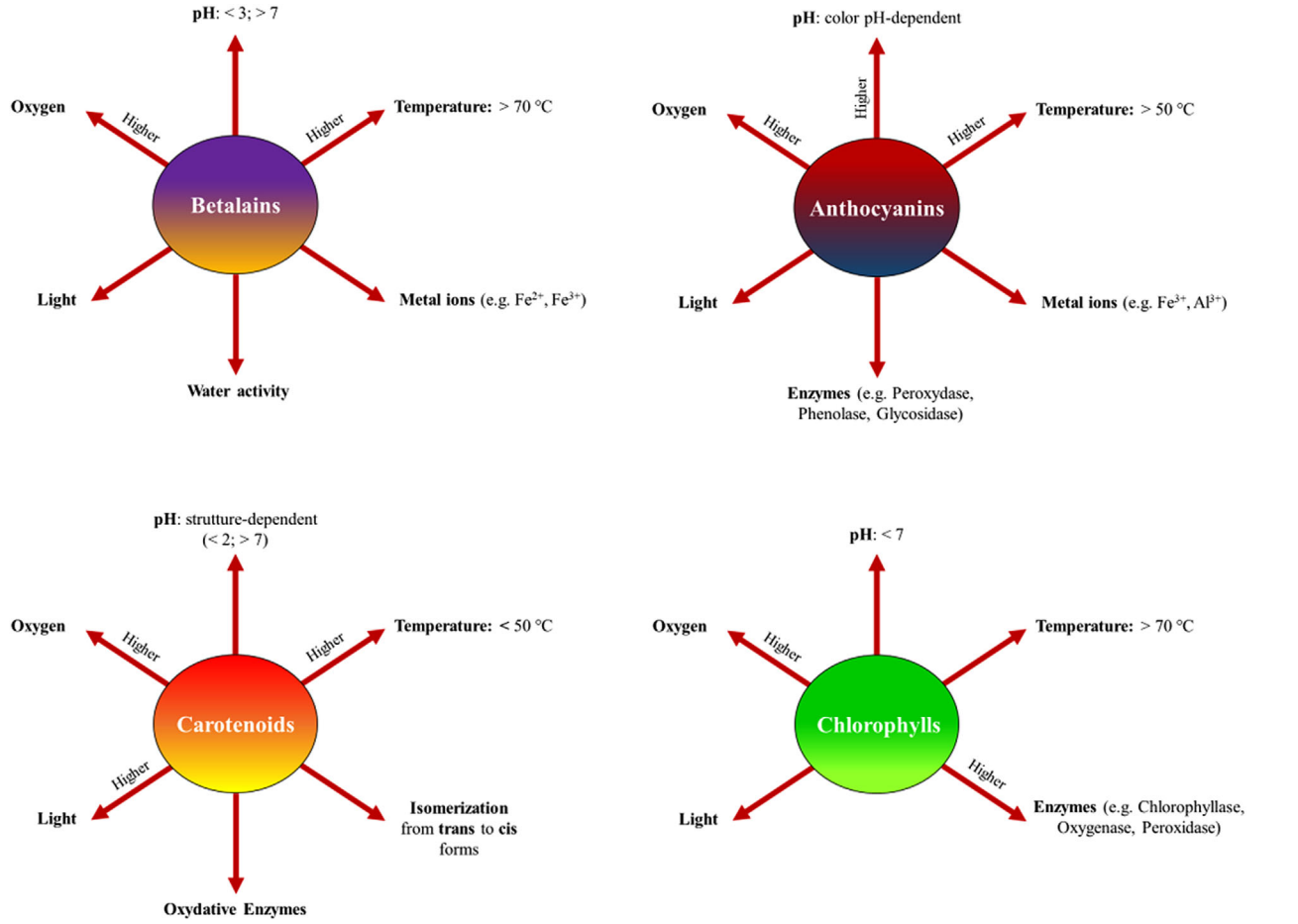

FIGURE 6 Physicochemical factors affecting the stability of pigments

The use of a natural colorant in food requires a detailed knowledge of its stability against potential degradation processes in order to retain as much color as possible throughout production, transport, and storage (Cerreti et al., 2020; Lombardelli, Benucci, & Esti, 2021). Indeed, natural pigments recovered from vegetable waste are highly unstable and susceptible to degradation phenomena caused by external (e.g., processing conditions) and internal factors (e.g., pigment concentration) (Sharma et al., 2021).

Figure 6 shows the main physicochemical parameters which provide instability to pigments, as affected by manufacturing and storage alteration factors.

5.1 | Carotenoids

With a view to the Circular Economy, several studies have been conducted to optimize the extraction of lycopene from tomato processing waste, obtaining a maximum recovery of 95% and reaching a high degree of purity (98%) (Poojary & Passamonti, 2015). As with most carotenoids, the stability of lycopene during food processing and storage is mainly affected by geometric isomerization (cis/trans), oxidation, and fragmentation into smaller volatile molecules (Figure 6) (Rodriguez-Amaya, 2016). The trans isomers of lycopene are commonly found in fruits and vegetables and are thermodynamically stable. However, when lycopene is subjected to heating or irradiation processes, it isomerizes into the cis form, with a consequent reduction in color intensity (Rodriguez-Amaya, 2016). Lycopene oxidation leads to the formation of undesirable small molecules such as acetone, methileptenone, laevulinic aldehyde, and probably glyoxal. In fact, the loss of color is associated with unpleasant tastes due to the presence of aldehydes (Galaffu et al., 2015). The most important degradation factors during the processing of food containing carotenoids are heat, light, and oxygen (Figure 6) (Rodriguez-Amaya, 2016).

The medium in which these reactions occur also affects the stability of carotenoids. Adding antioxidants in the formulation could be a solution for stabilizing such pigments at acidic pH. In this regard, Bou et al. (2011) proved that α-tocopherol was more effective than the other tested compounds (e.g., EDTA, citric acid, tripolyphosphate, propyl

gallate, and gallic acid) in stabilizing lycopene. Encapsulation strategies have been applied to provide stability to carotenoid-rich extracts under food processing and storage conditions (Corręa et al., 2019). It was observed that supercritical CO_2 extraction followed by encapsulation with β-cyclodextrins resulted in more stable forms of lycopene which were stable for 6 months (Blanch et al., 2007). Lobo et al. (2018) developed several inclusion complexes using yellow pepper pigments and β-cyclodextrin with two different procedures. Color stability in all isotonic beverage samples stored in the presence (1400 lux) and absence of light was assessed by the colorimetric method. The values of the coordinates L^*, a^*, and b^* demonstrated a higher color stability of the isotonic drinks added with the inclusion complexes compared to the colored drink with only the raw yellow pepper extract, proving that the inclusion may be a valid alternative for the application of natural additives in food. A significant improvement in the stability of lycopene was shown by Durante et al. (2020) following encapsulation in β-cyclodextrin, as it appeared by the increased stability of carotenoids over a period of 90 days at 25 and 4°C. The results showed that encapsulation significantly improves the stability of lycopene. de Freitas Santos et al. (2021) comprehensively reviewed the materials applied for the production of carotenoid-rich microparticles by spray drying, including carbohydrates (e.g., modified starch, maltodextrin, β-glucan, carboxy-methylcellulose, inulin, and pectin), proteins (e.g., chickpea protein, gelatin, whey protein isolate, and concentrate), and gum arabic (GA). They argue that the choice of extraction solvent, as well as the encapsulation technique and materials may contribute to the successful preservation of carotenoids. Indeed, as reported by Álvarez-Henao et al. (2018), lutein degradation after spray drying decreased from 97.62% to 8.06% when modified starch was substituted by GA as a carrier agent.

Nanoencapsulation seeks to form structures or particles that function as nano-carriers, which implies that at least one of the characteristic dimensions of the capsules is smaller than 100 nm. The most widely used nanoencapsulation methodology applied to carotenoids is nanoprecipitation (Fuenmayor et al., 2021). Afonso et al. (2020) reported that this technique has been used to encapsulate β-carotene in polymers as ethylcellulose (particle size: 60 nm, encapsulation efficiency: 74%) and zein (particle size: 83 nm, encapsulation efficiency: 93%).

Recently, the enzymatic recovery of carotenoids, keeping them inside chromoplasts, allowed us to obtain stable formulations without the need of further stabilization techniques (Lombardelli, Benucci, & Esti, 2021).

5.2 | Anthocyanins

One of the most interesting properties of anthocyanins is their ability to cover a broad spectrum of colors, ranging from red (at acid pH) to purple and blue (at neutral pH) (Figure 6). Red anthocyanins may be used to color soft drinks below pH 3. Also, some anthocyanin-based extracts are widely applied to dairy products to enhance the color of yogurt (pH 4.5). Selecting the right type of anthocyanin is essential for limiting color loss. For example, anthocyanins with particular substituents on the flavyl nucleus (e.g., sugar groups esterified with phenolic acids) maintain a better stability to heat and light (Pina et al., 2012). It may be explained by the fact that the substituted anthocyanins are reorganized in a secondary structural configuration that is able to protect the pigment from the hydration reaction, which normally determines the formation of colorless chalcones. This mechanism has been reported by numerous authors as a co-pigmentation in which a noncovalent interaction occurs between anthocyanin and a co-pigment (other colored or colorless molecules, metal ions or mixtures of these) (Cavalcanti et al., 2011). The resulting effect can generally be expressed as a bathochromic shift and is sometimes accompanied by a hyperchromic effect (Pina et al., 2012). In light of this, co-pigmentation is one of the most effective methods for obtaining stable anthocyanins for food applications (Galaffu et al., 2015). Furthermore, when anthocyanins are in the presence of transition metal salts (e.g., in nutritionally fortified products), important color changes may be observed. Although this mechanism is exploited in nature in flowers (Yoshida et al., 2009), it may represent a problem for the food industry (e.g., fruit-based preparations used in yogurt lose their characteristic color). One possible solution involves the use of chelating agents (EDTA), although their addition is not allowed in all countries (Galaffu et al., 2015). Recently, encapsulation by spray drying has been applied in order to enhance anthocyanins stability throughout storage, promoting their controlled release in food (Cortez et al., 2017). The microencapsulation of anthocyanins has been performed by using different coating materials. Correa et al. (2019) successfully tested maltodextrin, whereas Garcıa-Tejeda et al. (2015) used normal, modified, and waxy maize starches, showing the highest encapsulation efficiency when starch derivatives were applied. Guo et al. (2018) used alginate-pectin hydrogel particles to encapsulate anthocyanins extracted from blueberry and purple corn. The greatest encapsulation efficiency was observed for blueberry anthocyanins (116%) compared to purple corn anthocyanins (65%). The photodegradation rate was reduced in both encapsulates and at the same time the retention rate of anthocyanins during storage was increased, thus improving their

shelf life in low pH beverages (e.g., fruit juices or lemonade).

Liposomes may also be used to encapsulate anthocyanins due to their good ability to protect hydrophilic substances (Enaru et al., 2021). Gültekin-Özgüven et al. (2016) evaluated the encapsulation of spray-dried black mulberry in chitosan-coated nanoliposomes in dark chocolate. Anthocyanins were added to chocolates by varying the degrees of alkalinization (pH 4.5, 6, and 7.5) and the conching temperatures (40, 60, and 80°C). The results showed that, compared to the spray dried extract, the chitosan-coated liposomal powders provided better protection of the anthocyanin content when both temperature and pH increased.

Furthermore, anthocyanins can be easily incorporated into the dispersed phase of an emulsion to be protected in its continuous phase from harmful external environmental conditions, mechanical, chemical, or enzymatic degradation (Souza Simões et al., 2017). Almeida Paula et al. (2018) added a commercial anthocyanin extract and 1.25% guar gum, in the aqueous phase of a double emulsion and observed a high encapsulation efficiency (90.6%), an increase in thermal stability and the protection of anthocyanin molecules from degradation. Furthermore, the addition of guar gum increased the color stability.

5.3 | Betalains

Unlike anthocyanins, betalains are more stable to pH (Figure 6) and changes from 3.5 to 7 do not induce color alteration or hydrolytic cleavage. However, far from this range, degradation occurs rapidly (Rodriguez-Amaya, 2016). It has been observed that betalains are unstable in the presence of oxygen and that some antioxidants (e.g., ascorbic acid and tocopherol) seem to protect them only marginally from oxidation (Castro-Enríquez et al., 2020).

Exposure to light also has a negative effect on their stability. Indeed, it accelerates the oxidation reaction of betalains due to the excitation of electrons toward the orbitals at higher energy levels, resulting in a greater molecular reactivity. Heat treatment remains the most significant problem that needs to be addressed in the use of betalains in food. A significant loss of color begins to occur even below the pasteurization conditions of 70–80°C (Castro-Enrìquez et al., 2020).

Lombardelli et al. (2021b) contributed to increase knowledge about the arranged effect of heat and UV-light on the visual color of betalains recovered from unsold beetroot, proving that high temperature (40°C) leads to a greater pigment degradation especially under UV-light exposure.

To overcome these problems, a solution could be the addition of colorant in food matrices at the end of the process (Emerton, 2008).

Therefore, in order to prevent or limit the loss of betalains color during heat treatment, the absence of metal ions, light, and oxygen is necessary, thus requiring in turn the systematic use of antioxidants and chelating agents. Unlike anthocyanins, betalains may offer red colors at neutral pH conditions, and this property may be exploited to create more expensive and regulated color alternatives such as carmine (Herbach et al., 2006). Encapsulation of betalains has been performed by using different techniques and matrices (e.g., polysaccharides, proteins or in combination with polysaccharides) (Albuquerque et al., 2021; Castro-Enríquez et al., 2020). Ravichandran et al. (2014) reported the encapsulation of betalains from red beet in maltodextrin and its combination with guar gum, GA, xanthan gum, and pectin. Interestingly, the xanthan gum combination allowed to increase the stability (+21%) with respect to the control. Chranioti et al. (2015) encapsulated beet and saffron dye in maltodextrin, GA, modified starch (MS), chitosan and in combination, proving that encapsulation is suitable for color stabilization. The powders produced were evaluated in terms of color strength during storage at 40°C for 10 weeks. Chewing gum samples produced with color extracts encapsulated in GA–MS showed the highest values of a^* (for beet) and b^* (for saffron) indicating better protection. Vargas-Campos et al. (2018) studied the encapsulation of refined betalain extracts using native potato starch and its modification through phosphorylation or succinylation. The microcapsules were produced by spray drying and their stability was evaluated at 40°C for 39 days and using them as yoghurt pigmentants (pH 4.6) at 4°C for 32 days. Succinylated potato starch proved to be the best alternative for stabilizing betalains. Antigo et al. (2017) evaluated the stability of red beet extract microcapsules, dried by freeze-drying and spray drying and stored at room temperature as the pH varied (3–6). The microcapsules were manufactured using a combination of maltodextrin and xanthan gum as encapsulating agents and stored for 7 days. All microcapsules were characterized by a greater stability in terms of betanine content, phenolic compounds, and color parameters during storage at different pH.

5.4 | Chlorophylls and their derivatives

Given that in the culinary field the green color is considered a freshness index, the stabilization of green pigments (present in ingredients such as vegetables or spices) is a fundamental goal to be achieved. Chlorophylls and their derivatives (e.g., Cu-complexes) represent the main

natural green pigments, and their stability, as for other colorants, is affected by temperature and pH (Figure 6) (Galaffu et al., 2015). When the central magnesium ion is replaced by two protons to form phaeophytin, an undesirable color change occurs, which shifts toward grayish shades. The speed of this reaction is strongly affected by the acidity of the matrix and slows down to pH values around neutrality. Hence, chlorophyll stability is strongly affected by the control of this reaction. Operation processes do not cause instant color degradation, but rather initiate a slow color transformation which is observed at the beginning of the shelf-life (Galaffu et al., 2015). Several methods have been patented to prevent the degradation of chlorophyll, but they cannot be applied everywhere due to food legislation of each country. For example, the use of zinc to stabilize the green color of chlorophyll is only allowed in the United States for green beans and peas. In the European Union, the use of copper is allowed (Galaffu et al., 2015). These metal ions, replacing the magnesium of the central structure of chlorophyll, are able to produce more pH-stable green colorants (G. Ozkan & Bilek, 2015). Only few recent studies have been conducted on the stabilization methods of chlorophylls after extraction. Kang et al. (2019) encapsulated chlorophylls by using different blends of GA and maltodextrin by spray-drying, demonstrating that microcapsules coated with maltodextrin alone showed the highest storage stability (94.7%—97.5%). Zhang et al. (2019) produced cabbage leaf chlorophyll microcapsules and whey protein isolate (WPI-CH), in order to improve chlorophyll solubility and stability. The results proved that the encapsulation efficiency and chlorophyll solubility were increased by 3.78% and 7.79%. Raei et al. (2017) evaluated the effects of alfalfa chlorophyll microencapsulation on its stability with respect to high temperatures (80–100°C) and acidic pH (6.5), as well as its application in heated food. The results of this study presented a suitable way to improve the green pigment stability of alfalfa as a valuable and cost-effective source of chlorophylls.

6 | CONCLUSIONS

Emerging advances, innovations, and challenges in the extraction and application of natural pigments are the core focus of this review. Within the Circular Economy framework, the food industry needs to adopt measures to enhance vegetable waste, as a valuable source of natural pigments. In the modern food and biotechnology industry, the application of enzymes for the recovery of natural food colorants has been highlighted as a useful alternative to conventional techniques, being mild, eco-friendly, and economically efficient. The great challenge for the future is the exploitation of natural and health benefiting pigments from food waste that allows to minimize the environmental impact of waste supporting the Circular Economy concepts, also contributing to the sector company profit. Finally, more in-depth studies will be needed to further enhance the stability of recovered natural pigments to enable their usability as food colorants.

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AUTHOR CONTRIBUTIONS

Data curation, writing-original draft preparation, and investigation: Claudio Lombardelli, Caterina Mazzocchi. *Conceptualization, methodology, supervision, validation, writing-reviewing, and editing*: Ilaria Benucci, Marco Esti.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

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