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Review

Optimization of Extraction of Natural Antimicrobial Pigments Using Supercritical Fluids: A Review

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Abstract: It has become increasingly popular to replace chemically synthesized compounds with natural counterparts mostly found in natural sources, such as natural pigments. The conventional extraction processes for these compounds are limited by the toxicity and flammability of the solvents. To obtain pure extracts, it is always a longer process that requires several steps. Supercritical fluid extraction (SFE) is a cutting-edge green technology that is continuously increasing and expanding its fields of application, with benefits such as no waste produced, shorter extraction time, automation, and lower solvent consumption. The SFE of natural pigments has high potential in food, textiles, cosmetics, and pharmaceuticals; there are a number of other applications that can benefit from the SFE technique of natural pigments. The pigments that are extracted via SFE have a high potential for application and sustainability because of their biological and antimicrobial properties as well as low environmental risk. This review provides an update on the SFE technique, specifically as it pertains to the optimization of health-promoting pigments. This review focuses on antimicrobial pigments and the high efficiency of SFE in extracting pure antimicrobial pigments. In addition, the optimal conditions, biological activities, and possible applications of each category are explained.

Keywords: supercritical fluid; extraction; natural pigments; antimicrobial activity; optimization

Citation: Abou Elmaaty, T.; Sayed-Ahmed, K.; Elsisi, H.; Magdi, M. Optimization of Extraction of Natural Antimicrobial Pigments Using Supercritical Fluids: A Review. *Processes* **2022**, *10*, 2111. https://doi.org/10.3390/pr10102111

Academic Editor: Adriana Trifan

Received: 9 September 2022
Accepted: 13 October 2022
Published: 18 October 2022

1. Introduction

The use of sustainable natural products has become increasingly important in response to the growing awareness of the adverse effects of synthetic products on the environment [1]. Therefore, it is necessary to choose suitable extraction methods and conditions to obtain excellent extraction yields for natural products. Natural colorants, extracted from plants, animals, insects, and minerals, are bioresources for pigments and dyes with no negative environmental effects. Natural colorants have grown in acceptance as a result of their coloring qualities and health-promoting benefits, in addition to their low cytotoxicity compared with synthetic colorants [2,3]. These colorants can enhance the antimicrobial properties of textiles [4–9]. Natural pigments such as anthocyanins, carotenoids, and chlorophylls show outstanding antimicrobial activity against various pathogens, including different bacterial and fungal strains. These natural colorants exhibit a high potential for applications in various fields, particularly textiles, the food industry, and pharmaceuticals [10–15].

One of the most crucial processes in the production of natural colorants is pigment extraction [16]. The initial step of the extraction process is to separate the crude pigment from the starting material. In plant materials, most bioactive molecules are located inside plant cells that are enclosed with a pectocellulosic wall comprising a complex of cellulosic structures that consist of sugar alcohols and ether linkages between carbohydrates and proteins and is also strengthened by lignin [17]. Plant material can be extracted using various methods. Traditional techniques such as maceration and Soxhlet extraction are typically used [16]. Recently, nonconventional techniques such as pressurized liquid extraction (PLE), microwave-assisted extraction (MAE), ultrasound extraction (UAE), pulsed

electric fields extraction (PEFE), and, the subject of this review, supercritical fluid extraction (SFE) have allowed for the use of alternative solvents while ensuring a safe, cost-effective, and high-quality extraction [18,19]. Compared with traditional procedures, the extraction methods described above are preferable. These nonconventional methods can function in the absence of light and oxygen at high temperatures or pressures, with minimal organic solvent use and short extraction times. The main difficulties with traditional extraction methods are their extended extraction times, expensive toxic solvents, limited selectivity, and heat breakdown of thermally labile chemicals. These restrictions can be overcome using nonconventional extraction methods [20]. Therefore, it is necessary to select a suitable extraction method and conditions to obtain an excellent extraction yield for natural products. A suitable extraction technique helps to increase the extraction yield and prevent the degradation of extracted pigments, leading to the production of natural colorants of higher quality [16]. The SFE method has several advantages that make it a promising green alternative. This technology may provide nontoxic solvents that can be easily removed from the extract, with a low extraction temperature, high recovery of bioactive compounds (particularly for the extraction of heat-sensitive colorants), rapid mass transfer, excellent selectivity, continuous flow of fresh fluid, and scale-up for industrial processes [21]. Therefore, the extraction of natural antimicrobial dyes and pigments using SFE has attracted the interest of researchers. In addition, the optimization of SFE conditions is necessary to enhance the technique's extraction efficiency. In this respect, several SFE parameters, such as flow rate (FR), particle size (PS), temperature (Temp), time (T), pressure (P), sample weight (SW), and co-solvent ratio and type, can be optimized to enhance the overall extraction using supercritical fluids (SCFs). In addition, the SFE system involves the use of a supercritical fluid (SCF) (typically carbon dioxide (CO_2), which has supercritical properties higher than 31.1 °C and 7.38 MPa) to isolate the target compounds under optimal operating conditions [22–24].

This review compares the SFE technique and traditional extraction methods to illustrate the advantages and disadvantages of SFE compared with the conventional methods. In addition, SFE principles and mechanisms are mentioned, as well as the history of the development of SFE technology up to the present. The optimization of various extraction parameters, including the co-solvent ratio, flow rate, time, temperature, raw matrix, and pressure, is discussed in detail. This review focuses on the extraction of antimicrobial colorants from natural sources. The optimal SFE conditions for each category of antimicrobial pigments and dyes are summarized.

2. Green Extraction

Extraction is a technique of isolating components from natural materials using chemical or physical methods. Recently, the world has turned to using green extraction as part of its efforts to preserve the environment [25,26]. Green extraction is based on procedures that require less energy, allow for the use of alternative solvents and sustainable natural resources, and offer a safe and high-quality extract. SFE procedures are compatible with the principles of green extraction. It is considered the most effective alternative for traditional solvent extraction methods of bioactive substances, especially when supercritical carbon dioxide ($scCO_2$) is used as the green solvent. The six principles of the green extraction of natural products are as follows [27–29]:

(a) Innovation through using sustainable plant resources

Green extraction requires either intense culture or in vitro development of plant cells or organisms to protect natural products from extinction. Natural colorants can be obtained from natural resources such as plants, animals, and fungi, but in a manner that preserves the rights of future generations.

(b) Use of alternative solvents, principally water and safe solvents

Using the SFE technique, the active components can be produced without any solvent residue. CO_2, which is frequently used in the SFE method, is a nonflammable odorless gas

formed when fossil fuels are burned, alcohol is fermented, and during human and animal respiration. The SFE technique uses compressed supercritical CO_2 ($scCO_2$) at a pressure of up to 300 MPa and a temperature of 30 to 40 °C to replace organic solvents such as hexane in the extraction process.

(c) Reduced energy consumption by energy recovery using innovative technologies

Extraction is affected by economic and environmental issues, which require a drastic decrease in energy consumption and waste production. Compared with traditional extraction techniques, the SFE method is a rapid extraction. Further steps are not required to save time or energy.

(d) Production of co-products instead of waste to include bio- and agro-refining industries

Extraction operations produce a wide variety of additional materials such as co-products, by-products, or waste. According to the biorefinery concept, plant materials are used in an integrated manner. Plants contain various refined compounds. Each component of a plant can be isolated and used to produce a variety of products. The SFE procedure at low temperatures allows for the discovery of new compounds that enhance the value of the extract by producing co-products that can be functionalized.

(e) Reduced unit operations and safe-controlled processes

Reducing the number of stages in a production chain lowers costs and makes better use of the energy. The optimal procedure appears to be a single-stage process. Supercritical fluid extraction has the benefit of using a clean solvent and producing an extract using technology with a minimal number of discrete operations.

(f) Nondenatured and biodegradable extracts without contaminants

The extract must adhere to all the laws, regulations, quality standards, and market demands. In addition, the extract guarantees no harm to humans or the environment. According to this principle, the SFE technique protects thermally labile target compounds. In addition, the SFE extract has no solvent residue, making it highly pure.

3. Supercritical Fluids (SCFs)

3.1. History of Supercritical Fluids

The first discovery of a supercritical fluid (SCF) was made in 1822 by Baron Charles Cagniard de la Tour, who noticed that solvent behavior changed at a particular pressure and temperature [26,30]. In 1869, Thomas Andrews introduced the modern term "critical point" in his findings on the effects of temperature and pressure on partially liquefied carbonic acid in sealed glass tubes. He defined the critical point as the characteristic temperature (T_c) and pressure (P_c) on the phase equilibrium curve, where two distinctive phases do not exist [26,31]. In 1879, Hannay and Hogharth discovered that SCFs have a high potential to dissolve solid matter and fluids [26,32]. Subsequently, Zosel et al. [28] designed fundamental methods for extracting natural components using supercritical carbon dioxide ($scCO_2$). Therefore, this technology was used in the 1970s for the decaffeination of coffee and the extraction of oils from hops. In the 1980s, SFE technology was developed on an industrial scale in Europe, the USA, and Australia. Furthermore, the first scientific journal *The Journal of Supercritical Fluids* was published in 1988 because of an increase in the scientific research and patents of the SFE technique [32]. Currently, this technology is used to produce a wide range of products around the world [26].

A supercritical fluid (SCF) is any substance at a temperature and pressure above its critical point. The critical point is the maximum temperature and pressure at which a substance may exist in equilibrium as a vapor and liquid [33]. At this point, the fluid is represented by its gas and liquid phase characteristics. It diffuses through solids (similar to gases) and dissolves substances (similar to liquids) [34]. Figure 1 shows a standard phase diagram for pure carbon dioxide. Carbon dioxide has a critical point of 31.1 °C and 78.3 bar [26].

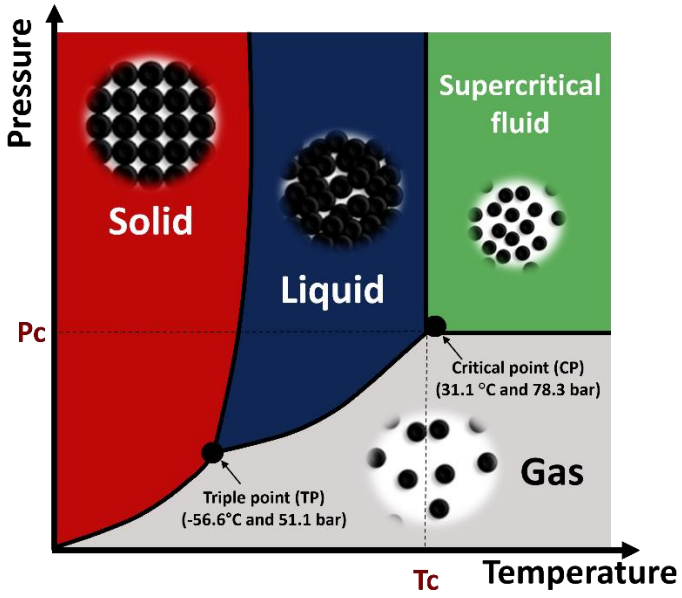


Figure 1. Standard phase diagram for pure carbon dioxide [25].

3.2. Properties of Supercritical Fluids

An SCF behave like a gas; hence, it fills and takes the form of a container. The mobility of the molecules is comparable to that of gas molecules. In addition, an SCF provides liquid properties as its density is close to that of a liquid which affects its dissolving power, as shown in Figure 2. Density, viscosity, and diffusivity are the three most essential parameters of an SCF [25,26]. The density of an SCF is between that of the gas and liquid. In the supercritical state, the density of the SCF increases when the pressure increases at a constant temperature; however, it decreases with rising temperature at a constant pressure. Density is a key parameter of SCFs when they are used as a solvent. Hence, the dissolution effect of SCFs is controlled by their density. The viscosity of the SCF is similar to that of a gas, but less than that of a liquid. Thus, low viscosity enhances the penetration power of an SCF and allows the components to readily flow through it. In addition, temperature has little effect on the liquid viscosity, but significantly influences the SCF's viscosity [34]. The diffusivity of the SCF is higher than that of liquids and lower than the diffusivity of gases. It is directly proportional to the temperature and inversely proportional to the pressure. Owing to the high diffusivity of SCFs, they have the potential to be suitable solvents and a fast carrier for extraction processes [34,35].

As previously stated, these characteristics are linked, and SFE is considered to be an excellent technique for the extraction of natural bioactive compounds [26]. Many chemicals have been utilized as supercritical fluids, and their critical characteristics are listed in Table 1.

Researchers are particularly interested in $scCO_2$ due to its green and sustainable properties [27]. CO_2 has been used as a supercritical solvent in more than 90% of SFE processes because of its low critical temperature (31.1 °C) and low critical pressure (72.8 bar) [36]. Water is superior to carbon dioxide in terms of sustainability; however, to become a supercritical solvent, special conditions are required that are difficult to easily provide (critical temperature of 374.15 °C and critical pressure of 220.64 bar). $scCO_2$ has unique properties that make it a desirable compound for the extraction of bioactive components from plant and animal materials [26]. The properties of CO_2 are shown in Figure 3.

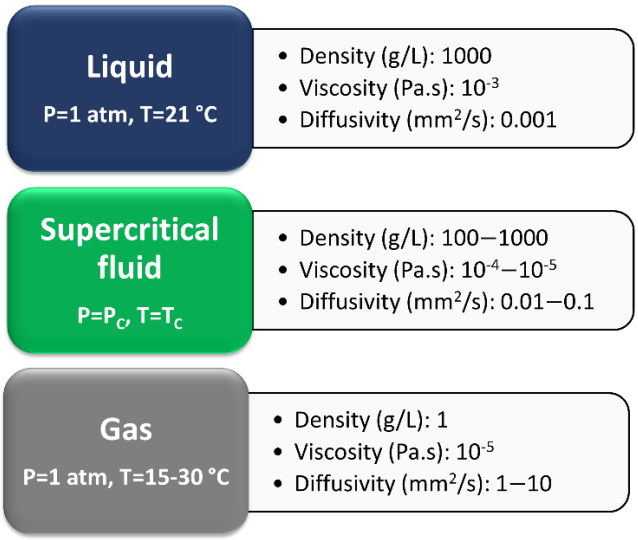


Figure 2. Comparison of the physical and chemical properties of liquids, supercritical fluids, and gases [35].

Table 1. Various compounds used as supercritical fluids and their properties [35].

Solvent	Molecular Weight (g/mol)	Critical Temperature (k)	Critical Pressure (MPa)	Critical Density (g/cm^3)
Carbon dioxide	44.01	304.1	7.38	0.469
Water	18.015	647.3	22.064	0.348
Methane	16.04	190.4	4.60	0.162
Ethane	30.07	305.3	4.87	0.203
Propane	44.09	369.8	4.25	0.217
Ethylene	28.05	282.4	5.04	0.215
Propylene	42.08	364.9	4.60	0.232
Methanol	32.04	512.6	8.09	0.272
Ethanol	46.07	513.9	6.14	0.276
Benzene	78.11	562	4.89	0.876
Acetone	58.08	508.1	4.70	0.278
Pentane	72.15	469.6	3.369	0.273
Butane	58.12	425.16	3.796	0.225
Hexane	86.178	507.44	3.031	0.233

$scCO_2$ applications can be listed as follows [33]:

- Extraction and fractionation of products such as cannabinoids from plants [37] and microbial natural products from myxobacterial strains [38];
- Dyeing of synthetic [39–45] and natural fabrics [46–51];
- Treatment of polluted soils;
- Manufacture of micron- and submicron-sized powders, as well as processes in or with SFCs;

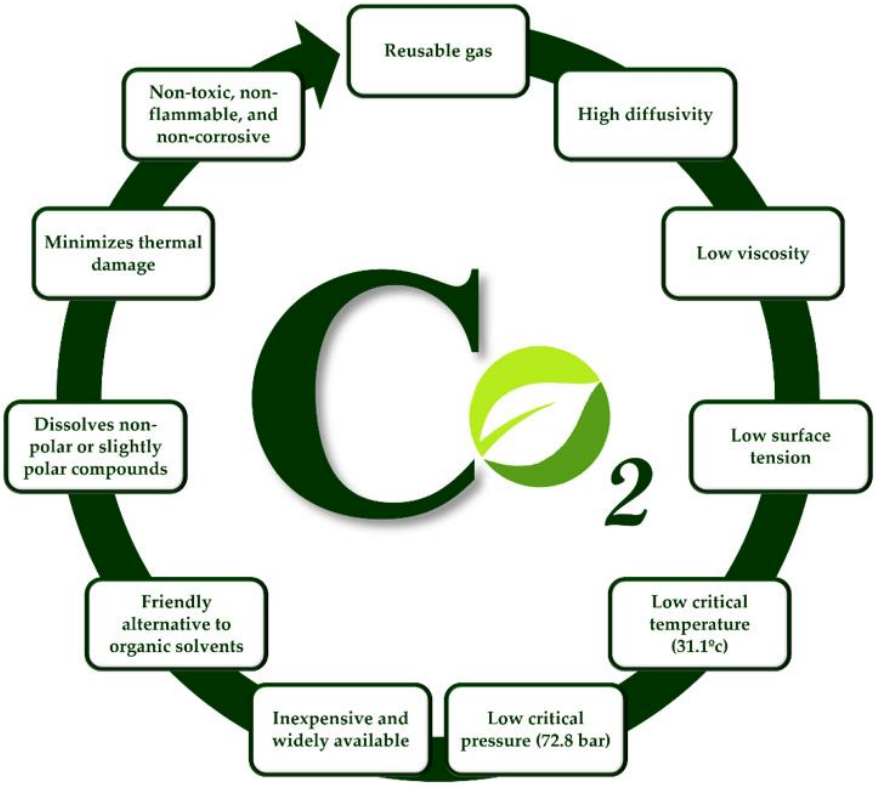


Figure 3. The properties of carbon dioxide [25,26,35].

4. Supercritical Fluid Extraction (SFE)

The SFE method is a separation technique in which natural chemical components are dissolved in a fluid that can change its dissolving power above a critical temperature and pressure under certain conditions [52]. SCFs can be used to selectively extract certain components from plants. This is a more sustainable alternative to conventional extraction methods [26,31].

4.1. Supercritical Fluid Extraction Principles

The SFE is determined by the solvating properties of a supercritical fluid, which can be generated by applying pressure and temperature above the critical point of a substance. Each compound exhibits a distinct critical point [53]. The yields of the SFE process, as well as the properties and chemical composition of the extracted material, are influenced by the type of SCF used and process parameters (temperature, pressure, extraction time, etc.) [32]. The extractability of the SCF can be adjusted by appropriately managing the SFE parameters, allowing this technology to find applications ranging from food to pesticide research.

4.2. Supercritical Fluid Extraction Instrument

The SFE apparatus is classified into four scales based on the vessel volume: analytical (1–24 mL), bench (200–500 mL), pilot (1–50 L), and production (350+ L). Regardless of complexity or cost, all extraction systems have the same fundamental components such as a CO_2 resource, chiller unit, CO_2 pump, co-solvent reservoir, co-solvent pump, valves, heater controller, extractor, heating jacket, back pressure regulator, vessel, equilibration coil, pressure vessels, and collector, as shown in Figure 4 [54,55].

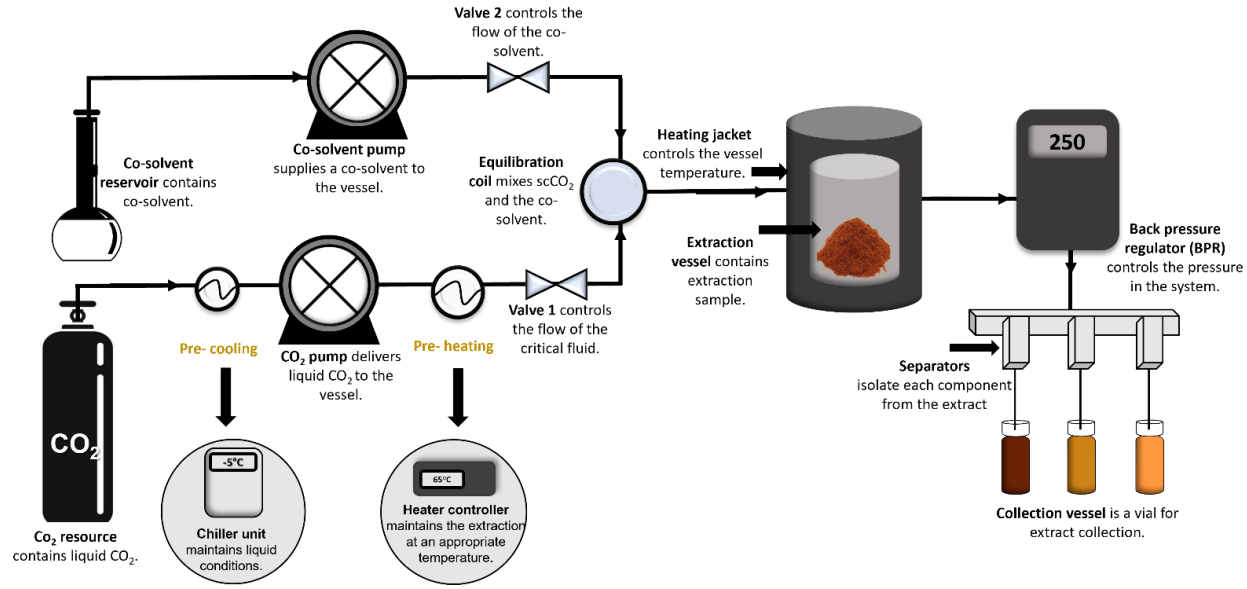


Figure 4. Schematic of the laboratory-scale supercritical fluid extractor.

4.3. Supercritical Fluid Extraction Mechanism

The SFE method has been applied to thousands of solid-sample matrices. The SCF extracts and transports target components that can be solubilized under the specified temperature and pressure parameters. The SFE procedure can be performed in two principal modes: static and dynamic extraction [54]. During static extraction, the plant matrix is exposed to a constant amount of $scCO_2$ for a certain amount of time. The static mode is utilized to allow fluid (e.g., $scCO_2$) to penetrate the plant matrix and dissolve the analytes. In dynamic extraction, the plant matrix is continuously fed fresh CO_2. Both the static and dynamic modes are combined in most SFE experiments. A static mode allows $scCO_2$ to penetrate the plant material and dissolve the analytes. The static mode is followed by dynamic mode, which sweeps the analytes through the restrictor from the extraction vessel into the collecting system [54]. After the fluid and dissolved compounds are carried to the separators, the products are collected through a tap placed at the bottom of the separators. After depressurization, the fluid is either cycled or released into the surrounding media; then, the extract can be collected [55,56].

5. Critical Parameters in the SFE

The initial step in the SFE technique is to optimize the experimental conditions to achieve an acceptable extraction of the targeted analytes while avoiding the co-extraction of other undesirable components. Because different process variables may influence the extraction efficiency, optimizing the operational parameters is an important step in the development of the SFE technique. The efficient extraction of bioactive components from plant materials is dependent on several SFE parameters, which can be optimized [57]. Temperature, pressure, co-solvent ratio, extraction time, flow rate, and raw matrix (particle size, moisture content, and pre-treatment) are the primary variables influencing extraction efficacy. The optimal ranges for each parameter are summarized in Table 2.

Table 2. The optimal ranges for critical parameters in the SFE technique.

Parameter	The Optimal Range	
Temperature	35–60 °C	
Pressure	Around 40 MPa (in the case of $scCO_2$)	
Co- solvent	Concentration	Below 1–10% (in the case of $scCO_2$)
	Type	Ethanol (in the case of food industry)Methanol (in the case of analytical operations)
Time	Less than 2 h	
Flow rate	1–10 L/min (in the case of $scCO_2$)	
Raw material	Particle size	0.25 to 2.0 mm
	Moisture content	4–14%
	Pre-treatment	Freeze-dried samples

5.1. Temperature

The extraction temperature has two varied effects at constant pressure. Increasing the temperature reduces the density of the solvent and its solvating capacity. However, increasing the temperature increases the vapor pressure of the desired compounds, improving the compound's solubility and extraction yield. Thus, this may cause the isotherms to cross, a phenomenon known as retrogradation, in which high temperatures result in low yields, and lower temperatures produce a rich extract. These opposing effects on the total extraction yield are responsible for the inversion of yield isotherms [26]. By considering the crossover feature, Mezzomo et al. proposed that the density impact is dominant at pressures below the crossover pressure, whereas the solute vapor pressure is the primary mechanism controlling the extraction process at higher pressures [25]. A higher temperature results in a lower extraction recovery of nonvolatile components. However, there is a competition between their solubility in an SCF and the volatility of their volatile components. Solubility decreases with increasing temperature, whereas volatility increases with increasing temperature. However, increasing temperature results in an increase in extraction efficiency; many heat-sensitive compounds may degrade or oxidize, losing their biological activity at higher extraction temperatures [30,53]. Therefore, the SFE temperature of thermolabile compounds must be set between 35 and 60 °C to avoid degradation [57].

5.2. Pressure

Pressure is one of the most critical parameters controlling the SFE process, owing to its effect on the solubility of a substance [58]. Pressure control in the SFE technique can be implemented using a back pressure regulator (BPR) that maintains the SCF pressure at the desired level [54]. At a constant temperature, increasing the pressure results in increased solvent power and corresponding extraction efficacy, which improves the solubility of the bioactive compounds and the extraction yield. Additionally, the density of the SCF increases with increasing pressure. Consequently, the solubility increases, which leads to a higher recovery of the target compounds [59]. Pressure affects the volatile components of the target compound. Hence, increased pressure results in a larger recovery of the volatile fractions and a lower recovery of the nonvolatile fractions. However, if the pressure is increased to a certain point, the solvent diffusivity may decrease. In addition, there may be less contact with the pores of the raw material, which may result in a reduced solute dissolution [25]. Therefore, high pressure is not recommended for all substances and targeted compounds as it can compress the raw material, which may negatively affect the extraction yield [60]. For example, the pressure should be around 40 MPa in the case of $scCO_2$ [26].

5.3. Co-Solvent

A co-solvent is defined as an organic solvent that can dissolve in an SCF at various ratios and can retain a significant amount of solvent power toward the targeted molecules [61]. For instance, $scCO_2$ is an excellent solvent for the extraction of nonpolar molecules because of its inherent polarity. However, pure CO_2 is not frequently utilized for the extraction of hydrophilic chemicals [58]. To increase the solvating power towards the target molecules, it is common practice in the SFE method to modify the polarity of the SCF by adding small amounts of organic co-solvents. The co-solvents have strong polarity-dependent interactions with the bio-components, such as hydrogen-bonding and dipole–dipole interactions, which significantly enhance extraction yields [25]. The type of sample, targeted molecules, and preliminary experiments should be considered when determining the optimal co-solvent for a particular extraction method [26]. Co-solvents or modifiers can be added to the SFE process using two main procedures: either by mixing the modifier with CO_2 flow or by mixing the modifier with the raw material in the extraction vessel.

The study of phase behavior in binary systems is a starting point for understanding the complexity of the phase behavior. The phase behavior of fluid mixtures under high pressure must be considered to design and enhance supercritical fluid processes. In supercritical fluid systems, pressure and temperature have a complex impact on the phase behavior and can lead to a variety of phase equilibria. Therefore, it is crucial to understand the behavior of fluid mixtures at high pressures within the context of a phase diagram. Studying the phase behavior of pure substances is crucial; however, it does not reveal much about the phase behavior of multicomponent mixtures. Fluid mixtures exhibit a variety of behaviors that are caused by interactions between various molecules and a large range of phase transitions that may occur in this environment [62].

The most commonly used organic solvents are ethanol and methanol [63]. According to the US Food and Drug Administration, ethanol is generally recognized as safe, making it the preferred co-solvent [64]. Although methanol is used in analytical-scale SFE operations, due to its toxicity, it is not used in the preparation of food or oil. Water, acetic acid, and formic acid are often used as SFE co-solvents [65]. The percentage of the co-solvent is significant in the extraction process. For example, the ideal extraction parameters for *Nannochloropsis gaditana* and *Dunaliella salina* were 400 bar and 60 °C, whereas for Synechococcus sp., the best results were obtained at 300 bar and 50 °C. The extraction yields of carotenoids increased with the addition of ethanol (5%) as a co-solvent to supercritical carbon dioxide. When *N. gaditana* was used as the raw material, the ideal pressure and temperature were 500 bar and 60 °C, respectively. The ideal conditions for *Synechococcus* sp. and *D. salina* were 400 bar and 60 °C, respectively, and the results did not clearly indicate a trend. However, using CO_2 + 5% ethanol resulted in low internal diffusion coefficients. This suggests that the mass transfer may have a significant impact on the extraction procedure [66]. The addition of a large volume of co-solvents can change the critical parameters of the fluid and reduce its selectivity [26].

5.4. Extraction Time

Extraction time is an essential variable in the SFE technique because it increases the efficiency of the recovery yield by increasing the contact between the supercritical solvent and the feed material [25]. The extraction time must be considered in the SFE method at both procedural and analytical scales [67] as it may change the extract content. Short extraction times may result in partial extraction. However, if the extraction period is too long, the time and solvent will be wasted, and bioactive chemicals will degrade. In addition, the extraction time is proportional to the flow rate. When, the flow rate is high, the extraction is fast, and the extraction time is short [26]. The total extraction time is calculated in two stages: static and dynamic extraction. For the SFE technique, extraction time is typically less than 2 h [55]. The extraction process was analyzed by considering the total extraction curve (yield vs. extraction time), which provides information on the time needed to obtain an efficient and favorable extraction procedure [25].

5.5. Flow Rate

The flow rate of the SCF affects the selectivity of bioactive compounds and the extraction efficiency [59]. The equilibrium between the fluid and the solid controls the mass transfer process. At the beginning of the extraction process, the recovery of the extract happens more quickly when the flow rate is raised. However, the recovery remains the same at low flow rates at the end of extraction. With an increase in the flow rate, the thickness of the film layer around the solid particles is minimized; thus, the mass transfer resistance surrounding the solid particles decreases, resulting in an increase in the total extraction yield [25]. For example, the CO_2 flow rates range from 1 to 10 L/min in most studies; this is based on the solubility of the components in $scCO_2$. The control of SCF flow rate is controlled using a back pressure regulator (BPR) and gas flow meter [67].

5.6. Raw Matrix

Several factors affect the solubility and mass transfer during the SFE technique, such as the nature of the raw material, particle size, moisture content, shape, surface area, and porosity [68]. The correct selection of these factors can more effectively enhance the complete extraction of targeted compounds [26]. The extraction yield increases as a result of particle size minimization. Grinding before extraction improves the interfacial area and releases solutes by breaking the interior structures of the particles, leading to a higher extraction rate [25]. However, it is imperative to avoid using particles that are too small as they might increase the internal mass transfer resistance. The range of the particle sizes of natural products for SFE is from 0.25 to 2.0 mm [26].

The extractable sample material needs to be dried to lower its moisture content. This is because moisture might compete with the extractable solute for association with the solvent, lowering the extraction yield. However, in some cases, the presence of water is necessary to allow suitable interactions between the solvent and the solute. The recommended moisture content range is 4–14% [69]. In addition, it is critical to understand how various pre-treatment techniques, including air flow drying, freeze-drying, and oven drying, may affect the recovered products [58]. For instance, freeze-dried samples had a significantly higher extraction yield than oven-dried ones [25].

6. Major Advantages and Disadvantages of SFE

SFE has many unique characteristics and is considered a viable alternative to traditional solvent extraction techniques. The advantages of using SCFs for the extraction of bioactive compounds are summarized as follows [70]:

- SCFs are highly diffusible and have relatively low viscosities. Therefore, they have a greater ability than liquid solvents to penetrate porous solid materials, leading to faster extraction;
- Compared with traditional procedures, SFE significantly reduces the amount of time required for extraction, from hours or days to a few minutes (less than 2 h);
- Continuous reflux of supercritical fluid into a sample can provide quantitative or complete extraction.
- SCFs have a better selectivity than liquid solvents because their solvation power can be tuned by changing the temperature and/or pressure.
- This adjustable solvation power of SCFs is helpful for extracting complicated substances, such as plant materials.
- The solute can be easily separated from the solvent using depressurization, which saves time.
- SFE is often performed at low temperatures, making it an excellent approach for studying thermally labile chemicals. This may lead to the identification of novel natural components.
- SFE uses no or significantly less toxic organic solvents and is considered to be environmentally friendly.
- SCFs can be recycled and reused to reduce waste generation.

- SFE may enable direct coupling with chromatographic techniques, which can be an efficient way to extract and immediately quantify extremely volatile compounds.
- For specialized purposes, SFE scales can be set up for small-scale analytical, preparative, pilot plant-scale, and large-scale industrial [70].
- However, there are some drawbacks of SCFs, which are listed as follows [71]:
- The phases of equilibrium between a solvent and a solute can be challenging.
- When co-solvents are used to change the polarity of a fluid, they remain in the extract and require further purification.
- It is challenging to continue adding solids to the raw material owing to the high pressure involved in this process.
- Compared with solvent extraction techniques, less material can be extracted.
- High operational costs.
- Low equipment availability.

7. Comparison between SFE and Traditional Methods

Traditional techniques such as Soxhlet extraction, maceration, and hydrodistillation are widely used to extract bioactive substances, essential oils, and natural pigments from a wide variety of natural sources. The efficiency of traditional extraction methods directly depends on the solubility of the solute and extraction temperature. Although conventional methods are simple, inexpensive, and easy to handle, they require large amounts of harmful organic solvents, long extraction times, and additional operations for solvent removal (Table 3). This lengthy operation of traditional methods may lead to pigment degradation and undesirable chemical extraction due to low purity and selectivity. Traditionally, organic solvents, such as acetone, hexane, chloroform, isopropanol, methanol, methylene chloride, and diethyl ether, have been used to extract pigments. Because most pigments exhibit polar to nonpolar properties, a mixture of solvents such as acetone/water or methanol/water is employed [16,21].

Table 3. Comparison of the SFE technique with traditional methods [70].

Parameter	SFE	Traditional Methods
Solvent	SCF, few amount of harmful organic solvents	Large volumes of harmful organic solvents
Speed	Rapid	Many steps and long processing time
Purity	Highly pure extracts (No solvent residue)	Less pure (Solvent residue)
Recovery	Simple	Need additional operations for solvent removal
Selectivity	Selective	Less selective
Dissolving power	Pressure-tunable dissolving power	Constant dissolving power
Cost	Expensive	Inexpensive

The SFE technique improves extraction by taking advantage of the benefits of supercritical fluids, which have qualities similar to those of gases and liquids. Despite being extremely effective, using little or no toxic solvent, being able to extract heat-sensitive pigments, having a continuous fresh fluid flow, having a quick mass transfer, and being automatable, the SFE method still has high capital and operational expenses owing to the high pressure needed for the process (Table 3). Supercritical CO_2 is a nonpolar solvent with a preference for low-polarity or nonpolar molecules such as carotenoids and chlorophylls. Therefore, its use for polar pigment extraction, such as that of anthocyanins, has some limitations. In this instance, the improvement of its affinity requires the addition of a co-solvent to change the nonpolar nature of supercritical CO_2 [16,21].

8. Extraction of Functional and Antimicrobial Pigments Using Supercritical Fluids

The use of natural colorants has gained wide acceptance worldwide due to their coloring qualities and health-promoting benefits [72]. Colorants derived from natural resources include dyes and pigments. Colorants contain two groups of molecules, chromophores and auxochromes. A chromophore is a conjugated double bond system that is delocalized. Light in the visible range of the electromagnetic spectrum is absorbed by the chromophore due to electron resonance. Auxochromes are color aids that shift wavelengths and control colorant solubility [73]. Based on their hues, origins, biological activities, and chemical structures, natural colorants can be widely classified as shown in Figure 5.

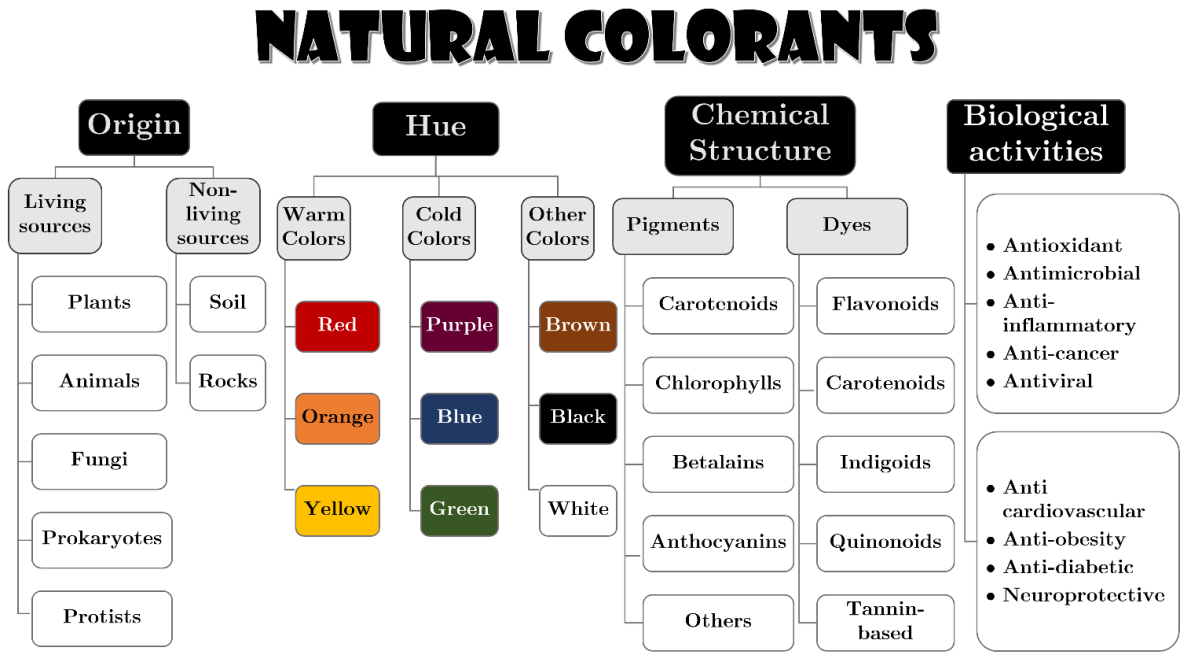


Figure 5. Classification of natural colorants.

The five kingdoms of living organisms can produce natural colorants. Animals, plants, fungi, protists, and prokaryotes are living sources of extractable colorants, while soil and rocks are nonliving sources. Plants, minerals, and animals are the three main sources of natural colorants [74]. Anthocyanins, carotenoids, chlorophylls, betalains, flavonoids, indigoids, quinonoids, tannins, and others can be classified based on their chemical structures [75]. Chemistry-based classification has defined chemical groups with specific characteristics based on the chemical structure [74]. Based on their hue, pigments are classified into warm, cold, and other color categories [76]. Based on their biological activities, natural colorants exhibit antimicrobial, antioxidant, anti-inflammatory, antiviral, anticancer, anticardiovascular, anti-obesity, antidiabetic, disease, and neuroprotective effects [77].

The quantity of colorants present in natural sources is limited. Therefore, a specific extraction procedure is required to remove dye-bearing components from their original sources [74]. The extraction procedure must be specified to achieve high yields and purity while maintaining functional properties such as color and bioactivity. In addition, it is desirable that the extraction procedure be inexpensive, require little energy, and use nontoxic solvents. The SFE technique is a highly effective method to extract natural pigments. The main natural pigments extracted by SFE are carotenoids, chlorophylls, and phycocyanins due to the strong affinity of these nonpolar molecules for $scCO_2$ [75]. Tables 4–7 show studies on functional natural pigments extracted using the SFE method, especially those with antimicrobial activity, as well as the general properties of these pigments.

Table 4. Previous studies on the SFE of carotenoids under the optimal conditions of solvent system, temperature (Temp), pressure (P), time (T), flow rate (FR), particle size (PS), and sample weight (SW).

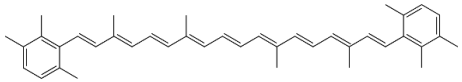
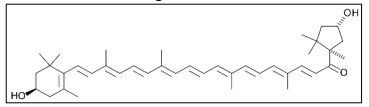
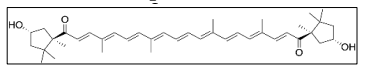
Raw Material	SFE Conditions (Solvent System, Temp, P, T, FR, PS, and SW)		Yield	Biological Activities	Remarks/Results	Ref.
			General structure 			
Mango 	Solvent system	CO_2 + Ethanol (5–15)%	Carotenoids: 1.9 g/kg	Antioxidant Antibacterial	Optimal conditions: 60 °C, 25 MPa, and 15% *w/w* ethanol	[78]
	Temp	40–60 °C				
	P	25–35 MPa				
	T	180 min				
	FR	6.7 g/min				
	SW	5 g				
	Solvent system	CO_2 + 50% Methanol	-		Mango leaf extract was used to impregnate polyester textiles using supercritical CO_2	[79]
	Temp	100 °C				
	P	12 MPa				
	T	180 min				
	FR	10 g/min				
	SW	30 g				
Paprika 	Solvent system	Pure CO_2	-	Antimicrobial	Main pigments: Capsanthin  Capsorubin  Optimal conditions: 65 °C, 40 MPa, 1 mm, and 90 min	[23]
	Temp	35–75 °C				
	P	10–50 MPa				
	T	60–180 min				
	FR	3 L/min				
	PS	0.25–1.25 mm				
	SW	25 g				

Table 4. *Cont.*

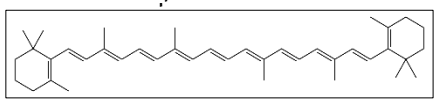
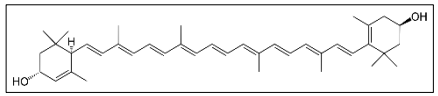
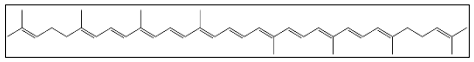
Raw Material	SFE Conditions (Solvent System, Temp, P, T, FR, PS, and SW)		Yield	Biological Activities	Remarks/Results	Ref.
Arthrospira platensis 	Solvent system	CO_2 + 96% Ethanol	β-carotene: 0.52446 g/kg Lutein: 0.00144 g/Kg	Antimicrobial Antioxidant	Main pigments: β-carotene  Lutein  Optimal conditions for β-carotene: 60 °C, 45 MPa, 15 min of static time, and 25 min of dynamic time Optimal conditions for lutein: 60 °C, 45 MPa, 5 min of static time, and 55 min of dynamic time	[80]
	Temp	40, 60 °C				
	P	15, 45 MPa				
	T	Static time: 5, 15 min Dynamic time: 25, 55 min				
	FR	25 g/min				
	PS	1 mm				
	SW	35 g				
Citrus 	Solvent system	Pure CO_2	Carotenoids: 1.952 g/Kg	Antimicrobial Antioxidant [81]	Optimal conditions: 25.196 MPa, 44.88 °C, and 1.91 mixing ratio.	[82]
	Temp	40–50 °C				
	P	20–30 MPa				
	T	120 min				
	FR	27 g/min				
Tomato 	Solvent system	Pure CO_2	Oleoresin yield: 251.15 g/kg (~62% lycopene)	Antioxidant Anti-inflammatory Anticancer	Main pigments: Lycopene  Optimal conditions: 52 °C, 55 MPa, and 180 min	[83]
	Temp	40–80 °C				
	P	20–55 MPa				
	T	120–240 min				
	FR	0.0018 g/mL				
	PS	< 0.20 mm				
	SW	15 g				

Table 4. *Cont.*

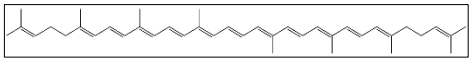
Raw Material	SFE Conditions (Solvent System, Temp, P, T, FR, PS, and SW)		Yield	Biological Activities	Remarks/Results	Ref.
Tomato 	Solvent system	Pure CO_2	Lycopene: 0.729 g/kg β-carotene: 0.016 g/kg	Antioxidant Anti-inflammatory Anticancer	Optimal conditions: 40 MPa, 80 °C, and 4g CO_2/min	[84]
	Temp	50–80 °C				
	P	30–50 MPa				
	T	105 min				
	FR	3–4 g/min				
	PS	0.3–1 mm				
	SW	10 g				
Tomato Watermelon Gac 	Solvent system	Pure CO_2	Lycopene: 63, 52, and 60% from gac, tomato, and watermelon, respectively.	Antioxidant	Main pigments: Lycopene  Dehydrated matrices are suitable for SFE.	[85]
	Temp	60 °C				
	P	35 MPa				
	T	30–180 min				
	FR	4 mL/min				
	SW	25 g				
Carrot 	Solvent system	CO_2 + 5, 10, and 15% Ethanol	Carotenoids: 86.1%.	Antioxidant	Optimal conditions: 59.0 °C, 34.9 MPa, and 15.5% ethanol	[86]
	Temp	50, 60 and 70 °C				
	P	15, 25 and 35 MPa				
	T	80 min				
	FR	15 g/min				
	PS	205 µm				
	SW	5.0 g				

Table 4. *Cont.*

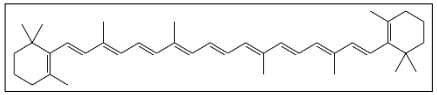
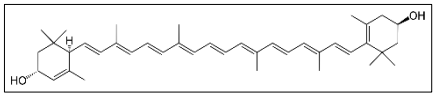
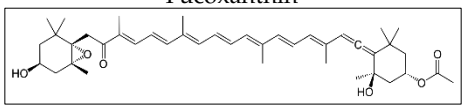
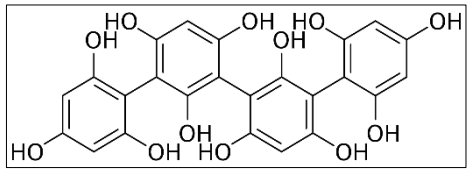
Raw Material	SFE Conditions (Solvent System, Temp, P, T, FR, PS, and SW)		Yield	Biological Activities	Remarks/Results	Ref.
Rowanberry 	Solvent system	Pure CO_2	Carotenoid: 6.630 ± 0.403 g/ Kg β-carotene: 3.295 ± 0.200 g/Kg	Antioxidant	Main pigments: β-carotene  Optimal conditions: 45 MPa, 60 °C and 180 min	[87]
	Temp	40–60 °C				
	P	25–45 MPa				
	T	360 min				
	FR	3.0 mL/min				
	SW	20 g				
Pumpkin 	Solvent system	CO_2 + Ethanol	β-carotene: 0.205 g/Kg	Antioxidant	Optimal conditions: 47.75 °C, 30 MPa and 67% mass of seeds	[88]
	Temp	40–50 °C				
	P	20–30 MPa				
	T	60 min				
	FR	15 L/ h				
	SW	100 g				
Corn gluten meal 	Solvent system	CO_2 + 5–15% ethanol	Lutein: 85.4 × 10^{-6} g	-	Main pigments: Lutein  Optimal conditions: 40 °C, 47.02 MPa, and 15% ethanol	[89]
	Temp	40–80 °C				
	P	37.92–51.71MPa				
	T	60–480 min				
	FR	2 mL/min				
	SW	2.5 g				

Table 4. *Cont.*

Raw Material	SFE Conditions (Solvent System, Temp, P, T, FR, PS, and SW)		Yield	Biological Activities	Remarks/Results	Ref.
Brown Seaweed 	Solvent system	CO_2 + (Ethanol/soybean oil /canola oil/ sunflower oil)	Fucoxanthin: 1.421 g/Kg. Phlorotannin: 0.927 g/Kg.	<u>Fucoxanthin:</u> Anti-inflammatory Antioxidant Anticancer <u>Phlorotannins:</u> Antioxidant Antibacterial Anti-inflammatory Anti-allergic.	Main pigments: Fucoxanthin  Phlorotannin  Optimal conditions for fucoxanthin: 50.62 °C, 30 MPa, and 2.00% sunflower oil as a co-solvent Optimal conditions for phlorotannins: 48.98 °C, 30 MPa, and 2.00% with water	[90]
	Temp	45–55 °C				
	P	20–30 MPa				
	T	120 min				
	FR	27 g/min				
	SW	100 g				

8.1. Carotenoids Extraction

Carotenoids are red, orange, and yellow pigments. The basic structure of carotenoids is linear and symmetrical, with approximately 40 carbon atoms. One or two cyclic structures are present at the ends of the conjugated chains. The extensively conjugated double bond system acts as a chromophore that absorbs light and produces red, orange, and yellow colors. In addition, the conjugated system gives the substance distinct molecular shapes and chemical reactivity [90]. The main types of carotenoids present in nature are hydrocarbon carotenoids (called carotene) and oxygen-containing xanthophylls. Lycopene, β-carotene, and α-carotene are examples of carotenes. Lutein, β-cryptoxanthin, and zeaxanthin are examples of xanthophylls [91]. Their conjugated double bond structure can confer high reactivity against oxidative stress, providing several bioactivities such as anti-obesity, antidiabetic, and anticarcinogenic affects, as well as cardiovascular and neuroprotective effects [75]. Owing to their high biological activities, carotenoids can be used in many applications such as food textiles, pharmaceuticals, and cosmetics.

The SFE technique was used to extract carotenoids from several raw materials as shown in Table 4. For example, Sánchez-Camargo et al. reported the results for carotenoids extracted from mango peels using SFE that have a high natural antioxidant activity. The co-solvent (ethanol) concentration was the most significant factor in the SFE optimization. The extraction temperature was also shown to have a favorable effect, whereas an increase in the pressure had a negative effect. The optimal conditions were found to be 60 °C, 25.0 MPa, and 15% w/w ethanol [78]. Furthermore, polyester textiles were impregnated with mango leaf extract as a source for carotenoids using $scCO_2$, which enhanced its antioxidant and antibacterial properties. In addition, the polar co-solvent and higher temperature (100 °C) improved the extraction yield of the phenolic compounds. However, the pressure did not significantly affect the extraction procedure [79]. These findings demonstrate the superiority of the SFE technique over traditional solvent extraction in terms of its selectivity for compounds containing carotenoids. Shah et al. investigated the optimization of the extraction of paprika oil and the extraction of capsaicin and pigments (capsanthin and capsorubin) from paprika powder using SFE. Unlike the mango peel case, pressure had a significant effect on yield, but other factors such as temperature, time, and particle size had no significant effect. The maximum pigment yield was obtained at 65 °C, 40 MPa, and 90 min. The extract of chili (paprika) exhibited antimicrobial activity because of the presence of multiple bioactive compounds, such as phytochemicals, terpenes, and capsaicin. Paprika extract showed antimicrobial activity against *Escherichia coli*, *Bacillus subtilus*, and *Staphylococcus aureus* [23]. Compared with traditional methods (organic solvents), supercritical carbon dioxide was more selective than organic solvents (n-hexane). Supercritical oleoresin with the highest pigment content was obtained at 40 MPa (14,134 mg/kg), which represents 44.9% of the yield obtained using the conventional method [92]. In another study, the effects of SFE parameters on the biological compounds present in *Arthrospira platensis* extracts were studied. *Arthrospira platensis* is a blue-green cyanobacterium that is extensively found in tropical and subtropical waters. The temperature, pressure, co-solvent, and static and dynamic times were assessed. The optimal conditions for β-carotene production were 60 °C, 45 MPa, 15 min of static time, and 25 min of dynamic time. The optimal conditions for lutein production were 60 °C, 45 MPa, static time of 55 min, and dynamic time of 5 min. In addition, the co-solvent was the most significant variable for all assessed impacts. However, the β-carotene concentration in the present study was lower than that previously reported for *Arthrospira platensis* [93]. Furthermore, the β-carotene content was higher than that obtained in the extraction without the co-solvent, which confirmed the significance of the co-solvent factor in the SFE of β-carotene. The SFE results in this study for 30 min were equivalent to the extraction in that study, which required 100 min. In addition, these extracts showed antimicrobial activity against *S. aureus, Pseudomonas aeruginosa, E. coli,* and *Candida albicans*. *Arthrospira platensis* is considered to be a sustainable source of functional extracts using SFE [80]. In a subsequent study [83], lycopene pigment extraction from tomato using the SFE technique was optimized. The optimal conditions for

cis-lycopene were 52 °C, 55 MPa, and 180 min. Lycopene is an important compound that has health-promoting properties such as antioxidant, anti-inflammatory and anticancer components. Additionally, Kehili et al. studied the SFE of lycopene and β-carotene from tomatoes, which was enhanced by increasing the temperature, pressure, and CO_2 flow rate. The maximum carotenoid (lycopene and β-carotene) recovery was obtained at 40 MPa, 80 °C, and 4 g CO_2/min [84]. In addition, the SFE of lycopene from the tomato peel was compared with that of traditional maceration extraction using hexane, ethyl acetate, and ethanol. The maximal SFE yield of lycopene reported in this study, 0.729 g/kg of dried tomato peels, is equivalent to or even greater than that reported in most previous studies on lycopene extraction from tomatoes using supercritical CO_2 and solvents. Moreover, a comparative study on the shades of red (lycopene-rich oleoresins) extracted from tomato, gac, and watermelon fruit was conducted using the SFE technique. According to the findings, all oleoresins provide a safe source of lycopene, with the added value of exhibiting significant lipophilic antioxidant activity that is enhanced by interactions with other biomolecules [85].

In addition, the effects of various SFE factors on carotenoid extraction from carrot peels have also been investigated. The most important factor that influenced carotenoid recovery was pressure. The temperature exhibited two different behaviors. Increasing the temperature had a positive effect on the extraction, but very high temperatures led to carotenoid degradation and isomerization. The maximum yield for carotenoid recovery was achieved at 59.0 °C and 34.9 MPa [86]. Bobinaitė et al. investigated the extraction of bioactive substances such as carotenoids from rowanberry pomace using consecutive extraction with $scCO_2$ and pressurized solvents. The consecutive pressurized solvent extraction of pomace residue using SFE recovered a polyphenol-rich extract with a strong antioxidant capacity. Furthermore, it was observed that pressure was the most significant factor influencing carotenoid recovery [87]. In Soxhlet extraction, the amount of total carotenoids and β-carotene recovered was 78.91 ± 3.50 mg/100 g and 38.69 ± 1.43 mg/100 g, respectively. Depending on the optimal SFE conditions for maximal extraction yield, the total carotenoid content in the extracts varied from 512.4 to 1913.9 mg/100 g and β-carotene content from 282.4 to 976.8 mg/100 g, whereas the total carotenoid and β-carotene recovery as compared with Soxhlet extraction constituted 49.7% and 52.5%, respectively. In addition, carotenoids can be extracted from citrus fruit using SFE. Carotenoid content was maximized by changing the pressure, temperature, and mixing ratio of the plant material [82]. Citrus fruit extracts have antioxidant and antimicrobial properties [81]. Wang et al. studied the SFE of carotenoids from pumpkin. The results indicated that the extraction of oil, α-tocopherol, and β-carotene was considerably affected by the mass ratio of pumpkin flesh to seeds. Optimal conditions were obtained at 47.75 °C, 30 MPa, and a 67% mass of seeds [88]. Xanthophyll extraction using SCFs has also been studied. Process factors were studied to determine the optimal lutein extraction from corn gluten meal. Lutein extraction was optimized at 40 °C, 47.02 MPa, and 15% ethanol [89]. Saravana et al. extracted fucoxanthin and phlorotannin from brown seaweed using $scCO_2$. Fucoxanthin is a xanthophyll carotenoid with outstanding properties. Fucoxanthins and phlorotannins exhibit antibacterial, anti-inflammatory, antioxidant, anti-allergic, and anticancer properties. Increasing the pressure, temperature, and co-solvent ratio significantly improved the yield of fucoxanthin and phlorotannin. The optimal conditions for fucoxanthin were 50.62 °C, 30 MPa, and 2.00% sunflower oil as a co-solvent, whereas for phlorotannins they were 48.98 °C, 30 MPa, and 2.00% water [90].

The SFE technique is a suitable green method for extracting carotenoids because of their hydrophobic nature. The nonpolar nature of carotenoids is consistent with that of CO_2. Therefore, it is an ideal solvent in this category. According to previous studies, the optimal conditions for this class are a temperature range of 40–65 °C and pressure of 25–55 MPa. To improve the extraction yields of carotenoids, it is suggested that carbon dioxide be combined with an organic solvent (such as methanol or ethanol) in small amounts (up to 10%).

Table 5. Previous studies on the SFE of chlorophylls under the optimal conditions of solvent system, temperature (Temp), pressure (P), time (T), flow rate (FR), particle size (PS), and sample weight (SW).

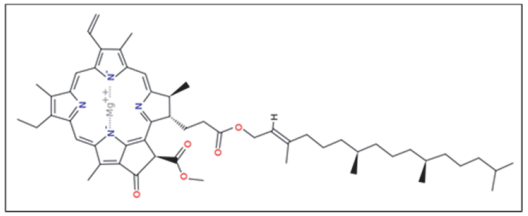
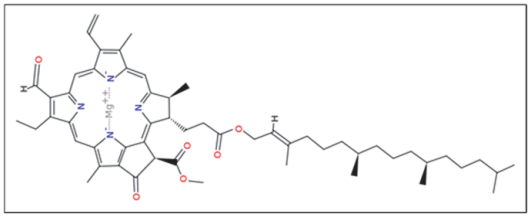
Raw Material	SFE conditions (Solvent System, Temp, P, T, FR, PS, and SW)		Yield	Biological Activities	Remarks/Results	Ref.
General structure						
						
	Chlorophyll (a)			Chlorophyll (b)		
Olive 	Solvent system	CO_2/Ethanol	-	Antioxidant Antimicrobial	Main pigments: Chlorophyll (a) Chlorophylls (b) Carotenoids	[94]
	Temp	60 °C				
	P	35 MPa				
	T	13 min				
	FR	10.00 mL/min				
Elaeagnus angustifolia 	Solvent system	Pure CO_2 for leaves	Carotenoids: 0.0183 g/Kg Chlorophyll a: 0.0438 g/Kg chlorophyll b 0.001 g/Kg	Antimicrobial	Main pigments: Chlorophylls Carotenoids	[95]
	Temp	55 °C				
	P	15 MPa				
	T	60 min				
	SW	7 g				
Hop cone 	Solvent system	Pure CO_2	-	Antimicrobial	Main pigments: Chlorophylls (a, b)	[96]
	Temp	50 °C				
	P	30 MPa				
Rosemary 	Solvent system	Pure CO_2 + 3%, 10% (EtOH: Water 50/50 *v/v*), or 30% (EtOH)	The yields of Carotenoids: 53 g/Kg Chlorophyll a: 100 g/Kg Chlorophyll b: 100 g/Kg	Antioxidant Antibacterial Antifungal	Main pigments: Chlorophylls Carotenoids Optimal conditions for carotenoids: Pure CO_2, 25 °C, and 20 MPa Optimal conditions for chlorophylls: 30% of ethanol as co-solvent.	[97]
	Temp	25 °C				
	P	20/10 MPa				
	T	20/40/50/30 min				
	FR	3.0 mL/min				
	SW	1 g				
	Solvent system	Pure CO_2	3.52%		Optimal conditions: 30 MPa and 50 °C	[98]
	Temp	30, 40, and 50 °C				
	P	10, 20 and 30 MPa				
	T	240 min				
	FR	0.2 kg/h				

Table 5. *Cont.*

Raw Material	SFE conditions (Solvent System, Temp, P, T, FR, PS, and SW)		Yield	Biological Activities	Remarks/Results	Ref.
Dunaliella salina 	Solvent system	Pure CO_2	Carotenoids: 0.115 g/Kg Chlorophylls: 0.033 g/Kg	Antioxidant	Main pigments: Chlorophylls Carotenoids Optimal conditions: 40 MPa and 55 °C	[99]
	Temp	40–60 °C				
	P	10–50 MPa				
	T	180 min				
	FR	3 L/min				
	SW	5.0 g				
	Solvent system	CO_2 + 5% (Ethanol/Hexane/ Acetone/ Methanol)	β-carotene: 25 g (for CO_2 + ethanol), 6 g (for pure CO_2)		Ethanol was the best co-solvent	[100]
	Temp	35, 45, and 55 °C				
	P	20 and 30 MPa				
	T	105 min				
	PS	<0.355 mm				
	SW	10 g				
Spinach 	Solvent system	CO_2 + 0, 5, and 10% Ethanol	72% lutein 50% chlorophylls	Anti-inflammatory Antioxidant	Main pigments: Chlorophylls Lutein Optimal conditions: 56 °C, 3.6 h, and 39 MPa, with 10% ethanol	[101]
	Temp	40, 50, 60 °C				
	P	10, 30, 50 MPa				
	T	60,120,180 min				
	FR	10 g/min				
	SW	25 g				
Chlorella sorokiniana 	Solvent system	CO_2 + Ethanol (0–10%)	-	Anti-obesity Antioxidant	Main pigments: Chlorophylls (chlorophyll a and chlorophyll b) Carotenoids Optimal conditions: 50.1 °C, 20.29 MPa, and 4.5% ethanol	[102]
	Temp	40–60 °C				
	P	10–30 MPa				
	T	180 min				
	FR	1 kg/h				
	SW	55 g				

Table 6. Previous studies on the SFE of anthocyanins under the optimal conditions of solvent system, temperature (Temp), pressure (P), time (T), flow rate (FR), particle size (PS), and sample weight (SW).

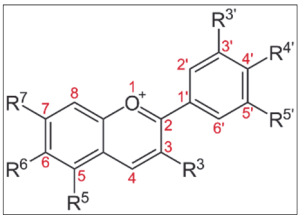
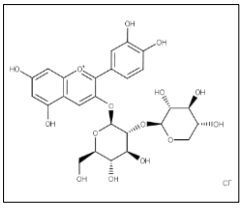
Raw Material	SFE Conditions (Solvent System, Temp, P, T, FR, PS, and SW)		Yield	Biological Activities	Remarks/Results	Ref.
			General structure 			
Roselle 	Solvent system	CO_2 + (5, 7.5, and 10)% Ethanol	Anthocyanins: 26.73%.	Antioxidant Wound healing Anticholesterol Antihypertensive	Optimal conditions: 8.90 MPa, 70 °C, and 9.49% ethanol	[103]
	Temp	50, 60, and 70				
	P	8, 10, and 12 MPa				
	T	70 min				
	FR	6 mL/min				
	PS	355 µm				
	SW	1.5 g				
	Solvent system	CO_2 + 5–10% Ethanol or Water	Anthocyanin: 11.97 g/Kg		Main pigments: Cyanidin 3-sambubioside  Optimal conditions: 27 MPa, 58 °C, and co-solvent ratio of 8.86%	[104]
	Temp	40–70 °C				
	P	10–30 MPa				
	T	120 min				
	FR	4 mL/min				
	PS	<0.355 mm				
	SW	1.5 g				
Juçara 	Solvent system	CO_2 + 10% acidified mixture of ethanol and water	Anthocyanins: 22 g/Kg	Antioxidant	SFE was more selective for anthocyanins.	[105]
	Temp	60 °C				
	P	20 MPa				
	T	46 min				
	FR	2.08×10^{-4} kg/s				
	SW	2.5 g				

Table 6. *Cont.*

Raw Material	SFE Conditions (Solvent System, Temp, P, T, FR, PS, and SW)		Yield	Biological Activities	Remarks/Results	Ref.
Chokeberry 	Solvent system	CO_2 + Ethanol (20, 50, and 80%)	Phenolic compounds: 15.2 g/Kg (Anthocyanins accounted for 50–67% of the total phenolics)	Antioxidant	Optimal conditions: 35 °C, 10 MPa, and 80% m/m ethanol addition Anthocyanins are impacted by the use of an acidified co-solvent.	[106]
	Temp	35, 50, and 65 °C				
	P	7.5,10, and 12.5 MPa				
	T	75 min				
	FR	1.8 g/min				
	SW	10 g				
Haskap berry 	Solvent system	CO_2 + Water	Anthocyanins: 52.7%	Antioxidant Anti-inflammatory Antitumor	Optimal conditions: 65 °C, 45 MPa, 15 min of static time, and 20 min of dynamic time	[107]
	Temp	35, 55 and 65 °C				
	P	10, 27.5, and 45 MPa				
	T	Static time:15, 60, and 120 min Dynamic time:0, 20, and 60 min				
	FR	10 mL/min				

Table 7. Previous studies on the SFE of other pigments under the optimal conditions of solvent system, temperature (Temp), pressure (P), time (T), flow rate (FR), particle size (PS), and sample weight (SW).

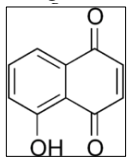
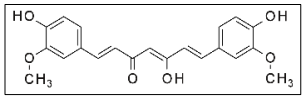
Raw Material	SFE Conditions (Solvent System, Temp, P, T, FR, PS, and SW)		Yield	Biological Activities	Remarks/Results	Ref.
Quinones						
Walnut green husk 	Solvent system	CO_2 + Ethanol	Juglone: 0.03726 g	Antibacterial Antifungal Antioxidant Antitumor	Main pigments: Juglone  Optimal conditions: 35 °C, 15 MPa, and 375 μm	[108]
	Temp	35, 40, 45 and 50 °C				
	P	9, 11, 13 and 15 MPa				
	T	Static time: 20 min				
	PS	375, 605, 855 and 1500 μm				
	SW	12 g				
	Solvent system	CO_2 + Ethanol	Juglone: 11.92 g/Kg	Antifungal Antioxidant	-	[109]
	Temp	50 °C				
	P	30 MPa				
	T	195 min				
	FR	10 mL/min				
	PS	≤1 mm				
Curcuminoids						
Turmeric 	Solvent system	CO_2 + Water or Ethanol	Curcumin: 23.4%	Antimalarial	Main pigments: Curcumin  The supercritical extract had a low curcumin content but significant antimalarial activity.	[110]
	Temp	40 °C				
	P	40 MPa				
	T	360 min				
	FR	4×10^{-2} g/s				
	PS	0.823 mm				

Table 7. *Cont.*

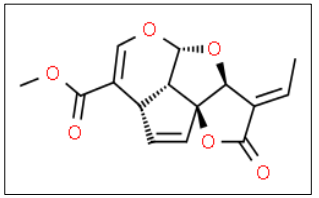
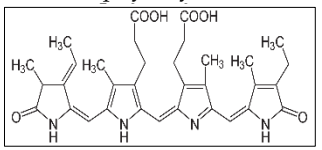
Raw Material	SFE Conditions (Solvent System, Temp, P, T, FR, PS, and SW)		Yield	Biological Activities	Remarks/Results	Ref.
Turmeric 	Solvent system	Pure CO_2	31 g/Kg	Antimicrobial Antifungal	Main pigments: Turmerones Optimal conditions: 30 MPa and 40 °C	[111]
	Temp	40 °C				
	P	9–66 MPa				
	FR	1.8 g/min				
	SW	60 g				
Iridoids						
***Momordica charantia* Vine** 	Solvent system	CO_2 + ethyl acetate or ethanol	-	Antibacterial	Main pigments: Plumericin  Optimal conditions: 50 °C, 25 MPa, and 5 L/min in the presence of ethyl acetate and ethanol as co-solvents. Plumericin exhibited antibacterial activity against 8 harmful bacterial strains, especially *Enterococcus faecalis* and *B. subtilis*.	[112]
	Temp	50 °C				
	P	25 MPa				
	T	180 min				
	FR	5 L/min for CO_2 0.003 L/min for co-solvent				
	SW	3000 g				

Table 7. *Cont.*

Raw Material	SFE Conditions (Solvent System, Temp, P, T, FR, PS, and SW)		Yield	Biological Activities	Remarks/Results	Ref.
Phycocyanins						
Spirulina maxima 	Solvent system	Pure CO_2	-	Antioxidant Anti-inflammatory Antiviral, Anticancer Cholesterol-lowering	Main pigments: C-phycocyanin  Chlorophylls Carotenoids Optimal conditions: 60 °C, 24.13 MPa	[113]
	Temp	40, 50, and 60 °C				
	P	24.13, 31.03, and 37.92 MPa				
	T	60,90, and 120 min Static time: 30,45, and 60 min Dynamic time: 30,45, and 60 min				
Black sesame pigment						
Sesame dregs 	Solvent system	Pure CO_2	Black sesame pigment: 3.58%	Antioxidant	Optimal conditions: 30 °C, 4 MPa, and 10.80 L/min	[114]
	Temp	20, 30, and 40 °C				
	P	10,14, and 18 MPa				
	FR	0.48, 0.80, and 1.12 L/min				

8.2. Chlorophylls Extraction Using SCFs

Plant leaves are mainly colored by chlorophyll. Chlorophylls are green, amphiphilic, oil-soluble pigments that are widely distributed in plants [10]. Their molecules contain cyclic pyrroles made up of four subunits formed from pyrroles, which are often complexed with a metal ion (e.g., Mg, Zn, and Cu) [75]. Chlorophyll is found in two types: chlorophyll a and chlorophyll b [10]. Chlorophylls are easily degraded and change color because of their great sensitivity to heat, light, oxygen, acids, and enzymes [16]. These colorants have anti-obesity, anti-diabetic, anti-cancer, and chronic disease prevention properties [75].

Many researchers have been interested in studying the optimal conditions during the SFE technique for various raw plant materials, as shown in Table 5. SFE was used to extract carotenoids and chlorophylls from *Dunaliella salina* by Hosseini et al. Pressure was found to have a greater impact on the extraction yield than temperature. The optimal conditions for carotenoids and chlorophylls were 40 MPa and 55°C. A carotenoid/chlorophyll ratio of 11.09 was achieved at 30 MPa and 30 °C, demonstrating the SFE technique's selectivity. The best extraction yield of carotenoids was obtained at 40 MPa and 55 °C (115.43 g/g dry microalgae), which was the most suitable operating setting. With supercritical extraction, the yield of carotenoids was only half (47%) of that obtained with solvent extraction. The selectivity of this method was demonstrated by the highest carotenoid/chlorophyll ratio (11.09) obtained at 30 MPa and 30 °C. Under these operating conditions, the two extracted pigments could be separated and purified more easily, and higher selectivity was attained [99,100].

Additionally, Derrien et al. extracted lutein and chlorophyll from spinach using SFE. The extraction process factors were then optimized. The co-solvent had a greater impact on chlorophyll content than lutein. Temperature did not have a significant effect on lutein and chlorophyll contents. Higher pressure resulted in a higher extraction yield of lutein and chlorophyll, but a decrease in the selectivity of the solvent. The best extraction conditions were 56 °C, 3.6 h of extraction time, 39 MPa of pressure, and ethanol concentration of 10%. Under these conditions, a yield of 50% chlorophyll and 72% lutein was obtained [101]. The SFE chlorophyll yield (recovery of 50% of the total chlorophyll) was lower than that of conventional green extraction (using water and ethanol as extraction solvents) under optimal conditions (96%). Chlorophylls and carotenoids from *chlorella sorokiniana* were also extracted using $scCO_2$, with ethanol as a co-solvent. Ethanol is essential for the efficient extraction of chlorophylls and modification of the polarity of supercritical solvents for carotenoid extraction. The temperature, pressure, and amount of ethanol also had statistically significant effects on the extraction process. The extract may exhibit antioxidant activity owing to its chlorophyll and carotenoid content [102]. In another study, carotenoids and chlorophylls were extracted from olive pomace using SFE; the extract exhibited antioxidant and antimicrobial effects owing to the presence of β-sitosterol. The highest yields of carotenoids and chlorophylls were obtained from freeze-dried samples, which efficiently preserve phytocompounds [94].

Carradori et al. showed that SFE extracts from the fruit and leaves of *Elaeagnus angustifolia* exhibited antimicrobial effects against *Salmonella typhimurium* and *E. coli*. These findings support the hypothesis that leaf extracts, which have a higher phenolic content and pigments such as chlorophylls than fruit extracts, have greater antioxidant activity [95]. According to Wasilewski et al., the hydrophobic extracts of hop cone obtained via SFE are suitable for use in all-purpose cleaners because of their antimicrobial activity, minimal irritating potential, and foam-like appearance [96]. In addition, selective sequential SFE was applied to extract bioactive compounds such as carnosic acid, rosmarinic acid, and pigments from rosemary. SFE optimization was performed using supercritical fluid extraction coupled with a supercritical fluid chromatography system. Consecutive conditions and sequential extraction were used to obtain various fractions that are rich in bioactive chemicals or pigments. Chlorophyll was extracted using 30% ethanol as the co-solvent [97]. Genena et al. confirmed that SFE extracts from rosemary leaves exhibited antioxidant,

antibacterial, and antifungal activities. In addition, the highest total SFE yield was obtained at 30 MPa and 50 °C.

Chlorophyll is easily degraded. Owing to singlet oxygen, chlorophyll content is reduced under light more quickly than in darkness. Therefore, chlorophylls can be effectively extracted using SFE owing to its controlled conditions. The SFE of chlorophylls can be performed within a few minutes at low temperatures in a closed system with high purity, which preserves the chlorophylls from degradation. The highest SFE yields of chlorophyll were achieved at an extraction temperature of 40–60 °C, pressure of 10–40 MPa, and time of 1–5 h. Most SFE processing of chlorophylls has been performed in pure CO_2. In addition, the solubility of chlorophylls can be improved by using a co-solvent with a limited percentage (up to 10%).

8.3. SFE of Anthocyanins

Anthocyanins are water-soluble pigments present in fruits and vegetables. They are red, blue, and purple colors. Their structures consist of two aromatic rings connected by an oxygen-containing, three-carbon heterocyclic ring. The conjugated double bonds of the anthocyanidin are considered the chromophore. Various factors have an impact on the color and stability of anthocyanin such as temperature, pH, light, oxygen, enzymes, and metallic ions [115]. Anthocyanins exhibit antioxidant, antimicrobial, anti-inflammatory, anticancer, antidiabetic, anti-obesity, and cardioprotective effects [75].

Anthocyanin pigments can be also extracted from different natural sources using SFCs under specific conditions, especially Temp and P, as shown in Table 6. For instance, roselle was extracted using $scCO_2$ and ethanol (as a co-solvent). The main anthocyanins extracted from roselle were dephinidin 3-sambubioside, cyanidin 3-sambubioside, delphinidin 3-glucoside, and cyanidin 3-glucoside. The findings proved that the total extraction yield was significantly influenced by all three main factors investigated. The optimal operating conditions for the red pigment were a pressure of 8.90 MPa, a temperature of 70 °C, and a co-solvent ratio of 9.49% [103]. Subsequently, Idham et al. investigated improvements in the extraction and stability of anthocyanins. It was found that increasing the co-solvent ratio resulted in an increase in anthocyanin yield, which gradually decreased at a co-solvent ratio of 10%. Increasing pressure increased the anthocyanin content; however, at higher pressures, the anthocyanin content decreased. Temperature did not significantly affect the anthocyanin content within the specified range (40–70 °C). The investigated conditions using $scCO_2$ treatment were more effective in protecting anthocyanins and maintaining color stability at refrigerated, room, or ambient temperatures [104].

Anthocyanins were also extracted using the SFE technique from juçara residues via $scCO_2$ and an acidified co-solvent, which increased the polarity of $scCO_2$ and positively affected the anthocyanin yield. The use of an acidified co-solvent increases the polarity of $scCO_2$, which positively affects the SFE of anthocyanins. The SFE was proven to be more selective for anthocyanins [105]. Woźniak et al. confirmed the significant effect of co-solvent in anthocyanins extraction from chokeberry using SFE, which makes $scCO_2$ more polar. However, the amount of organic solvent required for extraction was significantly reduced by using $scCO_2$. The effect of the operating parameters on the extraction yields was investigated, and the optimal conditions were found to be 35 °C, 10 MPa, and 80% m/m ethanol ratio [106]. Meanwhile, $scCO_2$ and water were used as co-solvents (green solvents) and increased the effectiveness of anthocyanins in the SFE of haskap berry pulp. Extraction factors of SFE including pressure, temperature, and amount of water were optimized. The highest yield of anthocyanins was achieved at 65 °C, 45 MPa pressure, 15 min static time, and 20 min dynamic time. It was observed that freeze-drying pretreatment was not necessary before the SFE processing. When $scCO_2$ and water were used, the anthocyanin extraction efficiency increased to 52.7%, and the antioxidant activity improved to 89.8%. The use of $scCO_2$ and water as co-solvents provided better anthocyanin extraction efficiency (52.7% vs. 38.3%) and increased antioxidant activity compared with the traditional extraction (89.8% vs. 72.2%) [107].

Unlike carotenoids and chlorophylls, the SFE technique is not highly appropriate for the extraction of highly polar pigments, such as betalains and anthocyanins. The use of various co-solvents can modify the polarity of the extraction process. According to previous reports, the use of a co-solvent is necessary in large amounts (up to 20%). The common co-solvents used for the SFE of anthocyanins are ethanol and water. Other parameters can be adjusted at a temperature of 35–70 °C, pressure of 7.5–45 MPa, time of 70–120 min, and high flow rate.

8.4. Quinones Extraction Using the SFE Technique

Quinones are chemical substances with a completely conjugated cyclic dione structure ($C_6H_4O_2$), which possess a sufficient conjugation to produce color [116]. Natural quinonoids are present in flowering plants, bacteria, fungi, sea urchins, lichens, and some insects [73]. Quinonoids can be classified into three basic types, benzoquinones, naphthoquinones, and anthraquinones, and into other quinonoids such as ubiquinones, plantoquinones and menaquinones [73]. The color spectrum of quinonoids is broad, ranging from yellow to red. Quinonoids are used as antimicrobial finishes for textiles [73].

Juglone was extracted from green walnut husks using SFE. The results demonstrated that pressure was the most significant parameter. Increasing the pressure improved the extraction efficiency, whereas temperature and particle size had a negative effect on the value of the extracted juglone. The highest juglone yields were obtained at 35 °C, 15 MPa, and 375 µm. In addition, the juglone dye exhibited antibacterial, antifungal, antioxidant, and antitumor properties. It can be used as a dye in natural and synthetic fabrics, skin-coloring preparations, hair dyes, and medicine [108]. Another study showed that the extract obtained using $scCO_2$ was rich in bioactive compounds, such as phenolic acids and juglone, and exhibited substantial antioxidant and antifungal activities. SFE with ethanol showed an extractive yield (27.18 g/100 g) comparable to that obtained with solvent (ethanol and methanol) extractions (36.13 and 39.27 g/100 g). This is due to the use of high pressures and the intermediate properties of CO_2 exhibited in the supercritical state (Table 7) [109].

8.5. Curcuminoids Extraction Using SCFs

Curcuminoids are water-insoluble compounds that are naturally present in turmeric. Two benzenemethoxy rings are linked by an unsaturated chain to produce the curcumins. The color of curcuminoids is yellow-orange. The bioactive features of this phenolic class include anti-inflammatory, antimutagenic, anti-Alzheimer, anticancer, antimicrobial, neuroprotective, and cardioprotective actions, in addition to helping to reduce obesity [75]. The SFE of curcumin from the rhizomes of *Curcuma longa* has been previously studied. The two step process ($scCO_2$ extraction, followed by ethanol or water extraction) resulted in the highest total yield when water was used. However, high concentrations of curcumin were obtained in the two step process when using ethanol as the solvent. The SFE had a low curcumin content but significant antimalarial activity [110]. Topiar et al. extracted turmerones from turmeric using SFE. The optimal conditions for the SFE technique were 30 MPa and 40 °C. Turmerones exhibited antimicrobial and antifungal effects owing to their high biological activity (Table 7) [111].

8.6. Iridoids Extraction via SCFs

Iridoids are water-soluble blue pigments. The bioactive properties of this class include antioxidant, anti-inflammatory, antimicrobial and anticancer properties [75]. Using SFE, Saengsai et al. extracted plumericin (an iridoid pigment) from *momordica charantia vine* (Table 7). The extraction parameters were optimized at 50 °C, 25 MPa, and 5 L/min in the presence of ethyl acetate and ethanol as co-solvents. The yield of the Soxhlet method was 8.55 g/100 g, while the SFE yield was 1.24 g/100 g. Plumericin exhibited antibacterial activity against eight harmful bacterial strains, especially *E. faecalis* and *B. subtilis*, with minimum inhibition concentration values compared with cloxacillin [112]. This pigment can be used in possible applications such as medicine, cosmetics, and pharmacology.

8.7. SFE of Phycocyanins

Phycocyanins are blue, water-soluble, fluorescent pigments produced by cyanobacteria and eukaryotic algae. Because of their sensitivity to heat and light, they have fewer potential uses in the food and pharmaceutical industries. They have antioxidant, anti-inflammatory, antiviral, anticancer, and cholesterol-lowering effects [75]. C-phycocyanin was extracted from *Sagina maxima* using SFE (Table 7). *Sagina maxima* is a biomass of blue-green algae that may be consumed by both humans and animals. Various conditions of SFE were studied to optimize the extraction process. The optimal $scCO_2$ pre-treatment conditions were 60 °C and 24.13 MPa. Due to C-phycocyanin, it was expected that the SFE extract would have several health benefits, such as antioxidant, anti-inflammatory, antiviral, anticancer, and cholesterol-lowering effects [113].

8.8. SFE of Black Sesame Pigment

Black sesame pigment (BSP) was extracted from sesame dregs using the SFE technique. It was found that pressure had a significant effect on its yield, and the maximum yield was obtained under the conditions of 30 °C, 4 MPa, and 10.80 L/min [114]. Under optimal conditions, the BSP yield produced using SFE was 3.58 ± 0.08%. Compared with synthetic black pigments, this pigment is considered safe and exhibits antioxidant activity. Therefore, BSPs can be used in medicine, pharmacology, food, cosmetics, and other fields.

9. Antimicrobial Effect of Natural Pigments

Among the natural compounds, natural colorants have demonstrated beneficial advantages, such as antioxidant, antibacterial, and antifungal activities. Although synthetic antimicrobial materials have received approval in many countries, natural compounds produced by microorganisms, animals, and plants have attracted the interest of many researchers. Natural colorants have antimicrobial properties owing to the presence of active biomolecules called phytochemicals. Phytochemicals and their manner of action can differ among sources. The mechanism by which natural colorants provide antimicrobial activity can be summarized as follows [117,118]:

- Coagulation of cytoplasmic contents
- Prevention of enzyme generation
- Inactivation of the function of the outer membrane
- Fluctuation of the proton engine force of the cells
- Interaction with extracellular proteins
- Alteration of cytoplasmic membrane
- Blockage of metabolic pathway

10. Future Trends

There are many aspects that require more research with respect to antimicrobial natural colorants, and the current challenge for researchers is to recover natural pigments from various sources, such as food industry byproducts, using environmentally friendly processes (for example, SFE) coupled with environmentally friendly solvents (e.g., water, CO_2, and ethanol) to achieve low cost and toxicity. Limited resources can affect the availability of natural plant dyes. Therefore, marine natural products have attracted the attention of scientists because marine microorganisms are considered as potential resources for new natural high-yield dyes with very broad application prospects. In this regard, researchers are attempting to discover and confirm the safety of new natural pigments; however, the regulatory clearance of these items is expensive and requires a long time. Hence, only a few naturally occurring colorants that are commercially available have been approved by the FDA for use in foods and beverages, such as curcumin and phycocyanin. As aforementioned, the limited stability and low solubility of natural colors in the application medium restricts their use in the food and pharmaceutical industries. An alternative is to use $scCO_2$-based formulation processes, which increases the water solubility of bioactive pigments and their bioavailability and absorption in the body. Additionally,

research has been conducted on co-pigments and encapsulation techniques to enhance their hyperchromic effect, stability, solubility, and bioavailability. Co-pigments have been utilized to improve color and stability because they create noncovalent complexes with different pigments, especially for anthocyanins. In the textile industry, antimicrobial fabrics dyed and finished with natural pigments are suitable for people with eczema skin allergies. However, the application of naturally colored antimicrobial-finished fabrics for the application of wound healing has yet to be studied, and further research is needed.

11. Conclusions

According to previous reports, the SFE technique is a practical substitute for traditional solvent-based extraction methods that add value by recovering high-quality health-promoting pigments in a green manner. SFE technologies have been used to develop distinctive natural antimicrobial colorants, which have led to the development of creative ideas for the widest possible use of these materials. Several dyes and pigment families ranging from the most hydrophobic chlorophylls and carotenoids to the most polar anthraquinones have been successfully extracted using SFE. Owing to its pressure-tunable dissolving power, simple recovery, high purity, high speed, nontoxicity, and absence of solvents, it is recognized as a green technology for extracting natural antimicrobial colorants from various sources. Each category of natural colorants (anthocyanins, carotenoids, chlorophylls, and others) has special SFE conditions, including Temp, P, T, FR, SW, Ps, and co-solvent ratio and type. The optimal conditions for the SFE of antimicrobial colorants increase the use of these pigments and control the factors that affect their stability. Regarding carotenoids, SFE is an excellent extraction method due to the chemical's hydrophobic nature. SFE preserves chlorophylls from degradation and oxidation because of the moderate and controlled extraction conditions. Despite the hydrophilic nature of anthocyanins, they can be extracted using SFE with some modifications in polarity using a co-solvent. Natural colorants extracted using SFE are considered safe ingredients in many industries, particularly those intended for human consumption, such as new functional food additives and dyes used in textiles and pharmaceuticals.

Author Contributions: Conceptualization and supervision, T.A.E.; writing—original draft preparation, M.M.; writing—review and editing, K.S.-A. and H.E. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Conflicts of Interest: The authors declare no conflict of interest.

Abbreviations

BPR	Back pressure regulator
P_C	Critical pressure
T_C	Critical temperature
FR	Flow rate
PS	Particle size
SW	Sample weight
$scCO_2$	Supercritical carbon dioxide
SCF	Supercritical fluid
SCFs	Supercritical fluids
SFE	Supercritical fluid extraction

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