 ISSN 2278-6783

Review Article

Green Extraction Technology and Its Application in Food Industry

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Abstract

The use of natural compounds in the food industry as well as the recovery of antioxidants, pigments, polymer and oils from agro-industrial waste have become increasingly popular. Conventional extraction methods involve large amounts of solvents, applied heat, time-consuming, decomposition of thermosensitive compound and security risks, such as toxicity of solvents and the presence of solvent residues in the extracts together with low yield, have stimulated the development of green technologies. Green extraction techniques also known as clean extraction techniques is a new concept of extraction of natural products from food, plants and agro-industrial waste to meet the challenges of the 21st century to protect both the environment and meet consumers demand and in the meantime enhance competition of industries to be more ecologic, economic and innovative. Green extraction techniques involve Ultrasound-Assisted Extraction (UAE), Microwave-Assisted Extraction (MAE), Pulsed Electric Field (PEF) Assisted Extraction, Enzymatic Extraction, Supercritical fluid Extraction and use of 'green' solvents are environmentally friendly, require less time, energy and allows the production of chemical free compounds than conventional extraction which are recognized as safe and are preferred by consumers. These extraction methods employed under controlled temperature or without the involvement of heat advantageous for extraction of thermolabile compounds preventing them from degradation. High capital investment, high running cost, complex configuration, training, maintenance cost are limitations to large scale application of green extraction techniques. Future research should address the cost minimization, integration of the novel technologies, using agri-food by-products and high value-added products, such as food additives to meet consumers increasing demand for cleaner labels and large-scale application of these techniques.

Keywords: Antioxidant, Extraction, Green solvent, Thermolabile compound, Toxicity

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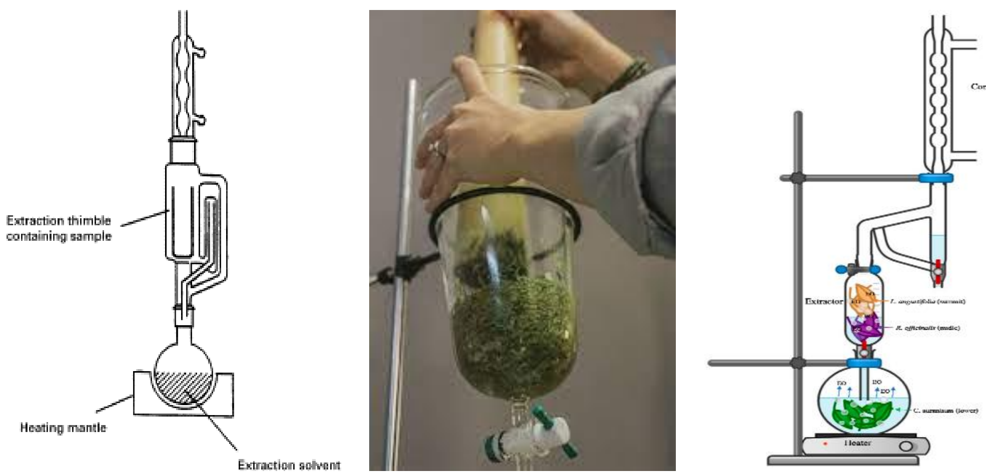
Introduction

Extraction can be considered as a stepping stone in analytical procedures and subsequent product development. Extraction of natural product like perfume, medicine or foods has been done since time immemorial by Arabs, Indians, Chinese, Greeks and Romans by conventional methods. Security risks, such as toxicity of solvents, solvent residues in the extracts and low yield of conventional methods have stimulated the development of other extraction technologies, such as clean or green technologies. Green extraction technology also called clean technology are the new methods for sustainable extraction for various compounds, which will reduce energy consumption, allows use of alternative solvents and ensure a safe and high-quality extract/product [1]. These extraction methods are employed under controlled temperature or without the involvement of heat hence advantageous for extraction of thermolabile compounds preventing them from degradation. The food industry is heavily involved in searching for green sources of valuable compounds to meet to the evolving consumers requirements for health-beneficial food ingredients [2]. Recent trends in extraction techniques have largely focused on finding solutions that minimize the use of solvents. This, of course, must be achieved while also enabling process intensification and a cost-effective production of high-quality extracts. The development of green technology has markedly influenced the recovery of natural compounds intended for the food industry. Innovation in extraction techniques has been fueled by green chemistry, as well as, the growing public interest for chemical free compounds. Such novel extraction methods include supercritical fluid extraction, microwave assisted extraction, ultrasound assisted extraction, enzyme assisted extraction, amongst others. Green technology coupled with the use of green solvents, such as, deep eutectic solvents, and ionic solvents, is regarded as a viable alternative for the recovery natural compound and also prevents the formation of toxic effluents. Apart from the environmentally friendly dimensions of green extraction techniques, the health and safety components are also taken into consideration. Novel extraction techniques are not without their own limitations. As such, the capital investment and running cost associated to these cutting technologies might be major challenges to their implementation and application [3]. Therefore, this review focused on low environmental impact technologies and green solvents towards the green extraction by discussing the main associated advantages and disadvantages, and the criteria of selection for process sustainability.

Conventional Extraction Technology

Conventional extraction techniques are considered classical methods of extraction and basic principle of these types of techniques is solvent extraction and an applied heat [4]. Some conventional extraction methods include Soxhlet extraction, hydro distillation, maceration, or solvent extraction, **Figure 1**. The solvent selection is critical in the extraction process and alcohols (methanol and ethanol) are the commonly used solvents for extraction in conventional methods as well as chlorinated solvents, such as chloroform, carbon tetrachloride, and chlorobenzene, and non-chlorinated solvents, such as acetone, and acetonitrile are also employed. The solubility, cost and security issues should be considered while selection of the solvent [5], a solvent with low boiling point, low toxicity and quick mass transfer capability can be a good option. Temperature also plays an important role as high temperatures enhance the diffusion and solubility, they can cause solvents loss, leading to possibly decomposition of thermosensitive compounds due to impurities formation [6]. The conventional methods have long processing times of extraction. However, increasing the time does not influence the extraction until the solvent balance is achieved within and outside the solid matrix [4].



Soxhlet extraction Maceration Hydrodistillation

Figure 1 Conventional extraction technology.

Soxhlet extraction

In Soxhlet extraction a small dry sample is put into a thimble and set in a distillation flask containing the desired solvents (such as ethanol, methanol, chloroform, acetone, acetonitrile, or hexane). Once an overflow level is reached, the thimble-holder containing solvent-solute mixture is suctioned by a siphon, empties the solution into the distillation flask. This solvent carries the separated extracts into the bulk liquid and solute (extracts) remains in the distillation flask and the solvent goes back into the plant tissue matrix. This procedure continues until the extraction is finished. Extraction time can be up to 48 hours and temperature ranges depend on the solvents used [4, 6, 7-9]. Is the basic model technique, commonly used for essential oils, lipids, fats and phenolics extraction and best suited for small scale industries. Disadvantages are requiring large quantities of solvents, time consuming, low efficiency and not suitable for heat-sensitive ingredients [6, 8, 10].

Maceration

Maceration based on the leaching of compounds from solid material, was traditionally used for the recovery of bioactive compounds from plant materials [11]. Maceration uses solvents in combination with heat and/or agitation to improve mass transfer and solubility of compounds [3]. The sample is grinded into small particles, this increases the surface area, facilitating the proper mixing with the solvent, the solvent is added in a closed vessel (water, organic solvents, or their combination). The liquid is discarded, and then the solid residue is squeezed to recover the prepared solution. Finally, the prepared solution is separated from impurities with a filtration process. Agitation is used in this process to facilitate the extraction. This action promotes the diffusion and removes the concentrated solution from the sample surface and brings new solvent for increase the extraction yield [6, 8, 9, 12]. It can be used in small scale extractions of essential oils and phenolics and is inexpensive method. However, its disadvantages are that it is time consuming and require large quantities of solvents [6, 10].

Hydro-distillation

The sample is packed in a still compartment, water is added and then boiled. At the same time, direct stream is injected into the sample, then a condenser cools the vapor mixture containing water and the compound of interest. The condensed mixture goes to a separator, to separate the compound from the water, the hot water and the steam act as the extractor factors. The physicochemical processes involved are hydro diffusion, hydrolysis, and decomposition by heat. This process should be performed before drying of sample. [6, 9, 10]. It is simplest and oldest technique and has different classifications: steam, water, or hydro-diffusion distillation, can be used to extract essential oils and phenolics. It can be used in small scale industries. and can uses water to facilitate the extraction. While disadvantages include not suitable for heat-sensitive ingredients, time consuming, consume high energy levels [6, 12].

Green Extraction Technologies

To overcome the disadvantages of conventional extraction techniques, there are other extraction methods called clean or green extraction techniques that exhibit shorter extraction times, high efficiency, and selectivity, as well as reduce the use of solvents techniques are [13]. The name “green extraction” is due to the less energy consumption, renewable natural products, the reduction of hazardous substances, and less time in the extraction process [1, 6]. These methods of extraction are the new trends for obtaining the bioactive compound from many plant sources and by-products and high value-added products, such as food additives to meet consumers increasing demand for cleaner labels. These techniques include microwave-assisted extraction (MAE), enzyme-assisted extraction (EAE), ultrasound assisted extraction (UAE), supercritical fluid extraction (SFE), and pulsed electric field extraction (PEFE) **Figure 2**.

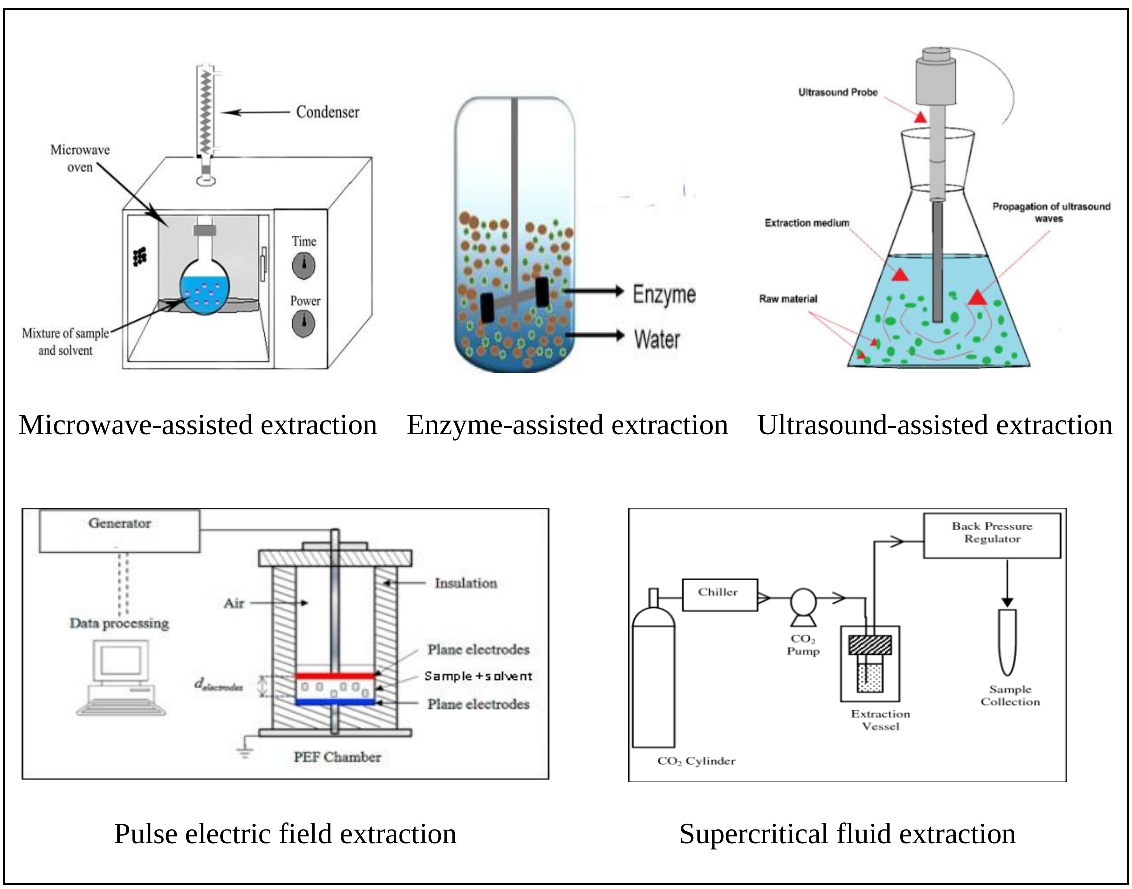


Figure 2 Green extraction technology

Green extraction techniques follow a series of principles and the Six Principles of Green Extraction involves the innovation in the selection of renewable plant resources such as fruit and vegetable wastes, use of alternative solvents (principally water or agro-solvents), reduce energy consumption by energy recovery and using innovative technologies, production of co-products, reduction of unit operations and aim for a non-denatured and biodegradable extract without contaminants. Most of the organic solvents are flammable, volatile and even toxic, use of alternative solvents is a good option to replace the petrochemical solvents and **Table 1** shows some examples of alternative solvents that can be used in green extraction [1, 13]. The novel techniques of extraction can also be used as a pretreatment or combined with alternative solvents to enhance the extraction and the extraction yield will depend on several factors, such as the process design, solvent selection, and type of matrix.

Table 1 Alternative solvents for green extraction techniques

Solvent	Description	Extraction technique	Application	Solvent Power	Cost	Environmental Impact
Solvent Free	-		Antioxidants, glucosinolates, essential oils, and pigments	Polar	Low	Medium
Water	It has polar nature for natural-water soluble compounds	MAE, UAE, and SFE	Essential oils and aromas	Polar	Low	Low
CO2	Non-flammable odorless gas produced during the burning of fossil fuels, by alcoholic fermentation	SFE	Phenolics	Weakly polar and non-polar	Low	Low
Ethanol	Obtained from the fermentation of sugar-rich materials (sugar beet and cereals)	MAE and SFE	Pigments and antioxidants	Weakly polar	Medium	Low
Glycerol	By-product from the trans-esterification of vegetable oils.		Polyphenols, oils, and fats	Weakly polar	Low	Low
n-Hexane	Petrochemical solvent.	MAE and SFE	Fats and oils	Non-polar	High	High

The recovery of antioxidants, polymer, pigments and oils from agro-industrial waste as well as use of natural compounds in the food industry have become popular day by day. The phytochemical rich extracts from food plants are also increasingly gaining acceptance owing to their nutraceutical, nutritional and functional applications. There are many natural compounds that can be obtained from natural sources like fruits, vegetable, plants etc. that can be used as food and feed additives as well as cosmetics ingredients. A great number of natural compounds with potential applications as preservatives to act either antioxidants, antimicrobials or to provide specific flavor, aroma or color to foods. To obtained these green extraction technologies are employed due to its advantages over conventional methods such as less time, minimize use of solvents and extraction yield improvements [14]. These extraction methods employed under controlled temperature or without the involvement of heat advantageous for extraction of thermolabile compounds preventing them from degradation.

Microwave – assisted extraction (MAE)

Microwaves provide dielectric heating and solute dissolution with electromagnetic fields in the range between 300 MHz to 300 GHz. In microwave-assisted extraction the solvent penetrates into the plant matrix, the components break down with the aid of electromagnetic waves and then the solubilized components are transferred from the insoluble matrix to the bulk solution, finishing with the liquid and residual solid phase separation [15]. The combination of mass gradients and heat is responsible for the acceleration and high extraction yield in Microwave – assisted extraction [16]. The frequencies that are commonly used in microwave ovens (for domestic uses and laboratories) are 0.915 GHz and 2.45 GHz [9]. Several bioactive compounds such as phenolics, carotenoids, and flavonoids can be obtained through microwave – assisted extraction method, [12]. MAE can be performed with or without the use of solvent, ethanol in combination with water due to its good compound solubilization and good capacity to absorb microwave energy is most commonly used. The factors such as the choice of microwave power, the temperature of extraction, time, and amount of solvent affect the MAE [15, 12]. MAE can be classified into two systems, closed and open systems, open MAE system is most commonly used in bioactive compound extraction due to its higher sample throughput, solvent addition and atmospheric system conditions [17]. The heat transfer from microwave radiation allows the moisture in the cell to evaporate, resulting in an increase in the pressure within the plant matrix and splits the cell membranes. This enables the penetration of the solvent to expel the bioactive compounds, dissipating the heat volumetrically within the irradiated medium [16]. There are several cases that prove the efficiency for MAE, for example, a study for isolating hesperidin from citrus skin using ethanol as solvent at 1 kW, 2.45 GHz of power with 140 ℃ for 8 min, reported an 86.8% (47.7 mg/g) of hesperidin yield, almost 10% more efficient than conventional extraction [18]. It is reported that mango peel has 1.5 to 6 times more phytochemicals and antioxidant power after 15 min of the MAE, compared to conventional solvent extraction (10 h), and required less extraction time [19]. 2.92% yield of polysaccharides from kiwi at 480W, 80℃ for 120min using 1:10 ethanol (80%) from kiwi [20], 7.1% of essential oil at 600W, 60℃ for 50min using water 20:1 from oregano [21] and 42.4% yield of Phenolics at 850W,164℃ for12,5 min using ethanol(45%) from *Hibiscus sabdariffa* [22]. Advantages includes short extraction time, enhanced extraction yield, low solvent consumption low power consumption, easy industrial escalation and possibility to develop a solvent-free process. However, high equipment cost and uneven heating or overheating of sample may reduce extraction efficiency or cause thermal degradation of phenolic acids are some of disadvantages [23, 24, 25].

Ultrasound – assisted extraction (UAE)

The ultrasound – assisted extraction also known as sonication, uses a sound wave from 20 kHz to 100 MHz, which travels through a medium and creates compression and expansion, producing a cavitation phenomenon [10]. The cavitation process involves the growth and collapse of tiny bubbles, the bubbles when exceed a critical diameter they break down inducing a high amount of energy that converts the kinetic motion into heat. The materials that have a cavitation effect are liquid and liquid-containing solids [6]. The physical phenomena of UAE include the diffusion through the cell wall and after the cell wall breakdown rising of the cell material. The time of sonication, moisture, particle size, solvent, temperature, and pressure are the main factors to consider for an effective extraction and frequencies between 20 and 100 kHz, and 80 to 200 W of power are used for bioactive compounds extraction [16]. The ultrasound can be applied directly with higher intensities or with indirect methods such as an ultrasonic water bath to the medium until the waves enter the sample [12]. A study that shows a 2.3-to-3-fold increase in total phenolic content from grape pomace in the temperature range of 20 to 50 C for 2.5 min using UAE at 55 kHz, water as a solvent, and 22.9 W/cm2 compared to conventional extraction [26]. Further, 279.9 mgGAE/g yield of phenolics at 20kHz, 110W/cm^3,48℃ for 49min using ethanol from *Malva sylvestris* leaves [27], 53.78 mg AA/100g yield of vitamin C at 20kHz, 400W/cm^3, 21℃ for 30min using ethanol from orange peels [28], carotenoids from pomegranate waste at 20kHz,130W/ cm^3, for 30 min using sunflower and soy oil as solvent yield 93.8% [29] and 23% polysaccharides yield from *Silvetia compressa* using ethanol at 3.8 W/ cm^3, 50℃ [30]. The combination of extraction techniques can be used to enhance the yield of extraction viz., Supercritical Fluid Extraction of 50 ℃ with 25 mPa of pressure followed by ultrasound assisted extraction with 400 W of power, using ethanol and water as a co-solvent lead to a global phenolic extraction yield of 9.87% twice the performance of only using Supercritical Fluid Extraction for blackberry bagasse [31]. Another study shows that a 97.4% extraction yield of lycopene can be obtained with microwaves (98 W) using ethyl acetate as a solvent from tomato paste delivered to an ultrasonic bath operating at 40 kHz and 50 W with 365s of extraction time and a temperature of 86.4 ℃ in comparison an 89.4% lycopene yield can be obtained by only using UAE [32]. These results show that a combined extraction method could be more efficient for obtaining bioactive compounds. UAE have several advantages such as the reduction in time, low energy and power usage, improved extraction yield and a good-quality extract, less solvent, low operating temperature, less thermal degradation and efficient extraction of thermolabile compounds [33-35].

Pulsed electric field extraction (PEFE)

The pulsed electric field extraction (PEFE) is a non-thermal process in which a living cell suspension is subjected to an electric field results in splitting the molecules in the cell membrane due to an electrical potential pass across the cell membrane. The charged molecules create pores in the weakest areas of the membrane once the transmembrane potential of 1 V is reached, increasing the membrane permeability and causing electroporation [6, 8]. The extraction yield depends on various parameters, such as the energy input, number of pulses, the field strength, temperature and plant matrix. It is estimated that 500 and 1000 V/cm of the electric field during 104 to 102 s can break the cell membrane for the releasing of bioactive compounds, with little temperature increment and also the degradation of thermolabile components can reduce. PEFE is mostly used as a preextraction treatment and can be combined with other extraction techniques to enhance the final extraction yield [6]. The PEFE of anthocyanins yield 50% higher than conventional extraction methods from grape skin at 3 kV/cm of pulsed electric fields showed a high selectivity after 1 h of extraction [36]. Another study shows that PEFE can be applied to obtain a 39% yield of carotenoid from tomato peel with a mixture of hexane, acetone, and ethanol (50:25:25) as solvent, using 5 kV/cm of intensity during 300ms [37]. 90% yield of pigments from red beetroot using 1 kV/cm, 270 pulses for 10 µs [38], anthocyanins from orange peel using 3.4kV/cm, 105 pulses for 3µs yields 65.8mg/g [39] and 1.85mg/g of alkaloids yields from potato peel at 0.75 kV/cm, 200pulse for 3 µs [40]. Some phenolic acids, such as chlorogenic, ferulic, and salicylic acids are found in apple pomace under a PEFE treatment obtaining a total phenolic yield content 37% higher than using conventional extraction technique, with optimal conditions of 2 kV/cm of electric field intensity, 500 ms of extraction time, and 12.5% w/v solid to water ratio [41]. Usually, PEFE operates at room temperature for less than 1 s [42] however, in certain situations the application of electric fields at room temperature is not adequate, so pulsed ohmic heating may be used through ionic motions in the series to raise the temperature [12]. PEFE enhances the bioactive compounds extraction from food and vegetable wastes as it uses less solvent, less time consuming and lower temperatures of extraction compared with conventional extraction techniques [43, 44]. Energetically efficient, selective, non-thermal, low operation cost, easy scaling up, low energy consumption, waste-free process, continuous operability, non-destructive, no thermal effect, continuous operability, these advantages make PEFE a good method for industrial application. Disadvantage are high cost, depend on medium composition (conductivity) and high cost of the equipment [45, 46].

Enzyme – assisted extraction (EAE)

The enzyme-assisted extraction (EAE) can be employed as a extraction method or pre-extraction in which the plant cell wall is destructed and the bounded bioactive compounds attached to the carbohydrate and lipid chains are released. This process is carried under the action of some enzymes such as a-amylase, cellulase, hemicellulose, pectin esterase, fructosyltransferase, pectinase and protease in the solvent extraction. Enzyme – assisted extraction due to the natural origin of enzymes and water instead of using hazardous solvents considered under the green extraction techniques category [10]. EAE is employed when the plant matrix compounds are not accessible to remove using a solvent in a traditional extraction process due to preserved by hydrogen or hydrophobic bonding in the polysaccharide-lignin network. The moisture content, the particle size of the material, chemical composition of the plant matrix, type and dosage of the enzyme, solvent amount, time, and temperature are the factors that affect enzyme-assisted extraction [6]. This technique has been studied for the extraction of several bioactive compounds from food and vegetable wastes, such as the phenolic extraction (18–20 mg/g) using pectinase from grape seeds [47] and phenolic extraction (0.152 mg/g) from apple pomace, using PectinexVR [48]. Lipids from raspberry using enzyme protease result 38% yield [49], polysaccharide from garlic using enzyme cellulase yields 35.3% [50] and lycopene from tomato using pectinase yields 1.1mg/g [51]. Enzyme-assisted extraction can be combined with other extraction techniques to enhance extraction yield for example, a combination of enzyme-assisted extraction and Supercritical fluid extraction can separate almost twice the total phenolic content from pomegranate peel than conventional methods, the process begins with a pretreatment with pectinase, protease, cellullase, and viscozyme, followed by supercritical CO_2, using ethanol as solvent [52]. Advantages are high selectivity; improved yield; environmentally friendly while disadvantage are enzymes are expensive; rigorous control of medium pH and temperature for optimal enzyme action [3, 53].

Supercritical fluid extraction (SFE)

The supercritical condition takes place when a material is exposed to a temperature and pressure above the critical limit. In this state, the fluid acquires gas/liquid properties of density, diffusion and viscosity, these properties make the extraction of the bioactive compounds possible in a short time. In supercritical fluid extraction (SFE) the analyte is distributed in two different stages: the separation and stationary phase [5]. SFE is a process in which an oven contains a mobile phase (normally CO2, ethane, propane, butane, water, or pentane), which is pumped until it pressurizes the gas, then a vessel with co-solvent is also pumped to the extraction vessel, working at high pressure [6, 15, 44]. Carbon dioxide is often used solvent for SFE, having a critical temperature and pressure of 31℃ and 74 bars, it offers stable working conditions for pressures between 100 to 450 bar however, the extraction is limited to mostly non-polar compounds due to CO_2 low polarity. To overcome this limitation a chemical modifier such as ethanol, water, methanol, and acetone can be added to enhance the polarity [15, 12]. The study report that bioactive compounds can be extracted using this technique with high extraction yields viz., 79% of catechin yield extraction can be obtained from grape seeds using CO2 and methanol as a modifier (40%) [54] and 61% of lycopene yield (7.19

mg/g) at 86 C, 34.47 MPa, 500 mL of CO2 at 2.5 mL/min, from tomato seeds and skin [55], 29.4 mg/g of vitamin E at 361 bar, 83℃ from *Spirulina* [56], 213mg/g of lycopene from tomato at 300 bar, 60℃ from tomato [57] and 60.57mg/g of flavonoids from mint at 200 bar, 60℃ [58]. Advantages includes increased selectivity, enhances mass transfer and yield, supercritical carbon dioxide can be re-used, fast extraction, no filtration required, no use of toxic solvents and allowing the recovery of thermolabile compounds at low temperature. Disadvantage includes high equipment cost, complex configuration of the system, high capital investment, required training and poor selectivity towards polar compounds due to low polarity of supercritical carbon dioxide [3, 23, 59, 60, 61].

Conclusion and Future prospects

Green extraction technology represents a promising tool than conventional extraction to recover high value compounds from natural sources and agro-industrial waste. Bioactive compounds extracted from natural sources have beneficial effects on human health and its applications in the food industry have become increasingly popular. Green extraction techniques are sustainable option for the extraction of natural compounds as they require less time, energy, and solvent. Moreover, the use of ‘green solvents such as water and even without any solvent in some cases allows the production of chemical free compounds which are recognized as safe and are preferred by consumers. The green techniques have better-quality extract, higher yields of extraction, less extraction time, reduce the use of solvents and a sustainable option for the extraction of bioactive compounds. This process are novel, environmentally-friendly and can be adopted for producing natural bioactive pigments for potential food industry applications. Conventional techniques generally require large amounts of organic solvents, high energy expenditure, and are time consuming, it has been proven that the replacement of conventional techniques by green technologies is promising. However, complex configuration, training, high capital investment, running and maintenance cost are limitations to large scale application of green extraction techniques. Integration of the novel technologies, minimization of investment costs, using agri-food by-products as cost-effective sources of natural compounds and high value-added products, such as food additives, should be address in future research contributing to meet consumers’ increasing demand for cleaner labels and for large scale application. Furthermore, the combination of more than one green extraction technique can possibly improve these processes and has better performance results in terms of yield of extraction and extract purity.

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Publication History

Received	19.02.2023
Revised	02.05.2023
Accepted	04.05.2023
Online	31.05.2023