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Insights on the use of alternative solvents and technologies to recover bio-based food pigments

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Abstract

This review will discuss, under the Circular Economy and Biorefinery concepts, the performance of the alternative solvents in the downstream process to recover natural pigments in a more sustainable way. Conventionally, pigments marketed on an industrial scale are produced through chemical synthesis by using petroleum derivatives as the main raw material. Also, the current production chain of the synthetic dyes is linear, with no solvent recycling and waste generation. Thus, the most promising processes of extraction and purification of natural pigments and strategies on the polishing of the solvents are here reviewed. In this review, the use of alternative solvents, namely, ionic liquids, eutectic solvents, aqueous solutions of surfactants, and edible oils, for recovering natural pigments was reviewed. Works discussing higher extraction yields and selectivity, while maintaining the stability of the target pigments, were reported. Also, a panorama between Sustainability and Circular Economy prospection was discussed for better comprehension of the main advances in the field. Behind the analysis of the works published so far on the theme, the most important lacunas to overcome in the next years on the field were pointed out and discussed. Also, the future trends and new perspectives to achieve the economic viability and sustainability of the processes using alternative solvents will be scrutinized.

KEYWORDS

aqueous biphasic systems, biorefinery, circular economy, downstream processes, eutectic solvents, ionic liquids

1 | INTRODUCTION

Pigments are responsible for assigning a specific color to a product like paints, plastics, textiles, cosmetics, foods, and others. Color is the first sensory attribute impacting the perception and influencing the consumer's emotional reactions contributing to product acceptance (Solomon, White, Dahl, Zaichkowsky, & Polegato, 2017; Spence, 2015). Natural pigments are defined as molecules able of emitting color, which are extracted from natural raw materials, namely, plants, animals, and algae, or produced by microorganisms (fungi, bacteria, cyanobacteria, and yeast). These transmit or reflect light as a result of the selective absorption by the chromophores in a specific wavelength (Sonar, Rasco, Tang, & Sablani, 2019). The different chromophores are indeed the main responsible for the huge diversity of natural compounds able to introduce color on products. The different colors are directly related to the chromophore structural arrangement, which is in general described by a molecular structure composed of

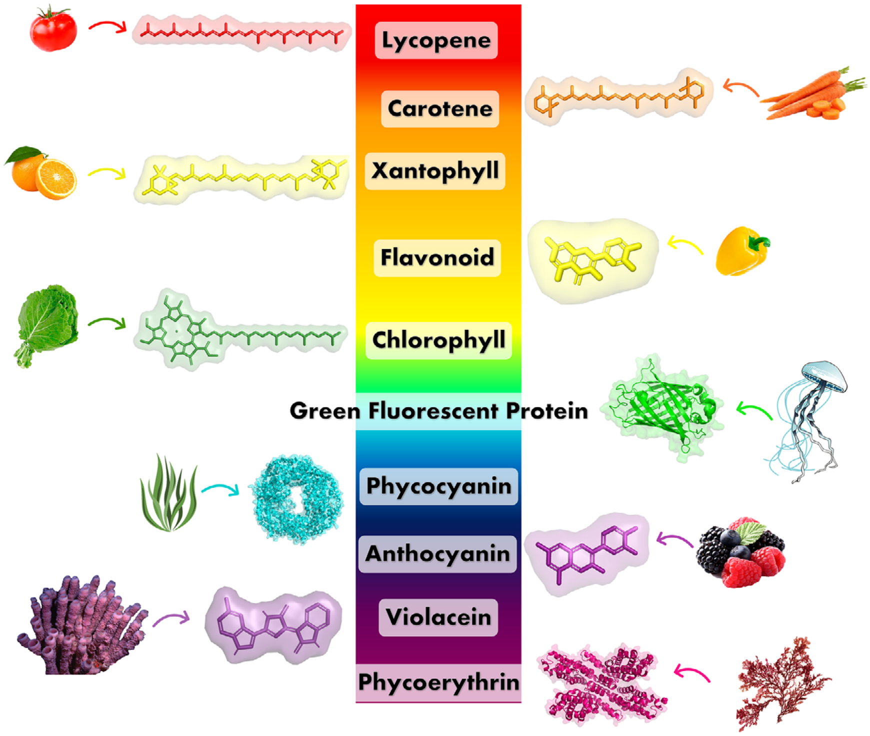

FIGURE 1 Examples of different natural pigments extracted from biomass and their sources

conjugated double bonds, or by a metal-coordinated porphyrin. As depicted in Figure 1, there is a huge variety of natural sources from where it is possible to recover natural colorants. Natural pigments are subdivided into three main categories: (i) terrestrial, from fruits, leaves from the forestry, and crops of superior plants; (ii) marine, pigments accumulated by algae, cyanobacteria, and marine organisms; and (iii) pigments produced by microorganisms, including fungi, bacteria, and in a lower extent, yeasts, which are commonly associated with the surfaces of other animals, like violacein found in sea sponges (Yang, Xiong, Lee, Qi, & Qian, 2007). Due to the wide structural variety of pigments and natural sources with different properties and structures, there is no single standardized downstream method for obtaining natural colorants, which is nowadays, as in other fields, an important request for industry and academia to solve.

Into a nutrition perspective, there is an increase in the demand for natural foods, free of allergens, and synthetic additives, which justify the replacement of synthetic to natural dyes (Yusuf, Shabbir, & Mohammad, 2017). However, even with this actual concern, synthetic colorants are very competitive compared to natural pigments. Today, natural dyes have a small niche market, because their distribution, availability, and properties are limited. In fact, presently only 16 natural pigments are allowed to be applied in food; these included in the classes of betalains, flavonoids, anthocyanins, carotenoids, isoprenoids, annatto (bixin, norbixin), paprika-based pigment, and porphyrins (chlorophylls, chlorophyllins, and copper complexes of these compounds), caramels, curcumin, and plant coal (Janiszewska-Turak, Pisarska, & Królczyk, 2016). Due to the limited number of approved natural pigments and the high demand for new natural dyes required by consumers worldwide (Martins, Roriz, Morales, Barros, & Ferreira, 2016), new strategies have been pursued. Also, driven by the need to get ecological and less hazardous pigments, there is a growing trend toward alternative chemistry strategies (Muhd Julkapli, Bagheri, & Bee Abd Hamid, 2014). Based on the 12 principles of Green Chemistry (Anastas & Eghbali, 2010; Erythropel et al., 2018), some theories report that the best way to create a sustainable downstream process to obtain natural added-value compounds should follow both Biorefinery and Circular Economy strategies (Clark, 2019; Clark, Farmer, Herrero-Davila, & Sherwood, 2016; Guillard et al., 2018). Additionally, aiming to optimize the disposal of biomass (forestry, food, marine, and freshwater sectors), there is a clear need to act on the creation of new products and integrated platforms for the efficient valorization of a large plethora of wastes/residues (Schanes, Dobernig, & Gözet, 2018; Zimmerman, Anastas, Erythropel, & Leitner, 2020). For example, as a worldwide concern, there is an international goal for sustainable development to halve per capita global food waste at the retail and consumer levels by 2030 (Reynolds et al., 2019). Furthermore, under the scope of this review, the most conventional methods to obtain dyes from natural matrices are not specific, impairing the selectivity of the process and the reuse of biomass for other purposes. Thus, the alternative

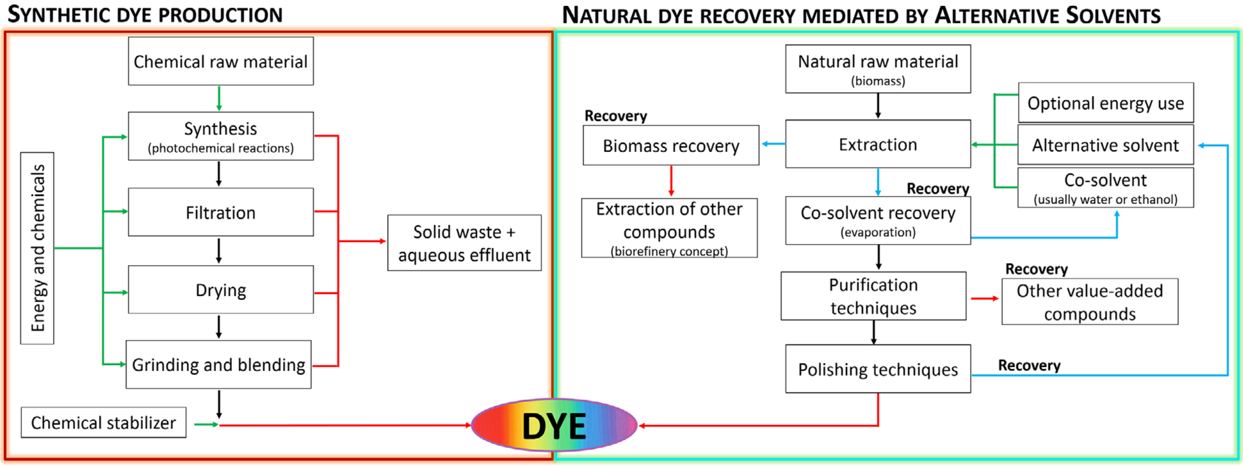

FIGURE 2 Representative scheme comparing different pigment production systems: synthetic dyes production steps (a) and natural dyes recovery steps mediated by alternative solvents (b). Black arrows: processes; green arrows: input; red arrows: output; blue arrows: alternative ways

methods, especially those based on ionic liquids (ILs), and more recently, eutectic solvents/mixtures, surfactants, edible oils, and copolymers, have been considered of utmost interest, including in the food sector (de Souza Mesquita, Murador, & de Rosso, 2019; Martins, Braga, & de Rosso, 2017; Toledo Hijo, Maximo, Costa, Batista, & Meirelles, 2016). Unfortunately, there are still no (or just a few) scale-up applications reported on natural pigments in industrialized products, which justifies the need for the development of new platforms to obtain these compounds. The market price of a natural pigment is much higher than its synthetic counterpart, because the extraction yield is generally low, whereas the synthesis of a pigment is easily reproducible by the industry on a large scale. However, the manufacture of a synthetic pigment is also representing a negative impact, namely, regarding their potential adverse allergenic and intolerance reactions, as well as their potential to cause respiratory problems (Amchova, Kotolova, & Ruda-Kucerova, 2015). It has been proved that some synthetic food colorants, such as tartrazine derivatives, may cause an intolerance reaction in susceptible people and have been linked to increased hyperactive behavior in children (McCann et al., 2007). Moreover, the manufacture of synthetic food pigments is extremely aggressive to the environment, which increases the public's desire toward environmentally friendly processes (Yusuf et al., 2017) and more sustainable pigments in their daily base products. For example, the production of the Allura Red Ac (E129), used to mimic the lycopene' coloration, has severe issues regarding not only the environment, but also in terms of social responsibility, regarding safe working conditions, consumers' expectance, health, and impurities (Gebhardt, Sperl, Carle, & Müller-Maatsch, 2020). The nonsustainable production processes of the synthetic pigments, in addition to their dependence on nonrenewable sources (petroleum derivatives), are also a concern. As shown in Figure 2, the manufacture of synthetic pigments follows a linear production, with a dependence of energy, high water consumption, and without reuse and recycle of raw materials, leading consequently to the production of solid wastes and aqueous effluents (Gebhardt et al., 2020; Natarajan, Bajaj, & Tayade, 2018). The industry receives the initial raw material (petroleum derivatives chemicals), and several steps with high energy impact are followed (photocatalysis reaction, filtration, drying, blending, and stabilization reactions). These steps represent a huge impact on the environment because the inadequate disposal of dyes (textile, food, and others) on the aquatic environment reflects in more than 7×10^5 tonnes per year of contaminated waters (Hasan & Jhung, 2015; Zamora-Garcia et al., 2018). On the other hand, also in Figure 2, it is possible to highlight the application of the Circular Economy concept under the biomass valorization. The alternative processes (apparently) have a more complex matrix compared to synthetic (and conventional) dye production. However, the alternative process is a retro sustainable system, which favors not only the production of dyes but also the possibility of recover other value-added compounds, associated with the possibility of recycling raw materials and solvents, under the guidelines of Sustainability, Green Chemistry, and Circular Economy. Therefore, processes based on the understanding of the risk for humans and the environment, associated with broadly physical and global hazards analysis, are urgent (Anastas & Miller, 2018). These analyses put in evidence the role of the alternative solvents in new and more sustainable extraction and purification schemes/processes, as previously reviewed by some of us (Ventura et al., 2017) and others (Clarke, Tu, Levers, Brohl,

& Hallett, 2018; Smith, Abbott, & Ryder, 2014; Zimmerman et al., 2020).

ILs and eutectic solvents have been defined as alternatives solvents able to replace (or reduce) the use of volatile and hazard organic solvents in many fields, because (at least) some of them have been considered environmentally friendly, easy to formulate, and with tunable properties, allowing their use as task-specific compounds. ILs started to be explored more than 20 years ago. They are recognized by their unique physicochemical properties such as the negligible vapor pressure, non-flammability, and high thermal and chemical stability (Anastas & Eghbali, 2010). On the other hand, eutectic solvents are defined as low-transition-temperature mixtures (Abbott, Boothby, Capper, Davies, & Rasheed, 2004), for which the eutectic temperature is below that of an ideal liquid mixture. Due to its selectivity and extraction potential (Cláudio, Ferreira, Freire, & Coutinho, 2013; Jablonský, Škulcová, Malvis, & Šima, 2018), both ILs and eutectic solvents are promising candidates to be applied on the development of more efficient and sustainable extractants (Smith et al., 2014; Ventura et al., 2017). The possible combination of the starting materials to synthesize the ILs and to prepare the eutectic solvents is virtually countless, and therefore, numerous combinations can be produced, with different chemical and biological properties (Kalhor & Ghandi, 2019; Passos, Freire, & Coutinho, 2014; Ventura et al., 2017). Supported by their "design solvent" credential, these have been applied as efficient solvents. This observation was done because some of the works have evidenced the high extraction yields, improved selectivity, and purification factors (Freire et al., 2012; Ventura et al., 2017) of processes based on these alternative solvents. Despite the important role of natural pigments in our life (Delgado-Vargas, Jiménez, & Paredes-López, 2000; Minich, 2019), the available methods of extraction and purification are not always appropriate, thus affecting the quality of the final product (Murador, de Souza Mesquita, Vannuchi, Braga, & de Rosso, 2019). For example, some studies have shown that the all-*trans*-*β*-carotene obtained from a natural source is more bioaccessible, providing much more confidence to the consumer than the synthetic one (Bogacz-Radomska & Harasym, 2018). Recently, Murador et al. (2020) concluded that carotenoids and chlorophylls extracted from orange peels by an IL-mediated process have shown similar bioefficacy compared to those extracted by organic solvents (acetone, and ether mixtures), indicating that the search for more sustainable alternatives to pigments extraction is extremely important, mainly considering a promising use of this natural pigments in the food sector.

Although not new, the improvements in the extraction yields using alternative solvents have gained prominence every year (Passos et al., 2014; Ventura et al., 2017). Nevertheless, target purification processes based on ecofriendly strategies are still scarce. This review takes into account the need for new natural pigments pointed out by the food sector. The progress and novelty of using alternative solvents on the extraction and purification of pigments of natural origin, especially those with much higher potential to be applied in new/improved food formulations, are thus here reviewed. This review is organized considering the works assessing the solid–liquid extraction of pigments from the biomass, followed by the purification techniques employed to separate the target pigments from the other contaminant, as depicted in Figure 3. In this figure, a list of the pigments reviewed is presented as well as the methodologies/techniques applied so far on the purification of the target pigments were identified. In this case, purification means the pigments' separation from other contaminants and their isolation/polishing represents the separation of pigments from the main solvents used, which simultaneously, and for some situations, allowed their recycle and reuse for new cycles of the downstream process. The polishing techniques, focusing the separation of the target compounds from the main solvents (Figure 4), are an important step to achieve a safer product to be used in sectors like food, cosmetics, and pharmaceutical. Considering the whole picture of the actual scenario, this work provides a critical perspective on how the sector of natural pigments may benefit with the development of the concept of process integration mediated by alternative solvents, thus allowing process gains with improved food formulations in a win–win relationship between natural pigments,

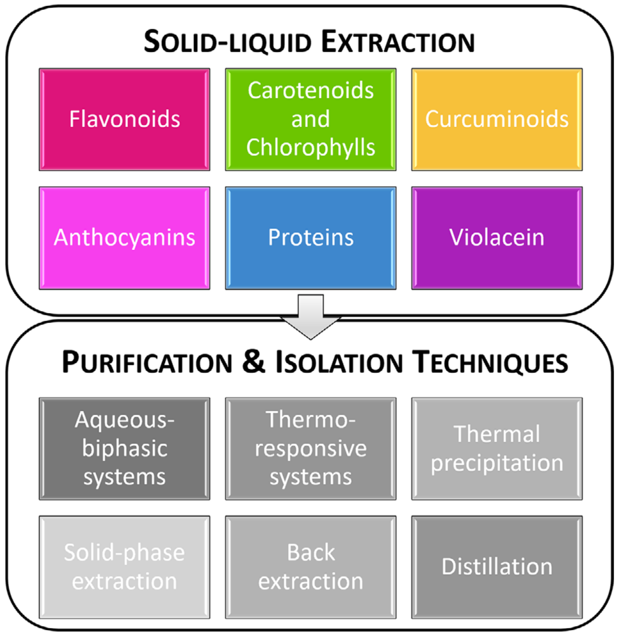

FIGURE 3 Summary of the pigments and techniques/methodologies studied in this work

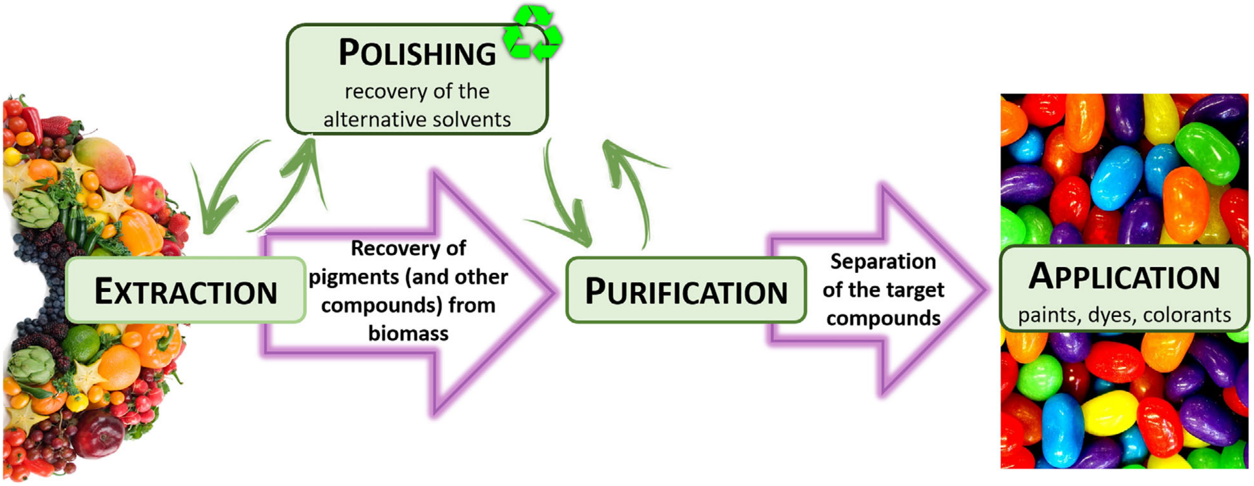

FIGURE 4 Main steps for the recovery and purification of natural colorants from raw materials, as well as the polishing to recover alternative solvents from extracts

sustainability, downstream process integration, food sector, and economic profit.

2 | (SOLID–LIQUID) EXTRACTION OF NATURAL PIGMENTS FROM BIOMASS

Most of the articles reporting the recovery of pigments from biomass use specific analysis to optimize the process aiming to achieve high extraction yields. By applying surface response tools, the authors do the simultaneous study of different conditions influencing the process, namely, temperature, extraction time, solid–liquid ratio ($R_{S/L}$), homogenization mechanisms, and solvents' concentration. The processes reported in literature differ mainly in the treatment of these independent variables, for example, regarding the type of alternative solvent and homogenization methods used. Choosing the most appropriate solvent is a crucial step, and for that, four main factors must be considered, namely, (i) the biocompatibility of the solvent with the target compound, (ii) toxicity, (iii) cost, and (iv) recyclability potential (Płotka-Wasylka, Rutkowska, Owczarek, Tobiszewski, & Namieśnik, 2017; Schuur, Brouwer, Smink, & Sprakel, 2019). Considering the extraction of carotenoids as an example, the use of tensioactive solvents is a facilitator due to their low polarity (Choi et al., 2019; Ulloa et al., 2012), contrarily to what happens for the extraction of anthocyanins and phycobiliproteins, for which hydrophilic solvents are the best approach. Another differential factor among the research works approached in this review is the homogenization techniques used to perform the release of pigments in solution (dependent or not on electricity), which must be chosen considering the initial conditions of the biomass (e.g., humidity). Therefore, Table 1 shows the main works already published on the extraction of natural pigments using alternative solvents (ILs and eutectic solvents), and more recently, the extraction mediated by aqueous solutions of surfactants, and edible vegetable oils (a very good option mainly for food proposes). Additionally, Figure 5 represents the distribution of the scientific works dealing with each alternative solvent used for the solid–liquid extraction of natural colorants.

The request for natural pigments instead of synthetic ones in some industries is increasing on a daily base (Rao, Prabhu, Xiao, & Li, 2017). Because the extractions usually have low yields (from µg to mg), and the amounts of colorants present in the biomass are not too high, there is a demand for the development of more efficient and integrated downstream processes. In this sense, the use of alternative solvents has gained increased attention in the most recent years. Because the colorants are part of the biochemical composition of the biomass, raw materials, and residues, the first step always encloses a solid–liquid extraction, where a solvent is added to the biomass, and after a specific time of contact, with or without mechanical techniques, an extract rich in the natural pigments is obtained. This section will be now analyzed by class/type of pigments.

2.1 | Flavonoids

Flavonoids are yellowish natural pigments, water soluble, and with a common core structure called benzopyrone ring (Rodriguez-Amaya, 2019). These are subdivided into five groups (flavanones, flavonols, flavanols, flavones, and isoflavones), which are divided according to the

TABLE 1 Summary of the publications using alternative solvents to solid–liquid extraction of natural pigments derived from different sources, and their respective operational extraction conditions (concentration of the solvent, co-solvent, $R_{(S/L)}$, and $Time_{(min)}$), and strategies of polishing and purification

		Extraction operational conditions								Polishing and purification strategies			
Class of pigments/ pigment	Biomass	Method (conditions)	Solvent	Co-solvent	Concentration in the co-solvent	$R_{(S/L)}$	Time (min)	T (°C)	Yield	Polishing	Recovery of the solvent (%)	Purification	References
Flavonoids	*Platycladi cacumen*	UAE (200 W, 40 KHz)	Cholinium chloride:Levulinic acid (1:2)	Water	90%	25	30	50	5 mg/g	n.e	n.e	SPE	Zhuang et al., 2017
	Pollen typhae	UAE (n.r.)	Cholinium chloride:1,2-propanediol (1:4)	Water	70%	50	35	n.r.	86.87% to 98.89%	n.e.	n.e.	n.e.	Meng, Zhao, Duan, Guan, & Zhao, 2018
	Red grape pomace	UAE (140 W, 37 KHz)	Lactic acid: sodium acetate (5:1)	Water	80%	0.3	90	80	3.32 mg/g	n.e.	n.e.	n.e.	Patsea, Stefou, Grigorakis, & Makris, 2017
	Apocynum venetum	UAE (200 W, 40 KHz)	1-Butyl-3-methylimidazoliu tetrafluoroborate [C_4mim][BF_4]	Water	72.43%	0.05	20	RT	93.35%	BE	n.d.	ABS	Tan, Yi, Wang, Zhou, & Wang, 2016
	Cercis chinensis	UAE (n.d.)	1-Butyl-3-methylimidazolium tetrafluoroborate [C_4mim][BF_4]	Ethanol	70%	0.03	50	n.d.	11.37 mg/g	n.e.	n.e	n.e.	Shi, He, Li, Li, & Kang, 2018
	Abutilon theophrasti	MAE (534 W) UAE (50 W)	1-Butyl-3-methylimidazolium bromide [C_4mim]Br	Water	2,000 mM	0.03	12	60	5.76 mg/g	n.e.	n.e.	n.e.	Zhao et al., 2014
	Sea buckthorn	MAE (600 W, 2,450 MHz)	1,4-Butanediol: cholinium chloride (3:1)	Water	20%	0.05	17	64	20.82 mg/g	SPE	91.15	SPE	Cui et al., 2018
	Bauhinia championii	MAE (800 W, 2,450 MHz)	1-Butyl-3-methylimidazoliu bromide [C_4mim]Br	Water	2,000 mM	0.05	10	70	0.2207 n	n.e.	n.e.	n.e.	Xu et al., 2012
	Chrysanthemum morifolium	MAE (450 W, 2,450 MHz)	1-Dodecyl-3-methylimidazolium bromide [C_{12}mim]Br	Water	100 mM	0.02	6.5	RT	1.02 mg/g	n.e.	n.e.	n.e.	Zhou et al., 2015

(Continues)

TABLE 1 (Continued)

Class of pigments/ pigment	Biomass	Extraction operational conditions: Method (conditions)	Solvent	Co-solvent	Concentration in the co-solvent	$R_{(S/L)}$	Time (min)	T (°C)	Yield	Polishing and purification strategies: Polishing	Recovery of the solvent (%)	Purification	References
	Chrysanthemum morifolium	MAE (450 W, 2,450 MHz)	1-Dodecyl-3-methylimidazolium bromide [C_{12}mim]Br	Water	200 mM	0.04	3	RT	1.05 mg/g	n.e.	n.e.	n.e.	
	Equisetum palustre	NPCE (−0.07 MPa)	Cholinium chloride-betaine hydrochloride-ethylene glycol (1:1:2)	Water	80%	0.04	20	60	89.25%	n.e.	n.e	SPE	Qi et al., 2015
	Cajanus cajan	NPCE (−0.07 MPa)	1-Octyl-3-methylimidazolium bromide [C_8mim]Br	Water	530 mM	0.05	15	74	0.5-5 mg/g	n.e.	n.e.	n.e.	Duan et al., 2013
	Scutellaria baicalensis	UHPE (400 MPa)	Cholinium chloride:lactic acid (1:1)	Water	60%	0.009	4	n.r.	116.8 mg	n.e.	n.e.	n.e.	Wang et al., 2018
	Ponkan peels (*Citrus reticulata*)	Pretreatment of the biomass using alternative solvents	Choline leucine	Water	54%	0.06	100	25	1.33%	TP	n.d.	ABS	Wang et al., 2016
Carotenoids	Carrots	UAE (22.5 W/cmš)	Sunflower oil	n.r.	100%	0.2	20	40	334.75 m	n.r.	n.r.	n.r.	Li et al., 2013
	Bactris gasipaes fruits (Peach palm by-product)	UAE (1,528 W/mš)	Sunflower oil	n.r.	100%	0.25	30	35	1.63 mg/g	n.r.	n.r.	n.r.	Ordóñez-Santos et al., 2015
		UAE (400 KHz, 400 W/mš)	Sunflower oil	n.r.	100%	0.16	5	RT	125 mg/L	n.r.	n.r.	n.r.	de Souza Mesquita, Neves, et al., 2020

(Continues)

TABLE 1 (Continued)

		Extraction operational conditions								Polishing and purification strategies			
Class of pigments/ pigment	Biomass	Method (conditions)	Solvent	Co-solvent	Concentration in the co-solvent	$R_{(S/L)}$	Time (min)	*T* (°C)	Yield	Polishing	Recovery of the solvent (%)	Purification	References
	Punica granatum (Pomegranate waste)	UAE (20 KHz, 130 W/mš)	Sunflower oil	n.r.	100%	0.10	30	51.5	3.255 mg/g	n.r.	n.r.	n.r.	Goula et al., 2017
	Shrimp waste	UAE (75 W/m^2)	1-Propylamine-3-methylimidazoliu bromide [C_3NH_2mim]Br	Ethanol	500 mM	0.1	60	RT	92.7 µg/g	n.e.	n.e.	SPE	Bi et al., 2010
	Bactris gasipaes fruits (Peach Palm by product)	UAE (400 KHz, 400 W/mš)	1-Butyl-3-methylimidazolium tetrafluoroborate [C_4mim][BF_4]	Ethanol	50%	1.0	48	RT	143 µg/g	TP	94	n.e	de Souza Mesquita, Ventura, et al., 2019
	Orange peels (waste)	UAE (400 KHz, 400 W/mš)	1-Butyl-3-methylimidazoliu chloride [C_4mim]Cl	Ethanol	33%	0.33	30	RT	32.08 µg	SPE	63.8	SPE	Murador, Braga, et al., 2019
	Tomatoes (waste)	UAE (400 KHz, 400 W/mš)	1-Butyl-3-methylimidazolium chloride [C_4mim]Cl	Ethanol	33%	0.25	60	0	8 µg/g	TP	90	n.e.	Martins & de Rosso 2016
	Rhodotorula glutinis	Rotatory orbital shaking (30 rpm)	3-Diethyl amino propylamine-hexanoic acid	Water	80%	x	60	25	333 µg/g	TP	100	TPP	Mussagy et al., 2019
	Neochloris oleoabundans	Rotatory elliptical shaking (50 rpm)	Tributyl(tetradecyl) phosphonium Chloride [$P_{4,4,4,14}$]Cl	Water	250 mM	0.025	60	RT	1.60 mg/g	Ultrafiltration	n.e.	ABS	Ruiz et al., 2020
	Pandalus borealis (shrimp waste)	Rotatory orbital shaking (400 rpm)	Methyl ester of sunflower oil and sunflower oil	n.r.	100%	0.1	60-160	70	32.9 mg/	n.r.	n.r.	n.r.	Parjikolaei et al., 2015

(Continues)

TABLE 1 (Continued)

Class of pigments/ pigment	Biomass	Extraction operational conditions: Method (conditions)	Solvent	Co-solvent	Concentration in the co-solvent	$R_{(S/L)}$	Time (min)	*T* (°C)	Yield	Polishing and purification strategies: Polishing	Recovery of the solvent (%)	Purification	References
	Bactris gasipaes fruits (Peach Palm by product)	Rotatory elliptical shaking (80 rpm)	1-Decyl-trimethylammonium bromide $[N_{1,1,1,10}]Br$	Water	140 mM	0.15	8.2	RT	88.7 µg/g	Antis-olvent	93.2	n.e.	de Souza Mesquita, Martins, et al., 2020
	Sargassum muticum	Rotatory elliptical shaking (250 rpm)	Tomadol 25–7	Water	20 mM	0.1	90	RT	1.86 mg/g	n.e.	n.e.	n.e.	Vieira et al., 2017
	Sargassum muticum	Rotatory elliptical shaking (250 rpm)	Pluronic P-123	Water	10 mM	0.02	90	RT	1.10 mg/g	n.e.	n.e.	n.e.	Vieira et al., 2017
	Sargassum muticum	Rotatory elliptical shaking (250 rpm)	Tween 20	Water	46 mM	0.02	140	RT	2.78 mg/g	n.e.	n.e.	n.e.	Vieira & Ventura, 2019
	Sargassum muticum	Rotatory elliptical shaking (250 rpm)	Sodium dodecyl sulfate (SDS)	Water	123 mM	0.04	90	RT	8.44 mg/g	n.e.	n.e.	n.e.	Vieira et al., 2018
	Haematococcus pluvialis	Pretreatment of the biomass using alternative solvents	1-Ethyl-3-methylimidazoliu dibutyl phosphate	Water	40%	4.7	90	45	>70%	Centrifu-gation	100	n.e	Desai et al., 2016
	Haematococcus pluvialis	Pretreatment of the biomass using alternative solvents	1-Ethyl-3-methylimidazolium-based ILs	Water	6.7%	1	60	30	>99%	n.e	n.e	n.e	Choi et al., 2019

(Continues)

TABLE 1 (Continued)

Class of pigments/ pigment	Biomass	Extraction operational conditions: Method (conditions)	Solvent	Co-solvent	Concentration in the co-solvent	$R_{(S/L)}$	Time (min)	*T* (°C)	Yield	Polishing and purification strategies: Polishing	Recovery of the solvent (%)	Purification	References
	Haematococcus pluvialis	Pretreatment of the biomass using alternative solvents	1-Butyl-3-methylimidazolium chloride $[C_4mim]Cl$	Water	40%	0.1	60	40	>80%	Centrifugation	n.d.	n.e	Liu et al., 2018
Chlorophylls	*Chlorella vulgaris*	Pretreatment of the biomass using alternative solvents	1-Ethyl-3-methylimidazoliu ethylsulfate $[C_2mim][EtSO_4]$	Water	100%	0.1	60	RT	17 µg/g	Centrifugation	98	n.e.	Orr, Plechkova, Seddon, & Rehmann, 2015
	Spinach leaves	Rotatory elliptical shaking (600 rpm)	Polyethylene glycol (PEG C11-C13 9EO's)	Water	12.4 mM	0.07	30	41	0.94 mg/g	n.e.	n.e.	ABS	Leite et al., 2017
	Neochloris oleoabundans	Rotatory elliptical shaking (50 rpm)	Tributyl(tetradecy Chloride $[P_{4,4,4,14}]Cl$	Water	250 mM	0.025	60	RT	2.90 mg/g	Ultrafiltration	n.e.	ABS	Ruiz et al., 2020
	Ulva spp.	Rotatory elliptical shaking (80 rpm)	Tributyl(tetradecyl)phosphonium Chloride $[P_{4,4,4,14}]Cl$	Water	250 mM	0.01	30	RT	5.96 mg/g	n.e.	n.e.	n.e.	de Souza Mesquita, Martins et al., 2020
Curcuminc	*Curcuma longa*	Pretreatment of the biomass using alternative solvents	1-Dodecyl-3-methylimidazoliu bromide $[C_{12}mim]Br$	Water	200 mM	0.6	1	RT	90.45% to 105.04%	Centrifugation	n.d.	n.e.	Xu et al., 2016
	Curcuma longa	Stirring	*N,N*-Dipropyl ammonium-*N*′, *N*′-dipropylcarbamate (DPCARB)	n.r.	100%	0.03	120	25	3.58%	Distillation	100	Chromatography	Sahne et al., 2017

(Continues)

TABLE 1 (Continued)

		Extraction operational conditions								Polishing and purification strategies			
Class of pigments/pigment	**Biomass**	**Method (conditions)**	**Solvent**	**Co-solvent**	**Concentration in the co-solvent**	**$R_{(S/L)}$**	**Time (min)**	**T (°C)**	**Yield**	**Polishing**	**Recovery of the solvent (%)**	**Purification**	**References**
	Curcuma longa	stirring	Citric acid:glucose (1:1)	Water	85%	0.1	30	50	21.18 mg/g	SPE	n.d.	SPE	Liu, Li, et al., 2019
Pigmented proteins: GFP, phycocyanin, and phycoerythrin	*Escherichia coli*	Rotatory orbital shaking (30 rpm)	Tributyl(tetradecy Chloride $[P_{4,4,4,14}]Cl$	Water	100 mM	0.025	30	RT	n.d.	n.e.	n.e.	ABS	Martins et al., 2018
	Gracilaria sp.	Elliptical homogenization	Cholinium chloride	Water	1,000 mM	0.7	20	RT	0.3 mg/g	n.e.	n.e.	n.e.	Martins et al., 2016
	Spirulina platensis	UAE (25 KHz)	2-Hydroxy ethylammonium acetate, 2-hydroxy ethylammonium formate and $[C_4mim]Cl$	Water	100 mM	0.12	30	25	14.91 mg	n.e.	n.e.	n.e.	Rodrigues et al., 2018
Anthocyanins	Wine	UAE (37 KHz, 341.5 W)	Cholinium chloride:malic acid (2:1)	Water	64.6%	0.1	30.6	35	6.55 mg/g	n.e.	n.e.	n.e.	Bosiljkov et al., 2017
	Grape skin (waste)	UAE	Citric acid:maltose (4:1)	Water	76.20%	0.12	9.23	RT	63.36 mg	n.e.	n.e.	n.e.	Jeong et al., 2015
	mulberry	HSH-CBE (12,000 rpm, −0.08 MPa)	Chloride–citric acid–glucose (1:1:1)	Water	70%	0.05	60	45	6.05 mg/g	n.e.	n.e.	n.e.	Guo et al., 2019

(Continues)

TABLE 1 (Continued)

		Extraction operational conditions								Polishing and purification strategies			
Class of pigments/ pigment	Biomass	Method (conditions)	Solvent	Co-solvent	Concentration in the co-solvent	$R_{(S/L)}$	Time (min)	T (°C)	Yield	Polishing	Recovery of the solvent (%)	Purification	References
	Catharanthus roseus	Stirring (n.r.)	1,2-Propanediol:cholinium chloride (2:1) and lactic acid:glucose (5:1)	Water	75%	0.03	30	40	55%	n.e.	n.e.	n.e.	Dai et al., 2016
	Grape-pomace	UAE (50 W) and MAE (300 W) combined	Cholinium chloride:citric acid (2:1)	Water	70%	0.03	10	< 80	1.77 mg/g	SPE	94.78	n.e.	Panić et al., 2019
	Grape skin (waste)	Stirring	1-Ethyl-3-methylimidazolium bromide [C_2mim]Br	Water	250 mM	0.04	240	RT	17.9 mg/g	n.e.	n.e.	n.e.	Ćurko et al., 2017
	Grape-pomace	Stirring	1-Ethyl-3-methylimidazoliu acetate [C_2mim]OAc	Water	12.5%	30.6	60	35	3.58 mg/g	n.e.	n.e.	ABS	Lima et al., 2017
Violacein	*Yarrowia lipolytica*	Rotatory elliptical shaking (1,000 rpm)	Tetrabutylammonium chloride [$N_{4,4,4,4}$]Cl	Water	100 mM	0.03	30	25	44.9%	AMTPS	n.d.	BE	Schaeffer et al., 2019
	Yarrowia lipolytica	Rotatory elliptical shaking (80 rpm)	Tween 20	Water	50 mM	0.025	240	RT	111 µg/mL	n.d.	n.d.	ABS	Kholany et al., 2019

Abbreviations: ABS, aqueous-biphasic system; AMTPS, aqueous micellar two-phases systems; BE, back-extraction; HSH-CBE, high-speed homogenization and cavitation-burst extraction; MAE, microwave-assisted extraction; n.d., not described; n.e., not evaluated; n.r., not required step; NPC, negative-pressure cavitation; RT, room temperature; SPE, solid-phase extraction; TP, thermal precipitation; TPP, three-phase partitioning; UAE, ultrasound-assisted extraction; UHPE, ultrahigh-pressure extraction.

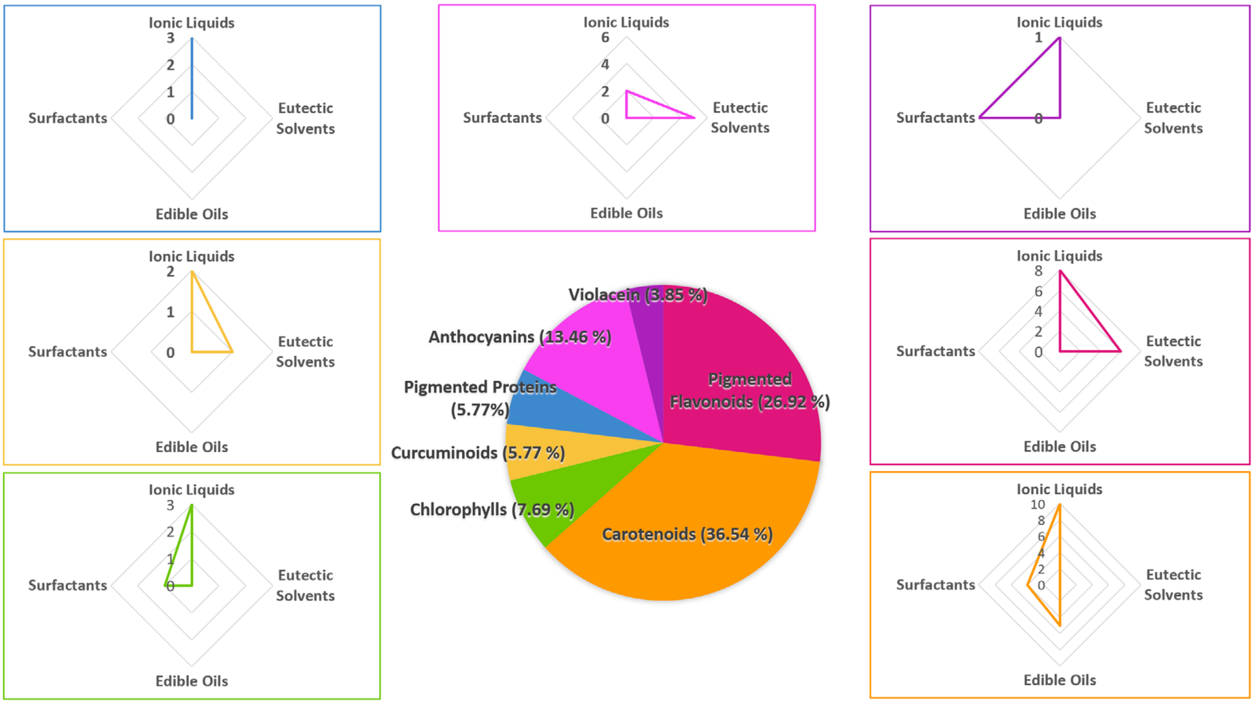

FIGURE 5 Distribution of the works dealing with each alternative solvent for the extraction of natural pigments. The radial graphs represent the number of scientific works dealing with the different alternative solvents, namely, ionic liquids, eutectic solvents, surfactants, and edible oils

presence of different substituent groups on the aglycone structure (Kasprzak, Erxleben, & Ochocki, 2015). These pigments display a very important role in human health, as natural antioxidants and anti-inflammatory agents. Some alternative homogenization methods like ultrahigh-pressure homogenization (UHPE) and negative-pressure cavitation (NPCE) were already used for the extraction of flavonoids, differing from other works using the most common ultrasound-assisted extraction or shaking homogenization. According to Wang, Ma, Cheng, Wang, and Zhang (2018), the UHPE technique (400 MPa, $R_{(S/L)}$ of 0.009, for 4 min) using the eutectic solvents based in cholinium chloride:lactic acid (1:1) was applied to promote a high yield of extraction of flavonoids (116 mg/g) from *Scutellaria baicalensis*, a Chinese herb used as infusions drinks. In addition to the contribution of eutectic solvents in the disruption of plant cells, this homogenization technique has many positive effects, such as the shorter processing times, lower energy consumption, and fewer impurities in the liquid extract. Moreover, it can be conducted at low temperatures, thus preventing chemical degradation reactions. The NPCE extraction, which consists of a device where the cavitation is generated by negative pressure with continuous nitrogen flow into the solid–liquid system, able to increase the turbulence in the medium and, consequently, the mass transfer coefficient while operating at low temperatures, was used by Qi et al. (2015). Quercetin from *Equisetum palustre* was extracted using a NPCE device by applying cholinium chloride:betaine hydrochloride:ethylene glycol (1:1:2) as the solvent, at −0.07 MPa, 60 °C, $R_{(S/L)}$ of 0.04, for 20 min. Similarly, but using an IL-based NPCE at −0.07 MPa, 1-octyl-3-methylimidazolium Bromide—[C_8mim]Br—(30 mM, at 74 °C, $R_{(S/L)}$ of 0.05, and extraction time of 15 min), genistein and apigenin flavonoids were recovered from pigeon pea roots (*Cajanus cajan*) (Duan et al., 2013). The authors reported the extraction of flavonoids from the group of apigenin, kaempferol, and quercetin with a yield of extraction from 0.5 to 5 mg/g. Although the authors consider the UHPE- and NPCE-assisted extractions as cost-effective and environmentally friendly techniques, no other works have done such evaluations to infer on the economic and environmental viability, which is indeed transversal to the majority of the works focusing the use of alternative solvents to recover pigments from biomass or residues of any kind. Furthermore, more conventional techniques were also used for the recovery of yellowish flavonoids from food matrices. For these proposes, there is no consensus on the best type of alternative solvent applied, and ILs (namely, cholinium- and imidazolium-based) or even eutectic solvents were already used. As an example, a cholinium leucine-based IL was used for the recovery of flavonoids from Ponkan peels (*Citrus reticulata*), by applying a pretreatment step to the biomass by promoting the biomass contact with

IL the during 100 min ($R_{(S/L)}$ of 0.06, 25 °C). At the end of this step, the authors have achieved an extraction of 1.33% of the total flavonoid content from Ponkan peels (Wang, Chang, Tan, & Li, 2016). Eutectic solvents were also used for the extraction of flavonoids from different food matrices, namely, cranberry, grapes, plum, orange peel, onion, broccoli, mustard, rosemary, and black pepper. Seventeen different aqueous solutions of eutectic solvents based on choline chloride, acetylcholine chloride, choline tartrate, betaine, and carnitine in different concentrations and molar ratios were evaluated. It was concluded that depending on the food matrix structure, and the food water content, some methods are quite effective, even for small concentrations of the alternative solvent (Bajkacz & Adamek, 2018). However, despite the interesting results obtained in the extraction of phenolic colorants in this large plethora of food matrices, some attention needs to be paid to nomenclatures exposed in some articles, especially when authors call eutectic mixtures as "deep" eutectic solvents without proper evaluation of the real melting point by thermodynamic assays.

2.2 | Carotenoids and chlorophylls

In a color spectrum similar to some flavonoids, the carotenoids also give yellowish–orange–reddish colors to products. However, in most of the cases in which the biomass produces more than one class of hydrophobic pigments, namely, carotenoids and chlorophylls, both are simultaneously extracted, because both classes are usually located in the same cell site, such as chromoplasts, chloroplasts, leucoplasts, and fat globules (Li & Yuan, 2013). Carotenoids and chlorophylls (green lipophilic pigments) are recognized by their many benefits to human health, mainly considering their antioxidant and anti-inflammatory activities (Cervantes-Paz et al., 2014). Chlorophylls are naturally found in marine algae, leafy vegetables, and some fruits. Carotenoids, on the other hand, are usually extracted from tomatoes, carrots, pumpkins, mangos, cassava, and other fruits (Biazotto et al., 2019; Minich, 2019), besides shrimp shell wastes and macro and microalgae. Thus, for having a similar structural hydrophobicity, their extraction is normally driven in a more efficient way using tensioactive solvents. Thus, the summary of what is being displayed in literature is the selection of tensioactive ILs and common surfactants, and more specifically those from the imidazolium family, conjugated with the tetrafluoroborate ($[BF_4]^-$), chloride (Cl^-), bromide (Br^-), and phosphate ($[PO_4]^{3-}$) anions, and cations conjugated with elongated alkyl chains substituted (>C7). Generally, organic solvents like acetone and ethanol are used as a control to compare the extraction efficiency of the proposed alternative solvents (Bi, Tian, Zhou, & Row, 2010; Choi et al., 2019; de Souza Mesquita, Ventura, et al., 2019; Desai, Streefland, Wijffels, & Eppink, 2016; Martins & de Rosso, 2016; Murador, Braga, Martins, Mercadante, & de Rosso, 2019; Vieira & Ventura, 2019; Vieira et al., 2017). The ultrasound-assisted extraction is one of the conventional techniques most widely used when ILs are used as solvents. Usually, the ultrasonic waves promote the highest yield of pigment extraction due to its intensity promoting the biomass disruption, and consequently, the release of pigments to the solvent. Additionally, some ILs have been selected regarding their additional capacity to promote the strong disruption of cells and/or dissolution of the compounds of interest, which facilitates the pigments' extraction (Choi et al., 2019; Desai et al., 2016; Liu, Zeng, Cheng, Liu, & Aadil, 2018). Also, some works report the positive action of the ILs to stabilize the carotenoids (better thermal stability and longer half-life time [$t_{1/2}$]) (Bi et al., 2010; de Souza Mesquita, Ventura, et al., 2019; Murador, Braga, et al., 2019). In another perspective, some works were reported using ILs to recover carotenoids from algae, to overcome the high structural rigidity of the biomass, hampering the assess to carotenoids (Gerken, Donohoe, & Knoshaug, 2013; Poojary et al., 2016). The cell wall and plasmatic membrane surrounding the cell, as well as the chloroplast membranes, are strong barriers, thus hindering the carotenoids release during the conventional extraction approaches (Poojary et al., 2016). Aqueous solutions of imidazolium-based ILs like 1-butyl-3-methylimidazolium chloride ($[C_4mim]Cl$) and 1-propyl-3-methylimidazolium bromide ($[C_3mim]Br$) were investigated as solvents to extract carotenoids from algae biomass and, after being compared with the conventional ethanolic solutions, it was concluded that the alternative process based on IL was much more energetically efficient (>50%) (Desai et al., 2016; Liu et al., 2018; Liu, Yue, Zeng, Cheng, & Aadil, 2019). Additionally, some authors concluded that the use of ILs are a better approach to dissolve the algae biomass, even when compared with the use of other strategies like electric fields and ultrasound techniques (Liu et al., 2018).

Recently, edible oils have been used to carry the carotenoids extraction (Goula, Ververi, Adamopoulou, & Kaderides, 2017; Ordóñez-Santos, Pinzón-Zarate, & González-Salcedo, 2015; Parjikolaei, El-Houri, Fretté, & Christensen, 2015). Although not considered as "classic" alternative solvents like eutectic solvents and ILs, the edible oils have also advantages over conventional organic solvents (COS), which usually have high volatility. From a food perspective, the use of edible oils as solvents for extraction is economically advantageous, because they can be maintained in the extract rich in carotenoids, thus being directly included in the new formulations (Yara-Varón et al., 2017). Following this approach, the downstream process is much simpler, safer, and cost-efficient, because there is no need for further polishing processes (Saini & Keum, 2018). The main results obtained by using edible oils (vegetable oil as mentioned by the authors) have shown average extraction yields between 60% and 90% (Goula et al., 2017). For the recovery of carotenoids from different kinds of food matrices, some recent studies have reported high extraction efficiencies using edible oils, namely, sunflower, flaxseed, and groundnut oil (Baria, Upadhyay, Singh, & Malhotra, 2019; Li, Fabiano-Tixier, Tomao, Cravotto, & Chemat, 2013; Ordóñez-Santos et al., 2015). Recently, it was performed a simple and fast method of carotenoids' extraction from the Amazonian *Bactris gasipaes* waste using sunflower oil assisted by applying ultrasonic homogenization technique (operational conditions: 400 W, 40 kHz, $R_{(S/L)}$ of 0.16, 5 min). Additionally, this carotenoid-rich oil was used in the development of a mayonnaise sauce with high antioxidant activity, thermal stability, and sensorial acceptance scores (color, aroma, taste, texture, and overall acceptance). Also, considering the sensory analysis carried out by 50 panelists, the new product had a high buy intention, demonstrating the commercial potential of the developed sustainable product. Thus, applying both, biorefinery and circular economy concepts, food waste was converted into a new food product with high nutritional and commercial value (de Souza Mesquita, Neves, Pisani, & de Rosso, 2020).

Surfactants are other classes of alternative solvents being investigated. Due to the high hydrophobicity of the carotenoids and chlorophylls, nonionic surfactant-based solutions (as Tween®, Tomadol®, Triton®, and Pluronic families) have demonstrated promising advantages considering their use in aqueous solution, thus reducing the environmental impact of the overall process, because hydrophobic pigments, such as carotenoids and chlorophylls, are usually extracted by COS (Leite et al., 2017; Vieira et al., 2018). Some recent works have used surfactant-based and tensioactive-IL aqueous solutions as solvents to obtain carotenoids and chlorophylls from food matrices (de Souza Mesquita, Martins, et al., 2020; Vieira & Ventura, 2019; Vieira et al., 2018). In these works, efficient and more selective processes were developed allowing to obtain twice as much in the yield of extraction of carotenoids, when compared with the organic solvents-mediated extractions. In a similar approach, Leite et al. (2017) extracted 0.94 mg/g of chlorophylls *a*/*b* from spinach leaves using aqueous solutions of nonionic surfactants, namely, polyethylene glycol (PEG C11-C13 9EO's). Thus, there are new possible technologies able to be applied to food, nutraceutical, cosmetic, and pharmaceutical products, without requiring an additional polishing step. For example, by applying carotenoids obtained from *Bactris gasipaes* fruits waste, it was developed a chitosan-based film as a possible alternative food-packaging materials using carotenoids extracted by aqueous solutions of tensioactive ammonium IL, 1-decyl-trimethylammonium bromide ($[N_{1,1,1,10}]Br$), with excellent results regarding their physical and mechanical parameters, as well as high antioxidant activity (de Souza Mesquita, Martins, et al., 2020). Additionally, it was concluded that besides the higher performance of the alterative process compared to COS, the physical–chemical and mechanical parameters of the films loaded with carotenoids extracted by IL were significantly better. Bio-based materials, especially those with functional activity, are nowadays considered as a hot topic, and a proof of concept of Biorefinery and Circular Economy concepts, because there is a complete valorization of the raw materials (Cazón, Velazquez, Ramírez, & Vázquez, 2017; Geyer, Jambeck, & Law, 2017). Recently, also as an example of a multiproduct Biorefinery concept, a single-step extraction process was performed to recover chlorophylls (*a* and *b*) and lutein (carotenoid) from the microalgae *Neochloris oleoabundans* using a tensioactive IL (Ruiz et al., 2020). An aqueous solution of tributyl-1-tetradecyl phosphonium ($[P_{4,4,4,14}]Cl$) was selected as the best solvent to recover the pigments (lutein: 1.60 ± 0.06 mg/g; chlorophylls *a* and *b*: 2.90 ± 0.05 mg/g) under the operational conditions optimized of $R_{(S/L)}$ of 0.025 and IL concentration of 250 mM, which represented at least twice the yield when compared to methanol-mediated extractions, and 10 times better results than the ones using water as the solvent. Besides, the authors have achieved high recovery of proteins (105 ± 2 mg/g) and carbohydrates (60 ± 9 mg/g), also important natural value-added compounds used in the production of food materials, food-packaging materials, and food supplements. In a different perspective, but also as an example of how to develop the Biorefinery concept, Orr, Plechkova, Seddon, and Rehmann (2017) have designed a simple and time-efficient process for lipids extraction from *Chlorella vulgaris* using an aqueous solution of 1-ethyl-3-methylimidazolium ethylsulfate ($[C_2mim][CH_3CH_2SO_4]$), which was also able to recover 0.94 mg/g of chlorophylls *a* and *b*. The authors have also developed a polishing step by centrifugation, recovering 98 % of the used IL and recycling for more four new extractions procedures, which represents an important task toward the economic viability of using an IL as a solvent. Recently, the economic viability has been discussed by Martins et al. (2020), who developed an alternative process mediated by an aqueous solution of $[P_{4,4,4,14}]Cl$ to extract chlorophylls from *Ulva* spp. This work allowed the authors to conclude that, besides having developed a more efficient process in terms of chlorophyll extraction yield, it was also considered more cost-effective (expressed in Cost of goods per $mg_{chlorophyll}$) and simpler (faster and with less raw materials), highlighting the promising role alternative solvents like ILs can have on the development of Biorefinery strategies. Moreover, this works also demonstrate that the processes under development in the laboratories all over the world should be designed not only considering their selectivity and yields of extraction of purification level, but also in terms of their economic and environmental viability, otherwise these works and merely academic exercises meaning inappropriate for future industrialization.

2.3 | Curcuminoids

Curcuminoids, obtained from the rhizomes of turmeric *Curcuma longa* (Zingiberaceae), are orange–golden pigments commonly used as a food colorant, with relevance in terms of their antioxidant, anti-inflammatory, and anticancer (Kocaadam & Sanlier, 2017) properties. Curcuminoids are constituted mainly by three lipophilic polyphenolic compounds, the bisdemethoxycurcumin, demethoxycurcumin, and curcumin (the most well-known) (D'Archivio, Maggi, & Ruggieri, 2018). Like carotenoids and chlorophylls, normally curcuminoids are not extracted using water, being the common organic solvents the ones most used in the curcuminoids' recovery. In 2016, the extraction of curcumin was evaluated by using aqueous solutions of the 1-dodecyl-3-methylimidazolium bromide ([C_{12}mim]Br) at 200 mM by applying the IL to pretreat the biomass, reaching final recoveries up to 90% of total content in curcuminoids. The authors reported that the ultrasound approach using traditional organic solvents promoted low selectivity and extraction yields. Moreover, higher concentrations of the selected IL contributed to increasing the system's viscosity, which has influenced the mass transfer of curcuminoids, leading to the poor interaction of IL with the cells (Xu et al., 2016). In 2017, a different IL was applied (a carbamate-based IL) on the extraction of curcumin, the *N*,*N*-dipropyl ammonium-*N'*,*N'*-dipropylcarbamate. After the optimization of the process' conditions ($R_{(S/L)}$ of 0.03, at 25 °C within 120 min), yield of 3.58% of curcumin was achieved using a shaker homogenization (Sahne, Mohammadi, Najafpour, & Moghadamnia, 2017). Eutectic solvents were also tested in the extraction curcuminoids by Liu, Li, et al. (2019). A process using a simple stirring method with an aqueous solution of citric acid:glucose (1:1 [85% in water]), with a $R_{(S/L)}$ of 0.1, 50 °C during 30 min was able to achieve a yield of extraction of total curcuminoids of 21.18 mg/g.

2.4 | (Pigmented) proteins

The phycobiliproteins, mainly the R-phycoerythrin (R-PE) and C-phycocyanin, are the principal class of pigments found in red macroalgae and cyanobacteria. Those are hydrophilic proteins linked to a chromophore called *bilin* responsible for a fluorescent pink color of R-PE and blue color of C-phycocyanin (Dumay & Morançais, 2016). Conventionally, these pigmented proteins have been extracted by maceration in buffers or aqueous solvents, an ecofriendly and effective method, although with low yields (Cuellar-Bermudez et al., 2015; Vernès, Granvillain, Chemat, & Vian, 2015). Despite the investigations carried out using other techniques like ultrasounds, microwaves, enzymatic digestions, and supercritical fluids, the application of ILs was recently assessed to recover phycobiliproteins (Kadam, Tiwari, & O'Donnell, 2013; Michalak & Chojnacka, 2014). Recently, an optimized method was developed using an aqueous solution of cholinium chloride in an orbital homogenization for recovering phycobiliproteins, and particularly, the R-PE from the red macroalga *Gracilaria gracilis*. Besides the good yields of extraction (46.5% better than the conventional method), the method was also selective because it avoided the extraction of chlorophylls (Martins, Vieira, et al., 2016). More recently, Rodrigues, de Castro, de Santiago-Aguiar, and Rocha (2018) used a protic-based IL (2-hydroxyethyl ammonium) through an acid–base neutralization reaction. A process using ultrasound-assisted extraction as the main homogenization technique for extraction of phycocyanin from *Spirulina platensis* was developed, being reported the extraction capacity of the protic IL over the conventional sodium phosphate buffer and the commercial 1-Butyl-3-methylimidazolium chloride ([C_4mim]Cl). However, it was not allowed to reach a final product of phycocyanin with food-grade purity, which makes it difficult for its application in this sector. Recently, through an integrated chain, supercritical CO_2 was applied to recover carotenoids and chlorophylls (in this case considered as contaminants) from *Spirulina* sp. before extracting the C-phycocyanin. After this step, the authors performed the extraction of C-phycocyanin using ethanolic solutions, and the high purity of the blue pigment was achieved ($A_{620}/A_{280} = 2.2$) (Marzorati, Schievano, Idà, & Verotta, 2020). Recently, also envisioning the recovery of pure C-phycocyanin, in this time from *Anabaena cylindrica*, Sintra et al. (2020) developed a process mediated by tensioactive ILs and aqueous solutions of surfactants and concluded that, despite the high-performance credential of the selected ILs, the aqueous solution of Na-phosphate buffer (20 mM, $R_{(S/L)}$ of 0.1, 35 °C, under rotatory elliptical shaking at 1,500 rpm) was the one showing the best results on the C-phycocyanin extraction, around 60 $mg_{phycocyanin}/g_{fresh\ biomass}$. Additionally, in this work, it was also recovered in a second-step of extraction 1.46 $mg_{chlorophylls}/g_{fresh\ biomass}$ by applying ethanol as solvent ($R_{(S/L)}$ of 0.13, 45 min, 25 °C, under rotatory elliptical shaking at 1,500 rpm). Even ethanol is not considered an alternative solvent, this work respected the Biorefinery concept, not treating the chlorophylls as a contaminant, but as an added-value product to be applied as well in the food sector.

Some fluorescent proteins are found in microorganisms, which represents a great advantage over the extraction from plants, animals, and general wastes as sources. Among those advantages, the easy genetic manipulation and independence of weather conditions can be highlighted (Venil, Aruldass, Dufossé, Zakaria, & Ahmad, 2014). The use of plant pigments has many processing drawbacks such as nonavailability across the year and the variations on the composition between different batches (Rao et al., 2017). Given that, green fluorescent protein (GFP) was isolated from the bioluminescent jellyfish *Aequorea victoria*, and commonly expressed by the *Escherichia coli* bacteria (Xia et al., 2002). The aqueous solution of the tensioactive $[P_{4,4,4,14}]Cl$ was used to develop an alternative solvent to obtain GFP from *E. coli* strains (BL 21). The IL was used to induce the GFP release, not only because of its high capacity to disrupt the cell wall of the bacteria but also due to its potential to easily dissolve the protein. The IL disruption method allowed also to overcome the scale-up limitations of ultrasonication (Martins et al., 2018).

2.5 | Anthocyanins

Anthocyanins are relevant pigments for both the food and pharmaceutical industries and appear with red, blue, and purple colors, which naturally occur in many species of fruits, flowers, and other vegetables (Castañeda-Ovando, de Lourdes Pacheco-Hernández, Páez-Hernández, Rodríguez, & Galán-Vidal, 2009). Structurally, anthocyanins are similar to flavonoids, only differing on a protonated oxygen atom of the C-ring basic aglycone, also called as flavylium ion (2-phenylchromenylium) (Dangles & Fenger, 2018). Anthocyanins have numerous biological activities, namely, the improvement of cardiovascular and macular health, as well as their anticancer, antidiabetic, antioxidant, antimicrobial, and anti-inflammatory activities (Khoo, Azlan, Tang, & Lim, 2017; Salehi et al., 2020). Recently, it was investigated the mixture composed of citric acid:D-(+)-maltose (4:1) able to extract around 40 mg/g of total anthocyanins from *Catharanthus roseus*, twice the yield achieved by the conventional methanol-mediated method, while maintaining the stability of the pigments under heating (Dai, Rozema, Verpoorte, & Choi, 2016). Similar results regarding the thermal stability of anthocyanins were observed in a study using fresh mulberry samples (Guo et al., 2019). In this study, a high-speed homogenization and cavitation-burst extraction using chloride–citric acid–glucose (1:1:1 [v/v/v]; 30% of water) was performed under operational conditions applying $R_{(S/L)}$ of 0.05, 12,000 rpm, 30 min of extraction, and negative pressure (−0.08 MPa), yielding 1.24-fold higher to those ethanol-mediated extractions. Thus, in addition to the high extraction performance of alternative solvents, the eutectic mixtures could also improve the thermostability of the anthocyanins extracted from natural sources. Recently, envisioning a scale-up production of natural anthocyanins, and also the valorization of grape pomace, a downstream process was proposed by Panić, Gunjević, Cravotto, and Redovniković (2019) using cholinium chloride:citric acid (2:1 [v/v], with 30% of water) with simultaneous ultrasound/microwave-assisted extraction. A yield of extraction of anthocyanin of 1.77 mg/g was obtained and the sequential scale-up (50×) tested achieving high efficiency around 90%. Another study, also using grape pomace samples, but with aqueous solutions of IL, namely, 1-ethyl-3-methylimidazolium acetate ($[C_2mim][CH_3CO_2]$) (operational conditions fixed under magnetic stirrer for 60 min, 35 °C, and $R_{(S/L)}$ of 30.6 mg/mL), the authors obtained up to 3.58 mg/g of total anthocyanin content (Lima, Soares, Paltram, Halbwirth, & Bica, 2017). Thus, in this case, using grape pomace rich in anthocyanins, the IL-assisted extraction has provided significantly higher extraction yields than those obtained using eutectic solvents. In other work, a series of aqueous solutions of imidazolium-based ILs were tested to extract anthocyanins from grape skins (1-alkyl-3-methylimidazolium bromide $[C_nmim]Br$ and 1-alkyl-3-methylimidazolium hydrogen sulfate $[C_nmim][HSO_4]$). Concluding, high selectivity in the extraction of individual anthocyanins was achieved when ILs were used, namely, those composed of acetylated and glycosylated radicals (Ćurko et al., 2017).

More conventional methods of homogenization, namely, ultrasound-assisted extraction using eutectic solvents, were also reported to extract anthocyanins, as depicted by Jeong et al. (2015). In this work, anthocyanins were extracted from wastes of grape skins using the eutectic solvent composed of citric acid:maltose (4:1 [76.20%]). After optimizing the process' conditions ($R_{(S/L)}$ of 0.12, during 9.23 min, at room temperature), a maximum of 63.36 mg/g of anthocyanins was recovered. In a similar approach, Bosiljkov et al. (2017) also developed an ultrasound-assisted extraction using wine lees-lyophilized samples. The process was mediated by cholinium chloride:malic acid (1:1) (operational conditions of 341.5 W, 30.6 min, $R_{(S/L)}$ of 0.1, 35 °C, and water content of 35.4% [v/v]), providing more than the double of the yield compared to conventional organic solvent-mediated extraction. However, despite the satisfactory results obtained regarding the yield of extraction of anthocyanins, the study is designed under conceptual mistakes, because the screening of eutectic solvents was performed using water content of 50%, 75%, and 90% (v/v), which makes it impossible to form an eutectic solvent.

2.6 | Violacein

With the similar color of anthocyanins, violacein is also a purple/violet-pigmented molecule that can be obtained from microorganisms (Aranda, Montes-Borrego, & Landa, 2011), animals, and even glaciers (Lu et al., 2009), with great potential, especially in the food, textile, and toy industries (Durán et al., 2012). To improve the bioactivity and production of microbial pigments for their commercial use in pharmacological and medical fields, ecofriendly downstream processes are again preferred (Tuli, Chaudhary, Beniwal, & Sharma, 2015). Recently, an integrated platform to recover and purify violacein from the yeast *Yarrowia lipolytica* was mediated by applying a tunable anionic surfactant-based system (Schaeffer et al., 2019). By tuning the tetrabutylammonium chloride ($[N_{4,4,4,4}]Cl$) to sodium dodecyl sulfate (SDS) surfactant (0.1 M, 30 min), the solid–liquid extraction was performed using a rotatory elliptical shaking (1,000 rpm) and allowing to recover 44.9% of the violet pigment, allowing the subsequent separation of 100% of the extracted violacein from the protein content of the extract (69.4%). A second platform for recovery of violacein from *Y. lipolytica* was recently described by Kholany et al. (2019), also using an aqueous solution of common surfactant; at this time, a nonionic surfactant (Tween 20) was used for recovering 111 ± 3 µg/mL (operational conditions: $R_{(S/L)}$ of 0.025, 240 min, 50 mM), which represents at least twofold higher yield than an ethanol-mediated extraction, considered the main conventional organic solvent used for recovery violacein. In addition, in this work, the viability of the cells from *Y. lipolytica* after exposed to Tween 20 aqueous solution and ethanol was evaluated, and concluded that even after disruption of the cells with the surfactant solution, *Y. lipolytica* cells can regrow, proving the biocompatibility of Tween 20, which has not happened in the cells exposed to ethanol.

3 | PURIFICATION AND ISOLATION OF PIGMENTS

One of the main drawbacks associated with the food sector is the assurance of safer food and for that, it needs to be guaranteed the complete removal of solvents and other possible contaminants from the pigments-rich extracts. If the purification of the colorants means their separation from other contaminants simultaneously extracted (other classes of bioactive compounds present in the cells composition, e.g., membrane proteins, polysaccharides, lipids, and other), the pigments' isolation (or polishing) consists of their separation from the main solvents applied on the development of the downstream process. Some polishing techniques were already reported to separate bioactive molecules from ILs (Mai, Ahn, & Koo, 2014). However, few works under the scope of recovering natural pigments have investigated these procedures. Again in Table 1, the details of the methods used so far to promote the purification and polishing of pigments are detailed. As it can be analyzed, most of the works summarized in this table focus on the solid–liquid extraction procedures, not considering the integration of the subsequent steps normally required to obtain a final product with high quality and purity. If it is true that in some cases if the needs of the application in terms of purity are not so demanding, the final product obtained after the step of solid–liquid extraction may be sufficient, and in this case, does not make sense to progress to purification and polishing tasks. However, most of the works generally argue that the processes will be developed to obtain pigmented products able to be applied in high-demanding applications, which does not make sense in the end, if extra steps of purification and polishing are not included in the final process. Meanwhile, and despite the lack of research on the field, from a circular economy perspective, the recovery and recycling of the alternative solvents are essential steps, because the environmental impact (Clark et al., 2016; Marion et al., 2017; Sheldon, 2016) and cost of the integrated process may benefit the scale-up while guaranteeing their safe application in food sector (Sun et al., 2017; Zhou, Sui, et al., 2018), very important conditions for an industrial application. Purification and isolation of the pigments from the alternative solvents represent a final important step for their predictable applications (as shown in Figure 4), which is crucial for the food sector, due to the huge exigence for the consumer safety. Thus, and for the processes of extraction represented by low purity and low selectivity, purification assays are as important as required (solid–liquid) extraction units. In this sense, the main aim of these works includes the efficient separation of the target pigments from the main contaminants present in the extracts. Within this context and, included in the successful techniques applied to purify pigments obtained from natural resources, are the aqueous biphasic systems (ABSs). Some ABSs are considered biocompatible platforms because they are mainly composed of water (up to 70–90%) (Benavides, Rito-Palomares, & Asenjo, 2011). For their preparation, two-phase forming mutually immiscible components, for example, salt–polymer, salt–salt, and polymer–carbohydrate, that form two immiscible phases above a critical concentration (Pereira, Freire, & Coutinho,

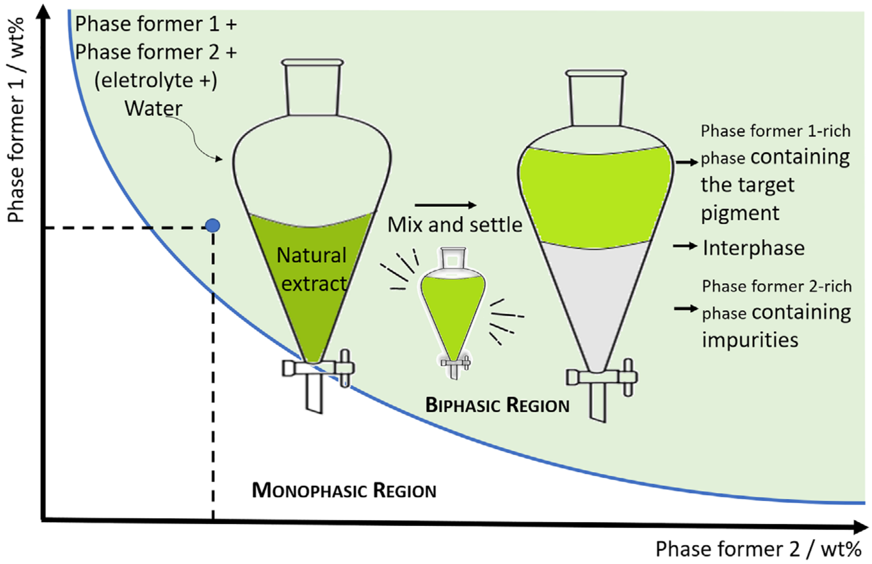

FIGURE 6 Schematic representation of a binodal curve and extraction performed using aqueous-biphasic systems (ABS)

2020) are mixed, as schematically represented in Figure 6. Their extractive capacity depends on several factors, mainly the biomolecules affinity for each aqueous phase, molecular size, and type of interactions between the biomolecule and the components of the system (Freire et al., 2012; Ventura et al., 2017). IL-based ABSs have been developed following different strategies regarding the addition of the ILs. In some cases, the ILs work as main phase formers; however, they can be considered as electrolytes, when just a small amount of ILs (1–5 wt%) is added to the system. These systems can be used to avoid problems, not exclusively, but mainly related to the poor polarity difference between both aqueous phases and high viscosity of the phases (Freire et al., 2012).

IL-based ABSs were reported for the first time in 2003 (Gutowski et al., 2003), and since then, the number of works using these systems is significantly increasing. The crescent interest in the IL-ABSs is related to the easier mass transfer between both phases by comparison with the most conventional polymeric ABS. Their high solvency power makes the authors believe that IL-ABSs can be applicable to the purification of a large range of molecules (Ventura et al., 2017). On the other hand, IL-ABS are considered as faster and more cost-effective purification technologies in comparison with traditional liquid–liquid extraction techniques. Engineering and existing equipment may be easily adapted to the process requirements allowing their expansion to scale-up separations, which is also a reality for proteins at an industrial level (Hatti-Kaul, 2000).

In 2013, a work using IL-ABS in the purification of pigments from natural resources was published (Ventura et al., 2013). In this work, it was intended to separate the red colorants from the remaining colorants and proteins in the fermented broth from *Penicillium purpurogenum* using systems composed of citrate buffer and IL. Systems based on tetraethylammonium bromide ($[N_{2,2,2,2}]Br$) were selected as the most promising systems with high selectivity and high protein removal (60.7%), by comparison with the polymer-based ABS. Among 2017 and 2018, three other publications arise. All have used similar purification processes to purify flavonoids (e Silva et al., 2017), GFP (dos Santos et al., 2018), and pigments involved in photosynthesis (Suarez Ruiz, Emmery, Wijffels, Eppink, & Van den Berg, 2018) from citrus juice, bacteria, and microalgae, respectively. In these works, ABSs composed of polymer and cholinium-based ILs were applied. Some authors have proposed a methodology to separate sugars from different flavonoids in a two-step methodology from citrus juice (e Silva et al., 2017). First, sugars were removed from rutin and naringin using a system composed of salt and the copolymer Pluronic L-35. The copolymer-rich phase containing the rutin and naringin was then applied in the formation of a second system by the addition of cholinium bicarbonate. At this point, the rutin and naringin were separated achieving recoveries of 89.6% and 32% for naringin and rutin, respectively. Additionally, dos Santos et al. (2018) proposed systems composed of polymers and cholinium chloride to purify GFP from other proteins present in the fermentation broth by applying ABS composed of cholinium chloride and polypropylene glycol 400. The authors achieved extraction efficiencies toward the cholinium chloride-rich phase of around 100% for GFP obtained with high purity. Recently, Suarez Ruiz et al. (2018) followed the use of polymer-IL-based systems to fractionate pigments from proteins, by processing a microalgae extract. A particular type of ABS, the three-phase partitioning systems (TPPS), was used in which the precipitation of biomolecules between the two phases (interphase) occurred. This precipitation allowed the separation and concentration of the proteins. With this process, the recycle step of the solvents was facilitated, being maintained the native conformation and structure of the proteins and pigments present. With the system composed of polyethylene glycol 400 and cholinium dihydrogen phosphate, the authors proved the high fractionation of proteins and pigments of *Neochloris oleoabundans* extracts, with 93% of the protein content in the interphase and bottom phase, and more than 97% of lutein in the top phase. Similarly, Mussagy, Santos-Ebinuma, Gonzalez-Miquel, Coutinho, and Pereira (2019) developed a TPPS based in protic ILs (3-diethylaminopropylamine-hexanoic acid), allowing the recyclability of the solvent for at least three more cycles of extraction, allowing at the same time the polishing of all-*trans*-*β*-carotene, torularhodin, and torulene from *Rhodotorula glutinis* by thermal precipitation with cold acetone.

Apart from the most conventional ABS, an alternative methodology, also based on ABSs, was carried out to fractionate phycobiliproteins from red macroalgae (Vicente et al., 2019) and violacein from *Yarrowia lipolytica* yeast, from the contaminant proteins present in the respective extracts (Schaeffer et al., 2019). In these works, the authors applied the concept of aqueous micellar two-phase systems (AMTPS), in which an aqueous solution of a surfactant (or more) above a certain concentration favors the formation of two aqueous phases (a surfactant-rich and a surfactant-poor phase) as a response to an increase in temperature. In these systems, ILs were applied as co-surfactants, meaning that they are present in a very small concentration, although facilitating the two-phase formation. The literature on the field has shown that the application of AMTPS with ILs as co-surfactants, for some specific processes, may enhance the performance of extraction, the selectivity of the extracted pigment, and/or reducing the use of other solvents (Vicente et al., 2019; Vicente, Lario, Pessoa, & Ventura, 2016). The authors have used these systems to develop a two-step method to fractionate phycobiliproteins from other nonfluorescent proteins, and in a second step, to separate R-PE from R-phycocyanin, the main phycobiliproteins composing the macroalgae treated (Vicente et al., 2019). In this work, the successful purification of each proteinic form was achieved, without compromising the structural integrity of proteins. A similar approach was explored for purple violacein (Schaeffer et al., 2019), but in this work, considering systems composed of common surfactant (SDS) and IL, namely, the tetrabutylammonium chloride $[N_{4,4,4,4}]$Br. In this system, IL plays an essential role because it is one of the main phase formers. This thermoresponsive system was successfully applied to the purification of violacein, for which, a high selectivity was obtained (meaning extracting more violacein and fewer contaminant proteins) by the integration of both extraction and purification units in a single step. Interestingly, and contrarily to the strategies previously described, this work also comprised a back-extraction step for the complete recovery and concentration of the violacein from the surfactant-rich phase to a menthol:thymol hydrophobic eutectic solvent (1:1) based on natural terpene precursors to isolate the target pigment, which is considered as common excipients in the pharmaceutical industry (Aroso et al., 2016).

Other strategies to isolate pigments from the alternative solvents were also described in the literature. As examples are the use of thermal precipitation (de Souza Mesquita, Ventura, et al., 2019), adsorbent columns in solid-phase extractions (SPE) (Murador, Braga, et al., 2019), antisolvents (de Souza Mesquita, Martins, et al., 2020), and back-extraction (Ruiz et al., 2020). In a recent work, de Souza Mesquita, Ventura, et al. (2019) proposed an efficient method to recover carotenoids from an Amazonian fruit waste, by using ethanolic-based solutions of 1-butyl-3-methylimidazolium tetrafluoroborate ($[C_4mim][BF_4]$). The easy reproducibility of the process was proved for 10 extraction cycles in an environmentally efficient way (very low carbon footprint). In this work, the IL ethanolic solution rich in carotenoids was frozen at −80 °C, causing the IL precipitation (94%). Later, the same authors have developed a new integrated process using aqueous solutions of an ammonium-based IL, the 1-decyltetramethylammonium bromide, $[N_{1,1,1,10}]$Br. Again, the polishing step was carried, however, and due to the nature of carotenoids and the IL solution, water was applied as antisolvent in a very efficient way (allowing to recycle around 96% of $[N_{1,1,1,10}]$Br), which was recycled up to three cycles, allowing to obtain pure carotenoids (99.9%) in the end (de Souza Mesquita, Martins, et al., 2020).

Enclosed in the techniques developed to perform the polishing of natural pigments is the use of a molecularly imprinted polymer successfully applied as a special sorbent for the solid–liquid extraction of astaxanthin from shrimp waste using an ultrasound-assisted approach (Bi et al., 2010). The IL ethanolic media (composed by 1-propylamine-3-methylimidazolium bromide—$[C_3NH_2mim]$Br—at 500 mM) was able to recover 92.7 μg/g of astaxanthin. However, despite the excellent affinity of the method proposed, the elution solvent used as a mobile phase to recover astaxanthin was dichloromethane, a hepatotoxic and volatile solvent (Wypych, 2019), which consequently restricts the application of the pigment in some commercial sectors, especially into the food segment. In a similar approach, in a SPE strategy, the adsorbent polymer resin Amberlite XAD-7HP with ethanol have been used as a mobile phase and achieved 60% of IL recovery ($[C_4mim]$Cl) with 55% of purification improvement of the total carotenoids extracted, mainly xanthophylls, from orange peels (Murador, Braga, et al., 2019).

For curcuminoids, Sahne et al. (2017) reported an interesting polishing technique mediated by distillation, which allowed one cycle of recovery and reuse of a newly synthesized IL (*N*,*N*-dipropyl ammonium-*N*′,*N*′-dipropylcarbamate). Besides, the purification of curcuminoids using a chromatographic column was also reported (purity of 96%, by applying silica gel [$(SiO_2)_x$]). Using this same strategy, however, with an eutectic solvent, Liu, Li, et al. (2019) have demonstrated a SPE mediated with a Waters® HLB Oasis column (Waters Corporation, Milford, Massachusetts, USA) for the recovery of both eutectic solvents and curcuminoids. Around 90% of the extracted pigments with high purity were achieved. However, methanol was used as the mobile phase for pigment elution, thus restricting the use of the final pigmented product in the food sector. Adsorption resins are also widely used to recover some pigments, namely, flavonoids and anthocyanins, particularly from the eutectic mixtures. Actually, there are some examples reported on the use of resins, which include the microporous AB-8, X-5, HP-20, HPD-750, LX-5, and LX-38. In general, good results have been obtained on the use of resins to recover and reuse the eutectic mixtures applied as solvents. In these works, the recoveries of eutectic mixtures reported were around 70%, the lowest value reported (Zhou, Wang, et al., 2018), 95% for the resin Sepabeads 825L using deionized water as mobile phase (Panić et al. (2019), and 99% for the microporous resin LX-38 (Zhuang, Dou, Li, & Liu, 2017).

4 | GFP AND R-PE: TWO INTERESTING CASE STUDIES

As highlighted in the works here reviewed, the economic viability of the downstream processes to obtain natural colorants depends very much on several factors, which include the advantages of the competing synthetic substitutes (e.g., color stability, chemical stability, and photostability), the availability and abundance of the natural raw materials (seasonality, variability between batches, just to mention a few), and the economic viability of the processes to obtain the bio-based colorants. It is well recognized that, independently of the field of application, all processes able to generate new products/compounds need to overcome some drawbacks and return some outputs to be considered commercially viable. Included in the commercial viability, in our opinion, it should be the efficiency, performance, economic and environmental impacts of the process to reach the product of interest, and the target characteristics of the final product (purity level, concentration, or content). But, like it happens when trying to reach all the 12 principles of Green Chemistry in a specific process/solvent/product (which up to now was still not done!), to develop a commercially viable but sustainable process to recover a natural-based colorant from a specific raw material is also very difficult. The difficulty lies on the impossibility of creating heuristic rules that allow the selection of the appropriate solvents, techniques, or even process conditions, without any experimental evidence. The conditions of the sample, the morphology of the cells and, consequently, the mechanisms behind the cells' disruption, and the properties of the colorant are some of the conditions that may be significantly different between distinct residues (e.g., peels of fruits), raw materials (e.g., macroalgae), or biomasses (e.g., microorganisms), which justifies the lack of heuristic rules on the field.

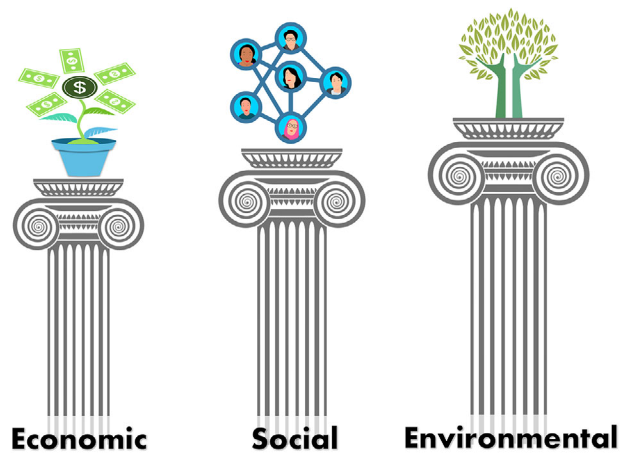

FIGURE 7 Three main pillars describing the concept of sustainability

If it is true that the sustainability concept is recurrently mentioned by authors, conferencists, and governments, when it is applied to industrial processes and services, the lack of a clear and detailed definition is still a major drawback of communication between regulatory entities, industry, academia, and governments. The three fundamental pillars behind the concept of sustainability are described as being the environmental, economic, and social sustainability (Figure 7), which is quite clear in the literature (Mensah & Casadevall, 2019) and on the Sustainable Development Goals (SDG) implemented by the United Nations (https://sdgs.un.org/goals). However, the recurrent application of this concept needs further clarification in what concerns its meaning in each sector of activity. In the case of products and process development, the United Nations define it as "Inclusive and sustainable industrial development has been incorporated, together with resilient infrastructure and innovation" (SDG 9—Industry, Innovation, and Infrastructure). Still, its meaning needs clarification in what regards the main issues it includes. Having this in mind and all works analyzed in this review, we came out with a summary of

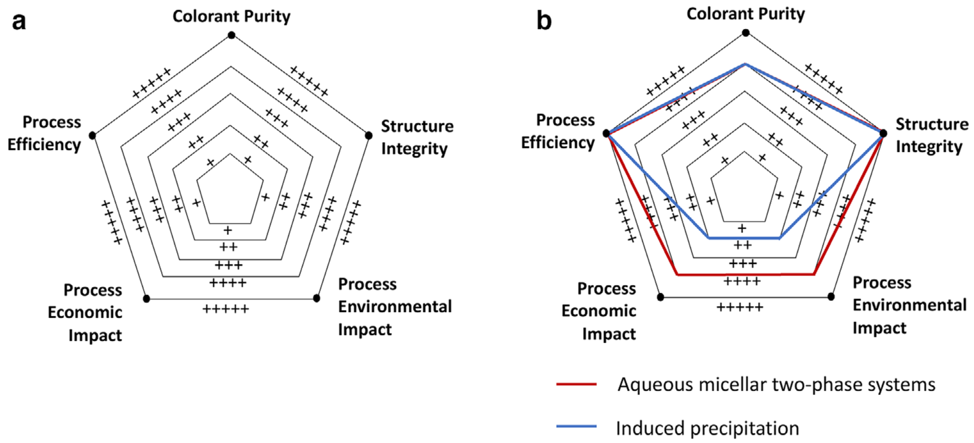

FIGURE 8 (a) Diagram representing the five main issues to manipulate aiming at the development of a sustainable (downstream) process; (b) diagram prepared for the comparison between both processes developed to recover R-PE, namely, the red line represents the process developed using aqueous micellar two-phase systems (Vicente et al., 2019) and the blue line represents the process using induced precipitation (Martins et al., 2020)

what are, in our opinion, the main issues that need to be considered as the ones composing the ideal scenario for the design and optimization of a sustainable downstream process to obtain a naturally based colorant (the focus of the present work). As it is easily identified in Figure 8a, the five main issues that need to be manipulated to achieve a sustainable process are the efficiency, economic and environmental impact of the process, and the colorant purity and structural integrity. Three are related with the process, and the other two represent the target compound/class of compounds to recover. It is obvious that several parameters are included in each issue, and thus the different criteria should be considered when the process is being designed and optimized. Actually, the final process to be sustainable, it should be the one demonstrating an equilibrium between the five issues presented and the final application/sector of activity. The typical example used is “the process complexity should be defined considering the demands of the final application.” This is true and it is somehow dictating a heuristic rule: low-income applications mean low purities and simpler processes; high-income applications mean high purities and, consequently, more complex processes. From the literature analyzed in this work, it is evident that most of the works are focusing mainly on the process efficiency, the colorant purity, and structural integrity. These are issues with high relevance because they include normally the selection of the best solvent (from the list of solvents screened) to recover the colorant with the high purity possible, and able to maintain the chemical structure of the colorant. However, even when these works are analyzing these three issues, in our opinion they are missing some steps. The final purity should be entirely related with the application, the best solvent should be the one with a high capacity to recover the colorant, but it does not need to be the one with the highest capacity, for example, if this implies to design a much more complex process. It may be the most indicated from efficiency but not the best option when analysis the entire process, because again, a more complex process means higher costs, which normally is also a sign of a higher environmental impact. Another very important question is related to the structural integrity of the colorant because this also implies the coordination with the application. As an example, if a fluorescent protein like R-PE is extracted from red algae to be used as an optically active center to be incorporated in luminescent solar concentrators, it should be guaranteed its chemical but also its photostability, both essential parameters for its final application. To improve our understanding of how all of these issues are interconnected, from the works discussed in previous sections, two case studies are highlighted, namely, R-PE and GFP.

In the case of GFP, systems using different types of ABS were tested and the best system selected considering its efficiency (dos Santos et al., 2018). Later, an economic analysis was also performed by comparing the economic impact of applying the different types of ABS (Torres-Acosta et al., 2020). A set of 14 ABS was studied to discriminate through production costs. In this work, the product purity parameter was also incorporated in the calculations. Two optimum ABSs were then selected and their costs were investigated at two different scales (1 and 100 L) to clarify the economic viability of using ABS at large scales to purify GFP. In the end, a sensitivity analysis was also performed to infer the impact of the recovery yield, material costs discount, and production titer. With this final analysis, the authors were able to conclude on the circumstances under which the IL-based system (less cost-effective) can overcome the production costs of the traditional

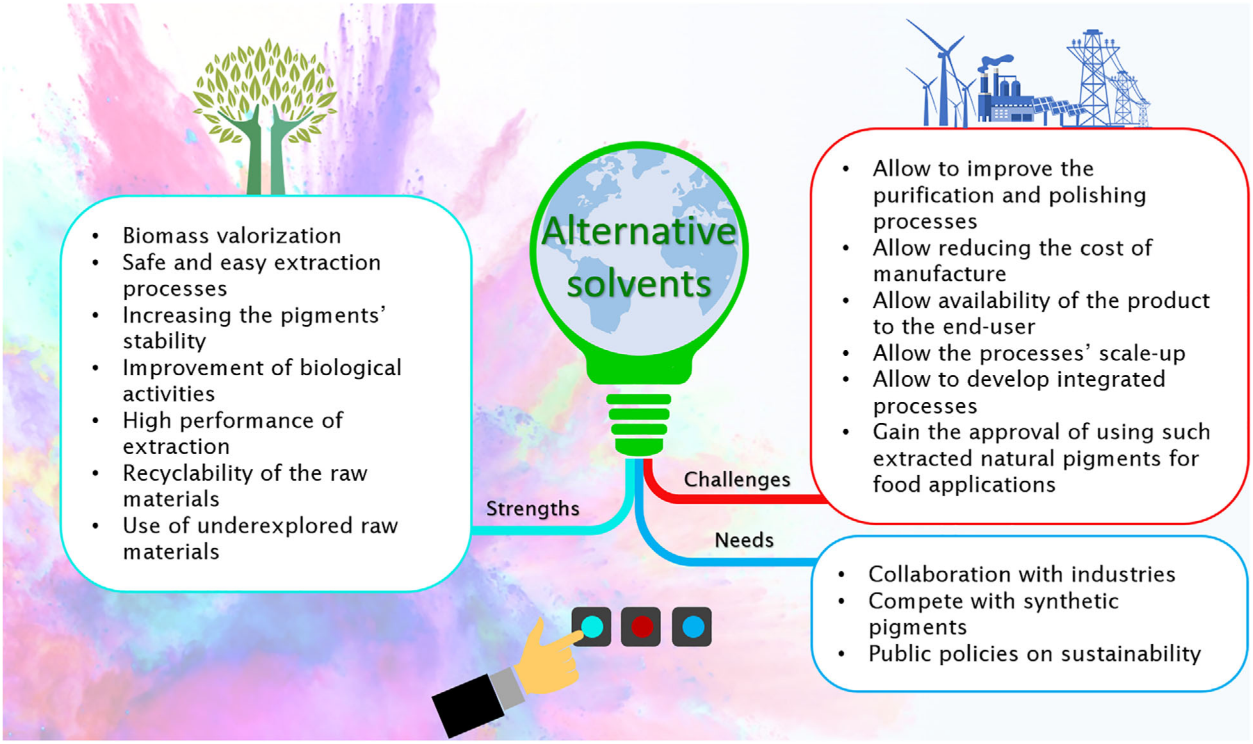

FIGURE 9 Representation of the current status, future challenges, and needs on the use of alternative solvents to recover bio-based pigments

PEG-based ABS (more cost-effective). The results indicated that, at high production titers, the IL-based ABS starts to be less expensive than the traditional polymeric system at all analyzed scales, which again demonstrates the need to study the different issues of the process under an interconnected strategy including solvent, the overall process, scale, and final product. However, in the work carried with GFP, the interconnection between the process and economic analysis and the final application demands was still missing. The GFP was applied as an optically active center in the development of luminescent solar concentrators (Carlos et al., 2020). Although the satisfactory results obtained, one of the conclusions was that the purity level applied to the luminescent solar concentrators was superior to the value required by the final application. In this sense, a new strategy was adopted by the authors for the recovery of R-PE from *Gracilaria gracilis*. In this case, two downstream processes were developed and optimized considering the separation of phycobiliproteins from the nonfluorescent proteins and also the R-PE from the other phycobiliproteins, to be able to answer the demands of application only for R-PE, but also applications interested in the phycobiliproteins as the final product. This objective was first achieved by applying AMTPS (Vicente et al., 2019) (as previously discussed in this review), but after the careful analysis of using R-PE in a more pure state also in luminescent solar concentrators (Frias et al., 2019), we came out with the conclusion that a simpler and more environmentally friendly process was much more useful than the one first developed (complex, with several units of operation and with a significant environmental impact; Vicente et al., 2019). After, and considering the low purity demands and chemical (Bharmoria et al., 2020) and optical stability required to R-PE as an optically active center, a new process was envisioned by induced precipitation, for which the environmental and economic impacts, even at larger scales of operation, are much more advantageous (Martins et al., 2020). Although this last work is still under preparation, Figure 8b shows the comparison between both processes developed to recover R-PE, in which it is concluded that, even if the structural integrity and purity of the target compound as well as the process efficiency are similar, the environmental and economic impacts may dictate the selection (Martins et al., 2020).

5 | CONCLUDING REMARKS AND FUTURE PERSPECTIVES

The extraction of natural pigments involves complex mechanisms that can be accomplished by various techniques and solvents. Green extraction concepts and principles are identified as potential allies to face the challenges imposed in the 21st century, and excellent candidates for meeting the sustainability goals in the industrial sector (Chemat et al., 2019, 2020). Based on the results reviewed on this work, it seems that ILs, eutectic solvents, and more recently, edible oils and surfactants may be good candidates to replace, or at least, reduce the use of conventional organic solvents, in the extraction of natural pigments from different biomass matrices. The data available highlight the advantages of the alternative over the conventional organic solvents, improving the physicochemical quality of the extracted pigments, ensuring better extraction yields, the thermal stability of the target compounds, and in some cases, increasing the safety of the process, by decreasing the environmental impact caused by the solvents. Besides, some works have been postulating that ILs and eutectic solvents can be recycled and reused in new cycles, thus reducing the negative environmental impact and the inherent cost of the process. However, most works are based on the concepts of "green solvents" and "safer," but do not effectively prove if the developed process justifies the use of such concepts. Therefore, the sustainability of the developed processes remains a challenge. This reflects the future application of the extracted pigments, because for many purposes, for example, the food industry, there is a need for purer extracts and products, that is, free of solvent. Given that, and in our opinion, future works should focus on the development of new purification strategies to ensure their scale-up and effective commercial viability, always considering the real environmental impact of the process. In most recent years, it has been an attempt to contradict this tendency. Also, as reported by Rodriguez-Amaya (2016, 2019), the instability of the natural pigments is a major problem, because the natural color of the pigment is lost over time, which reflects on the prevalence in the use of synthetic dyes in foods, especially in candies, cheeses, and soft drinks. However, some European countries, namely, Austria, Norway, Finland, the United Kingdom, and France, have forbidden the use of some synthetic dyes in foods, and the other countries have some restrictions to their use, which puts the EU as an expressive niche of the market of natural dyes, especially those with high color stability. Also, the EU is very receptive to products of natural origin, especially those obtained from wastes, which meets the European economic plan based on the Circular Economy for a cleaner and more competitive Europe (European Commission, 2020). In contrast, in some emergent countries like Brazil, a country with high social heterogenicity, there is a high increase in the rates of deforestation, use of agricultural pesticides, and food waste, which represents a global problem today, considering the recognized credentials of a country with huge biodiversity and as one of the largest exporters of food in the world (Braga, de Rosso, Harayashiki, Jimenez, & Castro, 2020). Thus, the sustainable strategies to recover natural colorants and other bioactive compounds from foods or to foods are one of the main challenges so far in the sector.

Researchers are indeed trying to find more sustainable ways to extract and purify diverse biomolecules, including natural pigments. It is recognized the urgency to find new sources and new colorant structures. Moreover, it is well known the issues associated with safety, stability, chromaticity, and/or opacity of common pigments. This quest is even more urgent for red pigments, as recently argued by Zach Schonbrun (Schonbrun, 2018). Citing Dr. Schonbrun, "a new pigment can generate hundreds of millions of dollars annually, affecting positively product categories from plastics to cosmetics, cars, construction," food, and nutraceutical. The truth is that the synthetic protocols used to prepare the most common pigments are unsafe (Oplatowska-Stachowiak & Elliot, 2017). Although the safety issues may be surpassed using naturally based pigments, their low abundance and low structural variability are inhibiting their more frequent use. That said, and in our opinion, one of the strategies to improve this sector passes, not only by the search for new sources of pigments, for example, focusing cyanobacteria and corals, but also looking for the exploration of some agroforestry raw materials and residues (as done with the Amazonian fruit waste). Moreover, the development of new metabolic routes of recombinant organisms allowing the production of added-value pigments in higher abundance may be one of the lines of deeper investigation. Actually, and despite the difficult task that is to design the metabolic routes to improve the production of a certain compound, it may make sense for some particular pigments. As a successful example of this strategy is the Portuguese company SilicoLife, Lda (http://www.silicolife.com/). The company's core business is the "design of optimized microorganisms and novel pathways for industrial biotechnology applications, based on computational metabolic engineering and synthetic biology approaches." Moreover, SilicoLife is also pursuing the "design of novel routes for the biological production of selected high-value chemicals with large market potential" (internal R&D program recently launched), which is in our opinion a very good initiative that helps researchers to achieve success on the search for added-value colorants.

If it is true that some of the works previously discussed show good strategies to obtain pigments from biomass, the processes' economic viability is questionable mainly due to the high complexity of the downstream process to recover a very low amount of pigments. Furthermore, complex designs make the processes scale-up a very difficult and uneconomical task. To surpass this limitation, there are some strategies and technologies that, in our opinion, should be deeply explored. One of these strategies will pass through the application of microfluidics with ABS (Hardt & Hahn, 2012). Some work was already done on the purification of GFP and bacteriorhodopsin (another fluorescent protein with natural origin), but conjugating microfluidics with traditional polymer-based ABS (Huh et al., 2010; Meagher, Light, & Singh, 2008). More recently, it was reviewed the separation and purification of biomacromolecules based on microfluidics (Vicente, Plazl, Ventura, & Žnidaršič-Plazl, 2020), and concluded that this innovative field may improve the purification processes with more efficient and selective purifications, mainly when the abundance of the target compounds is limited, thus allowing them to integrate miniaturization with an intensification of the downstream processes.

Thus, and as exposed in Figure 9, despite the large amount of experimental work reporting the extraction of natural pigments using alternative solvents, mainly ILs and eutectic solvents, there are just some advances on the effective assessment of the integration of processes, especially in what concerns the recycling of solvents and the life cycle analysis of the whole processes, which will have a big influence on the economic and environmental impact evaluation and, consequently, on the sustainability of the colorant application. Despite the efforts and data obtained so far, we encourage that the future works focus on the search for more ecological extraction processes, for example, the use of integrative and automatic systems (extraction–purification–polishing–solvents' recycling combined), such as those based on pressurized-liquid extraction (PLE), which consists in a modern method based on the use of high pressure (usually up to 600 MPa) at a specific temperature (which can also be around room temperature), and short times, able to reach higher extraction yields, is usually coupled with spectroscopy techniques for the identification of the compounds (da Silva et al., 2020). Thus, this method, combined with alternative solvents, could enhance the extractability and selectivity of bio-based compounds, while maintaining (or improving) the chemical stability and main properties of the pigments. Also, PLE-mediated extractions are known as good alternatives to preserve the organoleptic characteristics of the extracted compounds by the inactivation of enzymes and microorganisms, while extending their shelf life and impairing the formation of toxic compounds (Szczepańska, Barba, Skąpska, & Marszałek, 2020), which is very interesting in food-related processes. Additionally, and as recently reviewed by Chemat et al. (2020), the main sustainable and high-performance methods used to obtain food bio-based products should consider the application of innovative and integrated systems, able to recover natural compounds continuously and cost-effectively. Besides, sustainability as a real strategy will only be possible if academia and industries collaborate by integrating efforts on the development of balanced strategies. These balanced strategies need to integrate efficient processes to produce and obtain high-value products (meaning lucrative and high performant) to meet the society demands. In the end, claiming the need for competitive production of natural colorants, public policies focusing on industrial sustainability must be discussed.

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AUTHOR CONTRIBUTIONS

L. M. de Souza Mesquita and M. Martins conducted the literature search, conceptualized and executed the review, and drafted the first version. S. P.M. Ventura, L. P. Pisani, and V. V. de Rosso guided the development of the work. All the authors wrote the manuscript, approved the final version of the work, and takes responsibility for all aspects of the reliability, and interpretation of the data presented.

CONFLICTS OF INTEREST

The authors declare no conflicts of interest.

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REFERENCES

Abbott, A. P., Boothby, D., Capper, G., Davies, D. L., & Rasheed, R. K. (2004). Deep eutectic solvents formed between choline chloride and carboxylic acids: Versatile alternatives to ionic liquids. *Journal* *of the American Chemical Society*, *126*(29), 9142–9147. https://doi.org/10.1021/ja048266j

Amchova, P., Kotolova, H., & Ruda-Kucerova, J. (2015). Health safety issues of synthetic food colorants. *Regulatory Toxicology and Pharmacology*, *73*(3), 914–922. https://doi.org/10.1016/j.yrtph.2015.09.026

Anastas, N., & Miller, G. W. (2018). A farewell to harms: The audacity to design safer products. *Toxicological Sciences*, *161*(2), 211–213. https://doi.org/10.1093/toxsci/kfx288

Anastas, P., & Eghbali, N. (2010). Green chemistry: Principles and practice. *Chemical Society Reviews*, *39*(1), 301–312. https://doi.org/10.1039/B918763B

Aranda, S., Montes-Borrego, M., & Landa, B. B. (2011). Purple-pigmented violacein-producing *Duganella* spp. inhabit the rhizosphere of wild and cultivated olives in southern Spain. *Microbial Ecology*, *62*(2), 446–459. https://doi.org/10.1007/s00248-011-9840-9

Aroso, I. M., Silva, J. C., Mano, F., Ferreira, A. S., Dionísio, M., Sá-Nogueira, I., … Duarte, A. R. C. (2016). Dissolution enhancement of active pharmaceutical ingredients by therapeutic deep eutectic systems. *European Journal of Pharmaceutics and Biopharmaceutics*, *98*, 57–66. https://doi.org/10.1016/j.ejpb.2015.11.002

Bajkacz, S., & Adamek, J. (2018). Development of a method based on natural deep eutectic solvents for extraction of flavonoids from food samples. *Food Analytical Methods*, *11*(5), 1330–1344. https://doi.org/10.1007/s12161-017-1118-5

Baria, B., Upadhyay, N., Singh, A. K., & Malhotra, R. K. (2019). Optimization of 'green' extraction of carotenoids from mango pulp using split plot design and its characterization. *LWT*, *104*, 186–194. https://doi.org/10.1016/j.lwt.2019.01.044

Benavides, J., Rito-Palomares, M., & Asenjo, J. (2011). *Comprehensive biotechnology* (2nd ed.). Monterrey, Mexico: Elsevier.

Bi, W., Tian, M., Zhou, J., & Row, K. H. (2010). Task-specific ionic liquid-assisted extraction and separation of astaxanthin from shrimp waste. *Journal of Chromatography B*, *878*(24), 2243–2248. https://doi.org/10.1016/j.jchromb.2010.06.034

Biazotto, K. R., de Souza Mesquita, L. M., Neves, B. V., Braga, A. R. C., Tangerina, M. M. P., Vilegas, W., … De Rosso, V. V. (2019). Brazilian biodiversity fruits: Discovering bioactive compounds from underexplored sources. *Journal of Agricultural and Food Chemistry*, *67*(7), 1860–1876. https://doi.org/10.1021/acs.jafc.8b05815

Bharmoria, P., Correia, S. F., Martins, M., Hernández-Rodríguez, M. A., Ventura, S. P., Ferreira, R. A., … Coutinho, J. A. P. (2020). Protein cohabitation: improving the photochemical stability of R-phycoerythrin in the solid state. *The Journal of Physical Chemistry Letters*, *11*(15), 6249–6255. https://doi.org/10.1021/acs.jpclett.0c01491

Bogacz-Radomska, L., & Harasym, J. (2018). β-Carotene—Properties and production methods. *Food Quality and Safety*, *2*(2), 69–74. https://doi.org/10.1093/fqsafe/fyy004

Bosiljkov, T., Dujmić, F., Bubalo, M. C., Hribar, J., Vidrih, R., Brnčić, M., … Jokić, S. (2017). Natural deep eutectic solvents and ultrasound-assisted extraction: Green approaches for extraction of wine lees anthocyanins. *Food and Bioproducts Processing*, *102*, 195–203. https://doi.org/10.1016/j.fbp.2016.12.005

Braga, A. R. C., de Rosso, V. V., Harayashiki, C. A. Y., Jimenez, P. C., & Castro, Í. B. (2020). Global health risks from pesticide use in Brazil. *Nature Food*, *1*(6), 312–314. https://doi.org/10.1038/s43016-020-0100-3

Carlos, C. P., Correia, S. F., Martins, M., Savchuk, O. A., Coutinho, J. A. P., André, P. S., … Ferreira, R. A. (2020). Environmentally friendly luminescent solar concentrators based on an optically efficient and stable green fluorescent protein. *Green Chemistry*, *22*(15), 4943–4951. https://doi.org/10.1039/D0GC01742F

Castañeda-Ovando, A., de Lourdes Pacheco-Hernández, M., Páez-Hernández, M. E., Rodríguez, J. A., & Galán-Vidal, C. A. (2009). Chemical studies of anthocyanins: A review. *Food Chemistry*, *113*(4), 859–871. https://doi.org/10.1016/j.foodchem.2008.09.001

Cazón, P., Velazquez, G., Ramírez, J. A., & Vázquez, M. (2017). Polysaccharide-based films and coatings for food packaging: A review. *Food Hydrocolloids*, *68*, 136–148. https://doi.org/10.1016/j.foodhyd.2016.09.009

Cervantes-Paz, B., Yahia, E. M., de Jesús Ornelas-Paz, J., Victoria-Campos, C. I., Ibarra-Junquera, V., Pérez-Martínez, J. D., & Escalante-Minakata, P. (2014). Antioxidant activity and content of chlorophylls and carotenoids in raw and heat-processed Jalapeño peppers at intermediate stages of ripening. *Food Chemistry*, *146*, 188–196. https://doi.org/10.1016/j.foodchem.2013.09.060

Chemat, F., Abert-Vian, M., Fabiano-Tixier, A. S., Strube, J., Uhlenbrock, L., Gunjevic, V., & Cravotto, G. (2019). Green extraction of natural products. Origins, current status, and future challenges. *TrAC Trends in Analytical Chemistry*, *118*, 248–263. https://doi.org/10.1016/j.foodhyd.2016.09.009

Chemat, F., Vian, M. A., Fabiano-Tixier, A. S., Nutrizio, M., Jambrak, A. R., Munekata, P. E., … Cravotto, G. (2020). A review of sustainable and intensified techniques for extraction of food and natural products. *Green Chemistry*, *22*(8), 2325–2353. https://doi.org/10.1039/C9GC03878G

Choi, S. A., Oh, Y. K., Lee, J., Sim, S. J., Hong, M. E., Park, J. Y., … Lee, J. S. (2019). High-efficiency cell disruption and astaxanthin recovery from *Haematococcus pluvialis* cyst cells using room-temperature imidazolium-based ionic liquid/water mixtures. *Bioresource Technology*, *274*, 120–126. https://doi.org/10.1016/j.biortech.2018.11.082

Clarke, C. J., Tu, W. C., Levers, O., Brohl, A., & Hallett, J. P. (2018). Green and sustainable solvents in chemical processes. *Chemical Reviews*, *118*(2), 747–800. https://doi.org/10.1021/acs.chemrev.7b00571

Clark, J. H. (2019). Green biorefinery technologies based on waste biomass. *Green Chemistry*, *21*(6), 1168–1170. https://doi.org/10.1039/C9GC90021G

Clark, J. H., Farmer, T. J., Herrero-Davila, L., & Sherwood, J. (2016). Circular economy design considerations for research and process development in the chemical sciences. *Green Chemistry*, *18*(14), 3914–3934. https://doi.org/10.1039/C6GC00501B

Cláudio, A. F. M., Ferreira, A. M., Freire, M. G., & Coutinho, J. A. P. (2013). Enhanced extraction of caffeine from guarana seeds using aqueous solutions of ionic liquids. *Green Chemistry*, *15*(7), 2002–2010. https://doi.org/10.1039/C3GC40437D

Cuellar-Bermudez, S. P., Aguilar-Hernandez, I., Cardenas-Chavez, D. L., Ornelas-Soto, N., Romero-Ogawa, M. A., & Parra-Saldivar, R. (2015). Extraction and purification of high-value metabolites from microalgae: Essential lipids, astaxanthin and phycobiliproteins. *Microbial Biotechnology*, *8*(2), 190–209. https://doi.org/10.1111/1751-7915.12167

Cui, Q., Liu, J. Z., Wang, L. T., Kang, Y. F., Meng, Y., Jiao, J., & Fu, Y. J. (2018). Sustainable deep eutectic solvents preparation and their efficiency in extraction and enrichment of main bioactive flavonoids from sea buckthorn leaves. *Journal of Cleaner Production*, *184*, 826–835. https://doi.org/10.1016/j.jclepro.2018.02.295

Ćurko, N., Tomašević, M., Cvjetko Bubalo, M., Gracin, L., Radojčić Redovniković, I., & Kovačević Ganić, K. (2017). Extraction of proanthocyanidins and anthocyanins from grape skin by using ionic liquids. *Food Technology and Biotechnology*, *55*(3), 429–437. https://doi.org/10.17113/ftb.55.03.17.5200

Dai, Y., Rozema, E., Verpoorte, R., & Choi, Y. H. (2016). Application of natural deep eutectic solvents to the extraction of anthocyanins from *Catharanthus roseus* with high extractability and stability replacing conventional organic solvents. *Journal of Chromatography A*, *1434*, 50–56. https://doi.org/10.1016/j.chroma.2016.01.037

Dangles, O., & Fenger, J. A. (2018). The chemical reactivity of anthocyanins and its consequences in food science and nutrition. *Molecules*, *23*(8), 1970. https://doi.org/10.3390/molecules23081970

D'Archivio, A. A., Maggi, M. A., & Ruggieri, F. (2018). Extraction of curcuminoids by using ethyl lactate and its optimisation by response surface methodology. *Journal of Pharmaceutical and Biomedical Analysis*, *149*, 89–95. https://doi.org/10.1016/j.jpba.2017.10.042

da Silva, L. C., Souza, M. C., Sumere, B. R., Silva, L. G., da Cunha, D. T., Barbero, G. F., … Rostagno, M. A. (2020). Simultaneous extraction and separation of bioactive compounds from apple pomace using pressurized liquids coupled on-line with solid-phase extraction. *Food Chemistry*, *318*, 126450. https://doi.org/10.1016/j.foodchem.2020.126450

de Souza Mesquita, L. M. S., Neves, B. V., Pisani, L. P., & de Rosso, V. V. (2020). Mayonnaise as a model food for improving the bioaccessibility of carotenoids from *Bactris gasipaes* fruits. *LWT*, *122*, 109022. https://doi.org/10.1016/j.lwt.2020.109022

de Souza Mesquita, L. M., Martins, M., Maricato, É., Nunes, C., Quinteiro, P. S., Dias, A. C., … Ventura, S. P. (2020). Ionic liquid-mediated recovery of carotenoids from the *Bactris gasipaes* fruit waste and their application in food-packaging chitosan films. *ACS Sustainable Chemistry & Engineering*, *8*(10), 4085–4095. https://doi.org/10.1021/acssuschemeng.9b06424

de Souza Mesquita, L. M., Murador, D. C., & de Rosso, V. V. (2019). Application of ionic liquid solvents in the food industry. In S. Zhang (Ed.), *Encyclopedia of ionic liquids*. Singapore: Springer. Retrieved from https://doi-org-443.webvpn.fjmu.edu.cn/10.1007/978-981-10-6739-6_8-1

de Souza Mesquita, L. M., Ventura, S. P., Braga, A. R., Pisani, L. P., Dias, A. C., & de Rosso, V. V. (2019). Ionic liquid-high performance extractive approach to recover carotenoids from *Bactris gasipaes* fruits. *Green Chemistry*, *21*(9), 2380–2391. https://doi.org/10.1039/C8GC03283A

Delgado-Vargas, F., Jiménez, A. R., & Paredes-López, O. (2000). Natural pigments: Carotenoids, anthocyanins, and betalains — Characteristics, biosynthesis, processing, and stability. *Critical Reviews in Food Science and Nutrition*, *40*(3), 173–289. https://doi.org/10.1080/10408690091189257

Desai, R. K., Streefland, M., Wijffels, R. H., & Eppink, M. H. (2016). Novel astaxanthin extraction from *Haematococcus pluvialis* using cell permeabilising ionic liquids. *Green Chemistry*, *18*(5), 1261–1267. https://doi.org/10.1039/C5GC01301A

dos Santos, N. V., Martins, M., Santos-Ebinuma, V. C., Ventura, S. P., Coutinho, J. A. P., Valentini, S. R., & Pereira, J. F. (2018). Aqueous biphasic systems composed of cholinium chloride and polymers as effective platforms for the purification of recombinant green fluorescent protein. *ACS Sustainable Chemistry & Engineering*, *6*(7), 9383–9393. https://doi.org/10.1021/acssuschemeng.8b01730

Duan, M. H., Luo, M., Zhao, C. J., Wang, W., Zu, Y. G., Zhang, D. Y., … Fu, Y. J. (2013). Ionic liquid-based negative pressure cavitation-assisted extraction of three main flavonoids from the pigeon pea roots and its pilot-scale application. *Separation and Purification Technology*, *107*, 26–36. https://doi.org/10.1016/j.seppur.2013.01.003

Dumay, J., & Morançais, M. (2016). Proteins and pigments. In J. Fleurence & I. Levine (Eds.), *Seaweed in health and disease prevention* (pp. 275–318). Cambridge, MA: Academic Press.

Durán, M., Ponezi, A. N., Faljoni-Alario, A., Teixeira, M. F., Justo, G. Z., & Durán, N. (2012). Potential applications of violacein: A microbial pigment. *Medicinal Chemistry Research*, *21*(7), 1524–1532. https://doi.org/10.1007/s00044-011-9654-9

e Silva, F. A., Carmo, R. M., Fernandes, A. P., Kholany, M., Coutinho, J. A. P., & Ventura, S. P. (2017). Using ionic liquids to tune the performance of aqueous biphasic systems based on pluronic L-35 for the purification of naringin and rutin. *ACS Sustainable Chemistry & Engineering*, *5*(8), 6409–6419. https://doi.org/10.1021/acssuschemeng.7b00178

Erythropel, H. C., Zimmerman, J. B., de Winter, T. M., Petitjean, L., Melnikov, F., Lam, C. H., … Pincus, L. N. (2018). The Green ChemisTREE: 20 years after taking root with the 12 principles. *Green Chemistry*, *20*(9), 1929–1961. https://doi.org/10.1039/C8GC00482J

European Comission. (2020). *Communication from the commission to the European parliament, the council, the European economic and social committee and the committee of the regions—A new circular economy action plan for a cleaner and more competitive Europe*. Brussels, Belgium: Author.

Freire, M. G., Claudio, A. F. M., Araujo, J. M., Coutinho, J. A. P., Marrucho, I. M., Lopes, J. N. C., & Rebelo, L. P. N. (2012). Aqueous biphasic systems: A boost brought about by using ionic liquids. *Chemical Society Reviews*, *41*(14), 4966–4995. https://doi.org/10.1039/C2CS35151J

Frias, A. R., Correia, S. F., Martins, M., Ventura, S. P., Pecoraro, E., Ribeiro, S. J., … Carlos, L. D. (2019). Sustainable liquid luminescent solar concentrators. *Advanced Sustainable Systems*, *3*(3), 1800134. https://doi.org/10.1002/adsu.201800134

Gebhardt, B., Sperl, R., Carle, R., & Müller-Maatsch, J. (2020). Assessing the sustainability of natural and artificial food colorants. *Journal of Cleaner Production*, *260*, 120884. https://doi.org/10.1016/j.jclepro.2020.120884

Gerken, H. G., Donohoe, B., & Knoshaug, E. P. (2013). Enzymatic cell wall degradation of *Chlorella vulgaris* and other microalgae for biofuels production. *Planta*, *237*(1), 239–253. https://doi.org/10.1007/s00425-012-1765-0

Geyer, R., Jambeck, J. R., & Law, K. L. (2017). Production, use, and fate of all plastics ever made. *Science Advances*, *3*(7), e1700782. https://doi.org/10.1126/sciadv.1700782

Goula, A. M., Ververi, M., Adamopoulou, A., & Kaderides, K. (2017). Green ultrasound-assisted extraction of carotenoids from pomegranate wastes using vegetable oils. *Ultrasonics Sonochemistry*, *34*, 821–830. https://doi.org/10.1016/j.ultsonch.2016.07.022

Guillard, V., Gaucel, S., Fornaciari, C., Angellier-Coussy, H., Buche, P., & Gontard, N. (2018). The next generation of sustainable food packaging to preserve our environment in a circular economy context. *Frontiers in Nutrition*, *5*, 121. https://doi.org/10.3389/fnut.2018.00121

Guo, N., Jiang, Y. W., Wang, L. T., Niu, L. J., Liu, Z. M., & Fu, Y. J. (2019). Natural deep eutectic solvents couple with integrative extraction technique as an effective approach for mulberry anthocyanin extraction. *Food Chemistry*, *296*, 78–85. https://doi.org/10.1016/j.foodchem.2019.05.196

Gutowski, K. E., Broker, G. A., Willauer, H. D., Huddleston, J. G., Swatloski, R. P., Holbrey, J. D., & Rogers, R. D. (2003). Controlling the aqueous miscibility of ionic liquids: Aqueous biphasic systems of water-miscible ionic liquids and water-structuring salts for recycle, metathesis, and separations. *Journal of the American Chemical Society*, *125*(22), 6632–6633. https://doi.org/10.1021/ja0351802

Hardt, S., & Hahn, T. (2012). Microfluidics with aqueous two-phase systems. *Lab on a Chip*, *12*(3), 434–442. https://doi.org/10.1039/C1LC20569B

Hasan, Z., & Jhung, S. H. (2015). Removal of hazardous organics from water using metal-organic frameworks (MOFs): Plausible mechanisms for selective adsorptions. *Journal of Hazardous Materials*, *283*, 329–339. https://doi.org/10.1016/j.jhazmat.2014.09.046

Hatti-Kaul, R. (2000). *Aqueous two-phase systems: Methods and protocols* (Vol. 11). Berlin, Germany: Springer Science & Business Media.

Toledo Hijo, A. A., Maximo, G. J., Costa, M. C., Batista, E. A., & Meirelles, A. J. (2016). Applications of ionic liquids in the food and bioproducts industries. *ACS Sustainable Chemistry & Engineering*, *4*(10), 5347–5369. https://doi.org/10.1021/acssuschemeng.6b00560

Huh, Y. S., Jeong, C.-M., Chang, H. N., Lee, S. Y., Hong, W. H., & Park, T. J. (2010). Rapid separation of bacteriorhodopsin using a laminar-flow extraction system in a microfluidic device. *Biomicrofluidics*, *4*(1), 014103. https://doi.org/10.1063/1.3298608

Jablonský, M., Škulcová, A., Malvis, A., & Šima, J. (2018). Extraction of value-added components from food industry based and agro-forest biowastes by deep eutectic solvents. *Journal of Biotechnology*, *282*, 46–66. https://doi.org/10.1016/j.jbiotec.2018.06.349

Janiszewska-Turak, E., Pisarska, A., & Królczyk, J. B. (2016). Natural food pigments application in food products. *Nauka Przyroda Technologie*, *10*(4), 51. https://doi.org/10.17306/J.NPT.2016.4.51

Jeong, K. M., Zhao, J., Jin, Y., Heo, S. R., Han, S. Y., & Lee, J. (2015). Highly efficient extraction of anthocyanins from grape skin using deep eutectic solvents as green and tunable media. *Archives of Pharmacal Research*, *38*(12), 2143–2152. https://doi.org/10.1007/s12272015-0678-4

Kadam, S. U., Tiwari, B. K., & O'Donnell, C. P. (2013). Application of novel extraction technologies for bioactives from marine algae. *Journal of Agricultural and Food Chemistry*, *61*(20), 4667–4675. https://doi.org/10.1021/jf400819p

Kalhor, P., & Ghandi, K. (2019). Deep eutectic solvents for pretreatment, extraction, and catalysis of biomass and food waste. *Molecules*, *24*(22), 4012. https://doi.org/10.3390/molecules24224012

Kasprzak, M. M., Erxleben, A., & Ochocki, J. (2015). Properties and applications of flavonoid metal complexes. *RSC Advances*, *5*(57), 45853–45877. https://doi.org/10.1039/C5RA05069C

Kholany, M., Trebulle, P., Martins, M., Ventura, S. P., Nicaud, J., & Coutinho, J. A. P. (2019). Extraction and purification of violacein from *Yarrowia lipolytica* cells using aqueous solutions of surfactants. *Journal of Chemical Technology & Biotechnology*, *95*(4), 1126–1134. https://doi.org/10.1002/jctb.6297

Khoo, H. E., Azlan, A., Tang, S. T., & Lim, S. M. (2017). Anthocyanidins and anthocyanins: Colored pigments as food, pharmaceutical ingredients, and the potential health benefits. *Food & Nutrition Research*, *61*(1), 1361779. https://doi.org/10.1080/1654Sanlier6628.2017.1361779

Kocaadam, B., & Sanlier, N. (2017). Curcumin, an active component of turmeric (*Curcuma longa*), and its effects on health. *Critical Reviews in Food Science and Nutrition*, *57*(13), 2889–2895. https://doi.org/10.1080/10408398.2015.1077195

Leite, A. C., Ferreira, A. M., Morais, E. S., Khan, I., Freire, M. G., & Coutinho, J. A. P. (2017). Cloud point extraction of chlorophylls from spinach leaves using aqueous solutions of nonionic surfactants. *ACS Sustainable Chemistry & Engineering*, *6*(1), 590–599. https://doi.org/10.1021/acssuschemeng.7b02931

Li, Y., Fabiano-Tixier, A. S., Tomao, V., Cravotto, G., & Chemat, F. (2013). Green ultrasound-assisted extraction of carotenoids based on the bio-refinery concept using sunflower oil as an alternative solvent. *Ultrasonics Sonochemistry*, *20*(1), 12–18. https://doi.org/10.1016/j.ultsonch.2012.07.005

Li, L., & Yuan, H. (2013). Chromoplast biogenesis and carotenoid accumulation. *Archives of Biochemistry and Biophysics*, *539*(2), 102–109. https://doi.org/10.1016/j.abb.2013.07.002

Lima, Á. S., Soares, C. M. F., Paltram, R., Halbwirth, H., & Bica, K. (2017). Extraction and consecutive purification of anthocyanins from grape pomace using ionic liquid solutions. *Fluid Phase Equilibria*, *451*, 68–78. https://doi.org/10.1016/j.fluid.2017.08.006

Liu, Z., Yue, Z., Zeng, X., Cheng, J., & Aadil, R. M. (2019). Ionic liquid as an effective solvent for cell wall deconstructing through astaxanthin extraction from *Haematococcus pluvialis*. *International Journal of Food Science & Technology*, *54*(2), 583–590. https://doi.org/10.1111/ijfs.14030

Liu, Y., Li, J., Fu, R., Zhang, L., Wang, D., & Wang, S. (2019). Enhanced extraction of natural pigments from *Curcuma longa* L. using natural deep eutectic solvents. *Industrial Crops & Products*, *140*, 111620. https://doi.org/10.1016/j.indcrop.2019.111620

Liu, Z., Zeng, X., Cheng, J., Liu, D., & Aadil, R. M. (2018). The efficiency and comparison of novel techniques for cell wall disruption in astaxanthin extraction from *Haematococcus pluvialis*. *International Journal of Food Science & Technology*, *53*(9), 2212–2219. https://doi.org/10.1111/ijfs.13810

Lu, Y., Wang, L., Xue, Y., Zhang, C., Xing, X. H., Lou, K., … Su, Z. (2009). Production of violet pigment by a newly isolated psychrotrophic bacterium from a glacier in Xinjiang, China. *Biochemical Engineering Journal*, *43*(2), 135–141. https://doi.org/10.1016/j.bej.2008.09.009

Mai, N. L., Ahn, K., & Koo, Y.-M. (2014). Methods for recovery of ionic liquids—A review. *Process Biochemistry*, *49*(5), 872–881. https://doi.org/10.1016/j.procbio.2014.01.016

Marion, P., Bernela, B., Piccirilli, A., Estrine, B., Patouillard, N., Guilbot, J., & Jérôme, F. (2017). Sustainable chemistry: How to produce better and more from less? *Green Chemistry*, *19*(21), 4973–4989. https://doi.org/10.1039/C7GC02006F

Martins, M., Fernandes, A. P., Torres-Acosta, M. A., Collén, P. N., Abreu, M. H., & Ventura, S. P. (2020). Extraction of chlorophyll from wild and farmed *Ulva* spp. using aqueous solutions of ionic liquids. *Separation and Purification Technology*, *254*, 117589. https://doi.org/10.1016/j.seppur.2020.117589

Martins, M., Soares, B. P., Santos, J. H. P. M., Bharmoria, P., Torres-Acosta, M. A., Dias, A. C. R. V., … Ventura, S. P. M. (2020).

Phycobiliproteins purification by induced precipitation. *Green Chemistry*.

Martins, M., Ooi, C. W., Neves, M. C., Pereira, J. F., Coutinho, J. A. P., & Ventura, S. P. (2018). Extraction of recombinant proteins from *Escherichia coli* by cell disruption with aqueous solutions of surface-active compounds. *Journal of Chemical Technology & Biotechnology*, *93*(7), 1864–1870. https://doi.org/10.1002/jctb.5596

Martins, M., Vieira, F. A., Correia, I., Ferreira, R. A., Abreu, H., Coutinho, J. A. P., & Ventura, S. P. (2016). Recovery of phycobiliproteins from the red macroalga *Gracilaria* sp. Using ionic liquid aqueous solutions. *Green Chemistry*, *18*(15), 4287–4296. https://doi.org/10.1039/C6GC01059H

Martins, N., Roriz, C. L., Morales, P., Barros, L., & Ferreira, I. C. F. R. (2016). Food colorants: Challenges, opportunities and current desires of agro-industries to ensure consumer expectations and regulatory practices. *Trends in Food Science & Technology*, *52*, 1–15. https://doi.org/10.1016/j.tifs.2016.03.009

Martins, P. L. G., Braga, A. R., & de Rosso, V. V. (2017). Can ionic liquid solvents be applied in the food industry? *Trends in Food Science & Technology*, *66*, 117–124. https://doi.org/10.1016/j.tifs.2017.06.002

Martins, P. L. G., & de Rosso, V. V. (2016). Thermal and light stabilities and antioxidant activity of carotenoids from tomatoes extracted using an ultrasound-assisted completely solvent-free method. *Food Research International*, *82*, 156–164. https://doi.org/10.1016/j.foodres.2016.01.015

Marzorati, S., Schievano, A., Idà, A., & Verotta, L. (2020). Carotenoids, chlorophylls and phycocyanin from *Spirulina*: Supercritical CO_2 and water extraction methods for added value products cascade. *Green Chemistry*, *22*, 187–196. https://doi.org/10.1039/C9GC03292D

McCann, D., Barrett, A., Cooper, A., Crumpler, D., Dalen, L., Grimshaw, K., … Sonuga-Barke, E. (2007). Food additives and hyperactive behaviour in 3-year-old and 8/9-year-old children in the community: A randomised, double-blinded, placebo-controlled trial. *The Lancet*, *370*(9598), 1560–1567. https://doi.org/10.1016/S0140-6736(07)61306-3

Meagher, R. J., Light, Y. K., & Singh, A. K. (2008). Rapid, continuous purification of proteins in a microfluidic device using genetically-engineered partition tags. *Lab on a Chip*, *8*(4), 527–532. https://doi.org/10.1039/B716462A

Meng, Z., Zhao, J., Duan, H., Guan, Y., & Zhao, L. (2018). Green and efficient extraction of four bioactive flavonoids from Pollen Typhae by ultrasound-assisted deep eutectic solvents extraction. *Journal of Pharmaceutical and Biomedical Analysis*, *161*, 246–253. https://doi.org/10.1016/j.jpba.2018.08.048

Mensah, J., & Casadevall, S. R. (2019). Sustainable development: Meaning, history, principles, pillars, and implications for human action: Literature review. *Cogent Social Sciences*, *5*(1), 1653531. https://doi.org/10.1080/23311886.2019.1653531

Michalak, I., & Chojnacka, K. (2014). Algal extracts: Technology and advances. *Engineering in Life Sciences*, *14*(6), 581–591. https://doi.org/10.1002/elsc.201400139

Minich, D. M. (2019). A review of the science of colorful, plant-based food and practical strategies for "eating the rainbow". *Journal of Nutrition and Metabolism*, *2019*, 2125070. https://doi.org/10.1155/2019/2125070

Muhd Julkapli, N., Bagheri, S., & Bee Abd Hamid, S. (2014). Recent advances in heterogeneous photocatalytic decolorization of synthetic dyes. *The Scientific World Journal*, *2014*, 108653. https://doi.org/10.1155/2014/692307

Murador, D. C., de Souza Mesquita, L. M. S., Neves, B. V., Braga, A. R. C., Martins, P. L. G., Zepka, L. Q., & de Rosso, V. V. (2020). Bioaccessibility and cellular uptake by Caco-2 cells of carotenoids and chlorophylls from orange peels: A comparison between conventional and ionic liquid mediated extractions. *Food Chemistry*, *339*, 127818. https://doi.org/10.1016/j.foodchem.2020.127818

Murador, D. C., de Souza Mesquita, L. M., Vannuchi, N., Braga, A. R. C., & de Rosso, V. V. (2019). Bioavailability and biological effects of bioactive compounds extracted with natural deep eutectic solvents and ionic liquids: Advantages over conventional organic solvents. *Current Opinion in Food Science*, *26*, 25–34. https://doi.org/10.1016/j.cofs.2019.03.002

Murador, D. C., Braga, A. R. C., Martins, P. L., Mercadante, A. Z., & de Rosso, V. V. (2019). Ionic liquid associated with ultrasonic-assisted extraction: A new approach to obtain carotenoids from orange peel. *Food Research International*, *126*, 108653. https://doi.org/10.1016/j.foodres.2019.108653

Mussagy, C. U., Santos-Ebinuma, V. C., Gonzalez-Miquel, M., Coutinho, J. A. P., & Pereira, J. F. (2019). Protic ionic liquids as cell-disrupting agents for the recovery of intracellular carotenoids from yeast *Rhodotorula glutinis* CCT-2186. *ACS Sustainable Chemistry & Engineering*, *7*(19), 16765–16776. https://doi.org/10.1021/acssuschemeng.9b04247

Natarajan, S., Bajaj, H. C., & Tayade, R. J. (2018). Recent advances based on the synergetic effect of adsorption for removal of dyes from waste water using photocatalytic process. *Journal of Environmental Sciences*, *65*, 201–222. https://doi.org/10.1016/j.jes.2017.03.011

Oplatowska-Stachowiak, M., & Elliott, C. T. (2017). Food colors: Existing and emerging food safety concerns. *Critical Reviews in Food Science and Nutrition*, *57*(3), 524–548. https://doi.org/10.1080/10408398.2014.889652

Ordóñez-Santos, L. E., Pinzón-Zarate, L. X., & González-Salcedo, L. O. (2015). Optimization of ultrasonic-assisted extraction of total carotenoids from peach palm fruit (*Bactris gasipaes*) by-products with sunflower oil using response surface methodology. *Ultrasonics Sonochemistry*, *27*, 560–566. https://doi.org/10.1016/j.ultsonch.2015.04.010

Orr, V. C., Plechkova, N. V., Seddon, K. R., & Rehmann, L. (2015). Disruption and wet extraction of the microalgae *Chlorella vulgaris* using room-temperature ionic liquids. *ACS Sustainable Chemistry & Engineering*, *4*(2), 591–600. https://doi.org/10.1021/acssuschemeng.5b00967

Panić, M., Gunjević, V., Cravotto, G., & Redovniković, I. R. (2019). Enabling technologies for the extraction of grape-pomace anthocyanins using natural deep eutectic solvents in up-to-half-litre batches extraction of grape-pomace anthocyanins using NADES. *Food Chemistry*, *300*, 125185. https://doi.org/10.1016/j.foodchem.2019.125185

Parjikolaei, B. R., El-Houri, R. B., Fretté, X. C., & Christensen, K. V. (2015). Influence of green solvent extraction on carotenoid yield from shrimp (*Pandalus borealis*) processing waste. *Journal of Food Engineering*, *155*, 22–28. https://doi.org/10.1016/j.jfoodeng.2015.01.009

Passos, H., Freire, M. G., & Coutinho, J. A. P. (2014). Ionic liquid solutions as extractive solvents for value-added compounds from biomass. *Green Chemistry*, *16*(12), 4786–4815. https://doi.org/10.1039/C4GC00236A

Patsea, M., Stefou, I., Grigorakis, S., & Makris, D. P. (2017). Screening of natural sodium acetate-based low-transition temperature mixtures (LTTMs) for enhanced extraction of antioxidants and pigments from red vinification solid wastes. *Environmental Processes*, *4*(1), 123–135. https://doi.org/10.1007/s40710-016-0205-8

Pereira, J. F., Freire, M. G., & Coutinho, J. A. P. (2020). Aqueous two-phase systems: Towards novel and more disruptive applications. *Fluid Phase Equilibria*, *505*, 112341. https://doi.org/10.1016/j.fluid.2019.112341

Płotka-Wasylka, J., Rutkowska, M., Owczarek, K., Tobiszewski, M., & Namieśnik, J. (2017). Extraction with environmentally friendly solvents. *TrAC Trends in Analytical Chemistry*, *91*, 12–25. https://doi.org/10.1016/j.trac.2017.03.006

Poojary, M., Barba, F., Aliakbarian, B., Donsì, F., Pataro, G., Dias, D., & Juliano, P. (2016). Innovative alternative technologies to extract carotenoids from microalgae and seaweeds. *Marine Drugs*, *14*(11), 214. https://doi.org/10.3390/md14110214

Qi, X.-L., Peng, X., Huang, Y.-Y., Li, L., Wei, Z.-F., Zu, Y.-G., & Fu, Y.-J. (2015). Green and efficient extraction of bioactive flavonoids from *Equisetum palustre* L. by deep eutectic solvents-based negative pressure cavitation method combined with macroporous resin enrichment. *Industrial Crops and Products*, *70*, 142–148. https://doi.org/10.1016/j.indcrop.2015.03.026

Rao, N., Prabhu, M., Xiao, M., & Li, W.-J. (2017). Fungal and bacterial pigments: Secondary metabolites with wide applications. *Frontiers in Microbiology*, *8*, 1113. https://doi.org/10.3389/fmicb.2017.01113

Reynolds, C., Goucher, L., Quested, T., Bromley, S., Gillick, S., Wells, V. K., … Svenfelt, Å (2019). Consumption-stage food waste reduction interventions–What works and how to design better interventions. *Food Policy*, *83*, 7–27. https://doi.org/10.1016/j.foodpol.2019.01.009

Rodrigues, R. D. P., de Castro, F. C., de Santiago-Aguiar, R. S., & Rocha, M. V. P. (2018). Ultrasound-assisted extraction of phycobiliproteins from *Spirulina* (Arthrospira) platensis using protic ionic liquids as solvent. *Algal Research*, *31*, 454–462. https://doi.org/10.1016/j.algal.2018.02.021

Rodriguez-Amaya, D. B. (2016). Natural food pigments and colorants. *Current Opinion in Food Science*, *7*, 20–26. https://doi.org/10.1016/j.cofs.2015.08.004

Rodriguez-Amaya, D. B. (2019). Natural Food Pigments and Colorants. *Bioactive molecules in food*. Reference series in phytochemistry. Berlin, Germany: Springer. https://link.springer.com/referencework/10.1007%2F978-3-319-54528-8.

Ruiz, C. S., Martins, M., Coutinho, J. A. P., Wijffels, R., Eppink, M., Van den Berg, C., & Ventura, S. P. (2020). *Neochloris oleoabundans* biorefinery: Integration of cell disruption and purification steps using aqueous biphasic systems-based in surface-active ionic liquids. *Chemical Engineering Journal*, *399*, 125683. https://doi.org/10.1016/j.cej.2020.125683

Sahne, F., Mohammadi, M., Najafpour, G. D., & Moghadamnia, A. A. (2017). Enzyme-assisted ionic liquid extraction of bioactive compound from turmeric (*Curcuma longa* L.): Isolation, purification and analysis of curcumin. *Industrial Crops and Products*, *95*, 686–694. https://doi.org/10.1016/j.indcrop.2016.11.037

Saini, R. K., & Keum, Y.-S. (2018). Carotenoid extraction methods: A review of recent developments. *Food Chemistry*, *240*, 90–103. https://doi.org/10.1016/j.foodchem.2017.07.099

Salehi, B., Sharifi-Rad, J., Cappellini, F., Reiner, Ž., Zorzan, D., Imran, M., … Al-Sayed, E. (2020). The therapeutic potential of anthocyanins: Current approaches based on their molecular mechanism of action. *Frontiers in Pharmacology*, *11*, 1300. https://doi.org/10.3389/fphar.2020.01300

Schaeffer, N., Kholany, M., Veloso, T. L., Pereira, J. L., Ventura, S. P., Nicaud, J.-M., & Coutinho, J. A. P. (2019). Temperature-responsive extraction of violacein using a tuneable anionic surfactant-based system. *Chemical Communications*, *55*(59), 8643–8646.

Schanes, K., Dobernig, K., & Gözet, B. (2018). Food waste matters - A systematic review of household food waste practices and their policy implications. *Journal of Cleaner Production*, *182*, 978–991. https://doi.org/10.1016/j.jclepro.2018.02.030

Schonbrun, Z. (2018). The quest for the next billion-dollar color. Retrieved from https://www.bloomberg.com/features/2018-quest-for-billion-dollar-red/

Schuur, B., Brouwer, T., Smink, D., & Sprakel, L. M. (2019). Green solvents for sustainable separation processes. *Current Opinion in Green and Sustainable Chemistry*, *18*, 57–65. https://doi.org/10.1016/j.cogsc.2018.12.009

Sheldon, R. A. (2016). Green chemistry and resource efficiency: Towards a green economy. *Green Chemistry*, *18*(11), 3180–3183. https://doi.org/10.1039/C6GC90040B

Shi, M., He, N., Li, W., Li, C., & Kang, W. (2018). Simultaneous determination of myricetrin, quercitrin and afzelin in leaves of *Cercis chinensis* by a fast and effective method of ionic liquid microextraction coupled with HPLC. *Chemistry Central Journal*, *12*(1), 23. https://doi.org/10.1186/s13065-018-0391-8

Sintra, T. E., Bagagem, S. S., Ahsaie, F. G., Fernandes, A., Martins, M., Macário, I. P., … Ventura, S. P. (2020). Sequential recovery of C-phycocyanin and chlorophylls from Anabaena cylindrica. *Separation and Purification Technology*, *255*, 117538. https://doi.org/10.1016/j.seppur.2020.117538

Smith, E. L., Abbott, A. P., & Ryder, K. S. (2014). Deep eutectic solvents (DESs) and their applications. *Chemical Reviews*, *114*(21), 11060–11082. https://doi.org/10.1021/cr300162p

Solomon, M. R., White, K., Dahl, D. W., Zaichkowsky, J. L., & Polegato, R. (2017). *Consumer behavior: Buying, having, and being*. Boston, MA: Pearson. https://books.google.com.br/books?id=FCcXswEACAAJ&dq=Consumer+behavior:+Buying,+having,+and+being+2017&hl=pt-PT&sa=X&ved=2ahUKEwjevPK0jrjtAhULK7kGHduFD_wQ6AEwAHoECAAQAg.

Sonar, C. R., Rasco, B., Tang, J., & Sablani, S. S. (2019). Natural color pigments: Oxidative stability and degradation kinetics during storage in thermally pasteurized vegetable purees. *Journal of the Science of Food and Agriculture*, *99*(13), 5934–5945. https://doi.org/10.1002/jsfa.9868

Spence, C. (2015). On the physiological impact of food colour. *Flavour*, *4*(1), 21. https://doi.org/10.1186/s13411-015-0031-3

Suarez Ruiz, C. A., Emmery, D. P., Wijffels, R. H., Eppink, M. H., & Van den Berg, C. (2018). Selective and mild fractionation of microalgal proteins and pigments using aqueous two-phase systems. *Journal of Chemical Technology & Biotechnology*, *93*(9), 2774–2783. https://doi.org/10.1002/jctb.5711

Sun, J., Shi, J., Konda, N. M., Campos, D., Liu, D., Nemser, S., … Gurau, G. (2017). Efficient dehydration and recovery of ionic liquid after lignocellulosic processing using pervaporation. *Biotech-*

nology for Biofuels, *10*(1), 154. https://doi.org/10.1186/s13068-017-0842-9

Szczepańska, J., Barba, F. J., Skąpska, S., & Marszałek, K. (2020). High pressure processing of carrot juice: Effect of static and multi-pulsed pressure on the polyphenolic profile, oxidoreductases activity and colour. *Food Chemistry*, *307*, 125549. https://doi.org/10.1016/j.foodchem.2019.125549

Tan, Z., Yi, Y., Wang, H., Zhou, W., & Wang, C. (2016). Extraction, preconcentration and isolation of flavonoids from *Apocynum venetum* L. Leaves using ionic liquid-based ultrasonic-assisted extraction coupled with an aqueous biphasic system. *Molecules*, *21*(3), 262. https://doi.org/10.3390/molecules21030262

Torres-Acosta, M. A., dos Santos, N. V., Ventura, S. P., Coutinho, J. A. P., Rito-Palomares, M., & Pereira, J. F. (2020). Economic analysis of the production and recovery of green fluorescent protein using ATPS-based bioprocesses. *Separation and Purification Technology*, *254*, 117595.

Tuli, H. S., Chaudhary, P., Beniwal, V., & Sharma, A. K. (2015). Microbial pigments as natural color sources: Current trends and future perspectives. *Journal of Food Science and Technology*, *52*(8), 4669–4678. https://doi.org/10.1007/s13197-014-1601-6

Ulloa, G., Coutens, C., Sánchez, M., Sineiro, J., Fábregas, J., Deive, F. J., … Núñez, M. J. (2012). On the double role of surfactants as microalga cell lysis agents and antioxidants extractants. *Green Chemistry*, *14*(4), 1044–1051. https://doi.org/10.1039/C2GC16262H

Venil, C. K., Aruldass, C. A., Dufossé, L., Zakaria, Z. A., & Ahmad, W. A. (2014). Current perspective on bacterial pigments: Emerging sustainable compounds with coloring and biological properties for the industry—An incisive evaluation. *RSC Advances*, *4*(74), 39523–39529. https://doi.org/10.1039/c4ra06162d

Ventura, S. P., e Silva, F. A., Quental, M. V., Mondal, D., Freire, M. G., & Coutinho, J. A. P. (2017). Ionic-liquid-mediated extraction and separation processes for bioactive compounds: Past, present, and future trends. *Chemical Reviews*, *117*(10), 6984–7052. https://doi.org/10.1021/acs.chemrev.6b00550

Ventura, S. P., Santos-Ebinuma, V. C., Pereira, J. F., Teixeira, M. F., Pessoa, A., & Coutinho, J. A. P. (2013). Isolation of natural red colorants from fermented broth using ionic liquid-based aqueous two-phase systems. *Journal of Industrial Microbiology & Biotechnology*, *40*(5), 507–516. https://doi.org/10.1007/s10295-013-1237-y

Vernès, L., Granvillain, P., Chemat, F., & Vian, M. (2015). Phycocyanin from *Arthrospira platensis*. Production, extraction and analysis. *Current Biotechnology*, *4*(4), 481–491. https://doi.org/10.2174/2211550104666151006002418

Vicente, F. A., Plazl, I., Ventura, S. P., & Žnidaršič-Plazl, P. (2020). Separation and purification of biomacromolecules based on microfluidics. *Green Chemistry*, *22*, 4391–4410. https://doi.org/10.1039/C9GC04362D

Vicente, F. A., Cardoso, I. S., Martins, M., Gonçalves, C. V., Dias, A. C., Domingues, P., … Ventura, S. P. (2019). R-phycoerythrin extraction and purification from fresh *Gracilaria* sp. Using thermo-responsive systems. *Green Chemistry*, *2019*(21), 3816–3826. https://doi.org/10.1039/C9GC00104B

Vicente, F. A., Lario, L. D., Pessoa Jr, A., & Ventura, S. P. (2016). Recovery of bromelain from pineapple stem residues using aqueous micellar two-phase systems with ionic liquids as co-surfactants. *Process Biochemistry*, *51*(4), 528–534. https://doi.org/10.1016/j.procbio.2016.01.004

Vieira, F. A., Guilherme, R. J., Neves, M. C., Abreu, H., Rodrigues, E. R., Maraschin, M., … Ventura, S. P. (2017). Single-step extraction of carotenoids from brown macroalgae using non-ionic surfactants. *Separation and Purification Technology*, *172*, 268–276. https://doi.org/10.1016/j.seppur.2016.07.052

Vieira, F. A., Guilherme, R. J., Neves, M. C., Rego, A., Abreu, M. H., Coutinho, J. A. P., & Ventura, S. P. (2018). Recovery of carotenoids from brown seaweeds using aqueous solutions of surface-active ionic liquids and anionic surfactants. *Separation and Purification Technology*, *196*, 300–308. https://doi.org/10.1016/j.seppur.2017.05.006

Vieira, F. A., & Ventura, S. P. (2019). Efficient extraction of carotenoids from *Sargassum muticum* using aqueous solutions of Tween 20. *Marine Drugs*, *17*(5), 310–320. https://doi.org/10.3390/md17050 https://doi.org/10.3390/md17050310310

Wang, H., Ma, X., Cheng, Q., Wang, L., & Zhang, L. (2018). Deep eutectic solvent-based ultrahigh pressure extraction of baicalin from *Scutellaria baicalensis* Georgi. *Molecules*, *23*(12), 3233. https://doi.org/10.3390/molecules23123233

Wang, R., Chang, Y., Tan, Z., & Li, F. (2016). Applications of choline amino acid ionic liquid in extraction and separation of flavonoids and pectin from ponkan peels. *Separation Science and Technology*, *51*(7), 1093–1102. https://doi.org/10.1080/01496395.2016.1143006

Wypych, G. (Ed.). (2019). *Handbook of solvents, Volume 2: Use, health, and environment*. Amsterdam, the Netherlands: Elsevier. https://books.google.com.br/books?hl=pt-PT&lr=&id=VFuJDwAAQBAJ&oi=fnd&pg=PP1&dq=Handbook+of+solvents,+Volume+2:+Use,+health,+and+environment&ots=kwofBiMlj5&sig=m-q26KJHzTkXrDuM9IEuMg25S0A#v=onepage&q=Handbook%20of%20solvents%2C%20Volume%202%3A%20Use%2C%20health%2C%20and%20environment&f=false.

Xia, N.-S., Luo, W.-X., Zhang, J., Xie, X.-Y., Yang, H.-J., Li, S.-W., … Ng, M.-H. (2002). Bioluminescence of *Aequorea macrodactyla*, a common jellyfish species in the East China Sea. *Marine Biotechnology*, *4*(2), 155–162. https://doi.org/10.1007/s10126-001-0081-7

Xu, J. J., Li, Q., Cao, J., Warner, E., An, M., Tan, Z., … Liu, X. G. (2016). Extraction and enrichment of natural pigments from solid samples using ionic liquids and chitosan nanoparticles. *Journal of Chromatography A*, *1463*, 32–41. https://doi.org/10.1016/j.chroma.2016.08.012

Xu, W., Chu, K., Li, H., Zhang, Y., Zheng, H., Chen, R., & Chen, L. (2012). Ionic liquid-based microwave-assisted extraction of flavonoids from *Bauhinia championii* (Benth.) Benth. *Molecules*, *17*(12), 14323–14335. https://doi.org/10.3390/molecules171214323

Yara-Varón, E., Li, Y., Balcells, M., Canela-Garayoa, R., Fabiano-Tixier, A.-S., & Chemat, F. (2017). Vegetable oils as alternative solvents for green oleo-extraction, purification and formulation of food and natural products. *Molecules*, *22*(9), 1474. https://doi.org/10.3390/molecules22091474

Yang, L. H., Xiong, H., Lee, O. O., Qi, S.-H., & Qian, P.-Y. (2007). Effect of agitation on violacein production in *Pseudoalteromonas luteoviolacea* isolated from a marine sponge. *Letters in Applied Microbiology*, *44*(6), 625–630. https://doi.org/10.1111/j.1472-765X.2007.02125.x

Yusuf, M., Shabbir, M., & Mohammad, F. (2017). Natural colorants: Historical, processing and sustainable prospects. *Natural Products and Bioprospecting*, *7*(1), 1–23. https://doi.org/10.1007/s13659-017-0119-9

Zamora-Garcia, I. R., Altorre-Ordaz, A., Ibanez, J. G., Torres-Elguerra, J. C., Wrobel, K., & Gutierrez-Granados, S. (2018). Efficient degradation of selected polluting dyes using the tetrahydroxoargentate ion, Ag(OH)4−, in alkaline media. *Chemosphere*, *191*(2018), 400–407. https://doi.org/10.1016/j.chemosphere.2017.10.041

Zhao, C., Lu, Z., Li, C., He, X., Li, Z., Shi, K., … Zu, Y. (2014). Optimization of ionic liquid based simultaneous ultrasonic-and microwave-assisted extraction of rutin and quercetin from leaves of velvetleaf (*Abutilon theophrasti*) by response surface methodology. *The Scientific World Journal*, *2014*, 283024. https://doi.org/10.1155/2014/283024

Zhou, J., Sui, H., Jia, Z., Yang, Z., He, L., & Li, X. (2018). Recovery and purification of ionic liquids from solutions: A review. *RSC Advances*, *8*(57), 32832–32864. https://doi.org/10.1039/C8RA06384B

Zhou, P., Wang, X., Liu, P., Huang, J., Wang, C., Pan, M., & Kuang, Z. (2018). Enhanced phenolic compounds extraction from *Morus alba* L. leaves by deepeutectic solvents combined with ultrasonic-assisted extraction. *Industrial Crops & Products*, *120*, 147–154. https://doi.org/10.1016/j.indcrop.2018.04.071

Zhou, Y., Wu, D., Cai, P., Cheng, G., Huang, C., & Pan, Y. (2015). Special effect of ionic liquids on the extraction of flavonoid glycosides from *Chrysanthemum morifolium* Ramat by microwave assistance. *Molecules*, *20*(5), 7683–7699. https://doi.org/10.3390/molecules20057683

Zhuang, B., Dou, L.-L., Li, P., & Liu, E.-H. (2017). Deep eutectic solvents as green media for extraction of flavonoid glycosides and aglycones from *Platycladi Cacumen*. *Journal of Pharmaceutical and Biomedical Analysis*, *134*, 214–219. https://doi.org/10.1016/j.jpba.2016.11.049

Zimmerman, J. B., Anastas, P. T., Erythropel, H. C., & Leitner, W. (2020). Designing for a green chemistry future. *Science*, *367*(6476), 397–400. https://doi.org/10.1126/science.aay3060

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