

Processing Technologies for the Extraction of Value-Added Bioactive Compounds from Tea

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Abstract

Tea (*Camellia sinensis*) is the most widely consumed beverage in the world, with an excellent source of bioactive compounds such as catechins, caffeine, and epigallocatechin. There is an increasing trend to extract these bioactive compounds to deliver them as value-added products. Generally, the extraction of polyphenols and other functional compounds from different parts of tea is carried out using different solvents (e.g., water, water–ethanol, ethanol, methanol, acetone, ethyl acetate, and acetonitrile). The extraction efficiency of functional compounds from tea depends on the type and polarity of the solvent as well as the applied process. Several conventional techniques, such as boiling, heating, Soxhlet, and cold extraction, are used to extract bioactive ingredients. However, these procedures are unsuitable for achieving high yields and biological activities due to the long extraction times of cold brewing and the high temperatures in other heating methods. Many efforts have been carried out in food and pharmaceutical industries to replace conventional extraction techniques with innovative technologies (e.g., microwave (MAE), ultrasonic (UAE), pressurized liquid (PLE), pulsed electric field (PEF), and supercritical fluid (SFE)), which are fast, safe, energy-saving, and can present eco-friendly characteristics. These innovative extraction techniques have proven to improve the recovery rate of phenolic-based antioxidant compounds from tea and increase their extraction efficiency. In this review, the application of novel processing technologies for the extraction of value-added compounds from tea leaves is reviewed. The advantages and drawbacks of using these technologies are also highlighted.

Keywords Tea (*Camellia sinensis*) · Phenolic compounds · Bioactivity · Extraction · Ultrasound · Supercritical CO_2 · Pulsed electric field · Microwave · Pressurized liquid

Introduction

Tea (*Camellia sinensis* L.) is an ancient crop belonging to the Theaceae family. Although this evergreen plant originates from southeastern China, it has been widely distributed in countries with tropical and subtropical climate changes

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worldwide [1–3]. China's tea spread to India, Japan, Russia, and Europe [4, 5]. According to [6], tea production was 5.79 million tons, of which China and India contributed 72%. Depending on how the fresh leaves are processed, there are various kinds of tea (e.g., green, white, black, yellow, oolong, Pu'er, or Pu-erh) [7]. Two varieties of tea, green and black, are extensively consumed worldwide [8, 9]. There are thousands of chemical constituents in tea, where the concentration present can be substantially affected by the different heredity (e.g., genetic strain), environmental factors (e.g., weather, soil, irrigation method, growth altitude, and harvest season), horticultural practices, and processing technologies and conditions [1, 10, 11].

Recently, there has been an increasing trend toward extracting bioactive compounds from tea to produce valueadded products such as health supplements. Cold brewing of tea has also gained popularity due to increased consumer acceptance. In general, the extraction of polyphenols and other functional compounds from tea is carried out using a variety of solvents. Although water as a traditional solvent has been applied in most of the studies to extract polyphenols from green and black teas [12–16], the use of other solvents such as water–ethanol [17], ethanol [18, 19], methanol [20, 21], acetone [15, 22], ethyl acetate [23], and acetonitrile [15] has also been reported.

The extraction efficiency of bioactives without any chemical modification is not only a function of the type and polarity of the solvent used but is also influenced by the applied processing techniques for extraction [24]. Processing conditions such as tea: solvent ratio, particle size, agitation rate, and time/temperature have a significant effect on the extraction of bioactive compounds from tea [25]. Conventional techniques like boiling, heating, and reflux distillation are used for the extraction of bioactive ingredients from tea [26]. However, these conventional procedures are unsuitable for achieving high yields and biological activities due to the long extraction times and the high temperatures used [27].

Many efforts have been carried out to promote the use of innovative technologies such as microwave, ultrasound, pressurized liquid, pulsed electric field, and supercritical fluid with fast, safe, energy-saving, and eco-friendly characteristics that replace conventional solvent extraction methods (e.g., heat reflux) in food and pharmaceutical industries [26–35]. Applying novel technologies under mild processing conditions can decrease the impurity and structural changes of polyphenols sensitive to epimerization and oxidative oligomerization reactions and significantly enhance the extraction yield. In addition, these technologies also increase the solvent permeability rate in plant cells and the mass transfer coefficient of the target secondary metabolites [30]. Furthermore, there is a serious concern about tea by-products getting accumulated in the environment. Therefore, using economic and environmental approaches to reuse such agricultural and food processing waste seems necessary for the food industry.

The application of innovative extraction and separation systems such as nanofiltration membranes [2, 3], supercritical carbon dioxide [2, 36], microwaves [37, 38], ultrasound [39, 40], and pressurized liquid [3, 41] have been reported to significantly enhance the extraction of bioactive compounds from tea for the fortification of different foods such as bakery products [42]. Furthermore, studies have shown that tea can be a suitable substrate to produce activated carbon using combinations of chemical activation and microwave energy [43], as well as microwave and infrared energies [44]. Moreover, the discoloration process of dye wastewater by pulsed discharge plasma combined with charcoal derived from tea has been previously reported [45]. This paper provides a comprehensive summary of the literature published on the application of innovative processing technologies like ultrasound, microwave, pulsed electric field, pressurized liquid, and supercritical fluid to extract and recover bioactive compounds from tea.

Tea: Nutrition and Health Properties

Tea, the most popular and oldest non-alcoholic beverage, has a unique flavor with some health benefits [46]. The global average consumption of this healthy functional drink is about 120 mL per day per person, while this value for Great Britain's inhabitants is 4.5 times higher (≈540 mL/ day) [47]. Tea primarily contains polyphenols, and catechins and theaflavins are tea's primary and secondary polyphenols. Through the enzymatic browning by polyphenol oxidase, catechins present in tea leaves are converted to theaflavins during fermentation [48-50]. There are two optical isomers for each geometrical isomer of catechin (trans-catechins and cis-epicatechins), including (+,-)catechin and (+, -)-epicatechin. Esterification of (-)catechin with gallic acid (GA) can lead to the synthesis of (-)-gallocatechin-3-gallate (GCG), (-)-catechin-3-gallate (CG), epicatechin-3-gallate (ECG), and (-)-epigallocatechin-3-gallate (EGCG) from tea leaves [51, 52]. Moreover, four different kinds of theaflavins, namely theaflavin (TF), theaflavin-3-gallate (TF3G), theaflavin-3'-gallate (TF3'G), and theaflavin-3,3'-digallate (TF33'G), can be formed with the polymerization through oxidative coupling [53, 54]. The presence of other flavonoids (e.g., quercetin), alkaloids (theophylline, theobromine, and caffeine), long-chain aliphatic alcohols (e.g., policosanols), amino acids (e.g., glutamic acid, aspartic acid, and theanine), and minerals (e.g., fluorine, chlorine, calcium, and manganese) in various tea products has been demonstrated [5, 55].

The daily drinking of tea can significantly reduce the incidence rate of cancer types such as skin [56-59], breast [2, 3, 60–63], ovarian [64, 65], prostate [66, 67], lung [68, 69], oral [70, 71], colon [72, 73], stomach [70, 74], and pancreatic [75-77] induced by the consumption of alcohol and tobacco. The presence of polyphenols such as EGCG can notably inhibit the activation of carcinogens and, consequently, cancer initiation due to its antiradical and antioxidant activities and its implication in the detoxification system activation. The robust mechanism associated with the modulation in membrane organization, the formation of intercellular interactions with some functional macromolecules (e.g., proteins and nucleic acids), the epigenetic alteration, and the regulation of cellular replicative potential can highly limit the progress of carcinogenesis. This mechanism is accomplished by preventing the self-renewal, proliferation, and viability of the predominant tumor-initiating clones and, thus, the consequent growth [78]. Earlier, the effects of anti-mutagenic, anti-diabetic, anti-inflammation, antibacterial, anti-viral, anti-arthritis, anti-obesity, and neuroprotective of tea polyphenols have been comprehensively

reported by other researchers [49, 79–88]. Additionally, the immune response by these bioactives was recently identified against severe acute respiratory syndrome coronavirus type 2 (SARS-CoV-2) [49, 89].

Several bioactive compounds, such as polyphenols and caffeine, can be extracted using conventional and novel extraction systems [23, 90]. The tea dust generally contains 2.5% decaffeinated tea, serving as a value-added source to extract bioactive compounds such as theanine [42]. This amino acid has many health benefits, such as relaxing and anti-tumor effects, learning capability enhancement, weight and nervousness decrease, reduction of blood pressure, tri-glyceride and cholesterol levels, immune system improvement, and inhibition of tobacco and nicotine addiction [42, 91–96].

Conventional Extraction of Bioactive Compounds from Tea

Selecting a proper extraction technique is essential for recovering the maximum amount of bioactives from tea. The conventional solid–liquid extraction (CSLE) methods are commonly used due to their ease and broad applications [97] and liability [97–101]. Soxhlet extraction is a standard technique for extracting phenolic compounds from tea using organic solvents such as methanol, ethanol, acetone, diethyl ether, and ethyl acetate. Flavonoids can be extracted with polar solvents such as ethanol, methanol, water, and combinations of these solvents [102–105]. The choice of solvent depends on the solvent's ability to solubilize the solute, the extraction temperature, and the particle size of the solute [106].

Chang et al. [107] reported the possibility of extracting phenolics from green tea using co-solvents combined with carbon dioxide-assisted Soxhlet extraction. Although water is the commonly used solvent for extracting phenolics from tea, applying other non-polar green solvents (such as butanol, ethyl acetate, and ethanol) has been reported to extract bioactives and decaffeinating. It was reported in another study that using ethanol as a solvent improved the extraction of catechins than those extracted with water [36].

Goksu and Poyrazoglu [108] investigated the effect of using 80% methanol on total phenolic content (TPC) extraction from caffeinated and non-caffeinated green and black teas. There was a significant difference in TPC contents between caffeinated (159.4 mg/kg) and non-caffeinated (32.81 mg/kg) black teas. Similar results were observed for caffeinated (128.22 mg/kg) and non-caffeinated (43.16 mg/ kg) green teas. Ethyl acetate, compared to n-butanol and n-hexane, was found to be a better solvent for isolating catechins from green tea [109]. The optimum extraction conditions with water were reported to be at the solid-to-solvent ratio of 1:30, the temperature of 80 °C, and the extraction time of 40 min for the extraction of catechins, which were then isolated using ethyl acetate and decaffeinated using citric acid. The authors reported that this treatment could lead to a substantial reduction in the caffeine content by up to 78.8%. The application of liquefied dimethyl ether removed the total caffeine from dried green tea leaves before extraction, whereas catechins were retained up to 56% [110].

Another study used the green, deep eutectic solvent (DES) to extract catechins from Chinese green tea [111]. The results showed that the efficiency values of catechins, (+)-epicatechin gallate (EG), and (-)-epigallocatechin gallate (EGCG) were 82.7, 92.3, and 97.0%, respectively. Nadiah and Uthumporn [112] characterized catechins, caffeine, and gallic acid (GA) in tea leaves and spent tea. They evaluated the effect of various extraction conditions, such as boiling water, 50% ethanol concentration, and different extraction times. Compared to water, ethanol resulted in higher extraction efficiency of phenolic compounds from tea extracts, probably due to the higher polarity of ethanol that influenced the extractability rate.

The effect of particle size (intact and pulverized) and solvent type (methanol and acidified methanol) on the TPC, total flavonoid content (TFC), tannin content, and antioxidant activities of leaves of yellow, green, and black tea was studied by Kopjar, Tadić, and Piližota [113]. Pulverized tea leaves treated with acidified methanol exhibited the highest values of functionalities among the different tea leaves. The yellow tea leaves had higher bioactivity than the leaves of green and black teas. The antioxidant activities of extracts obtained from the yellow and green tea leaves were higher than those obtained from black tea leaves. Nibir et al. [114] have recently studied the antioxidant, and antimicrobial properties of aqueous extracts of flowery broken orange pekoe, broken orange pekoe, red dust, and green tea prepared with a solid-to-water ratio of 1:6. The aqueous extract of green tea exhibited promising anti-bacterial properties with a maximum level of phenolic content corresponding to 26.33 mg GA equivalent (GAE)/g extract.

Optimizing the operating parameters involved in the extraction process is essential to obtain bioactive compounds' maximum efficiency and functionality from plantbased food matrices [25]. The optimization of phenolic compounds extracted from tea fruit peel biomass (TFPB) was carried out by Xu et al. [115]. The highest TPC (47.5 mg GAE/g) was obtained at the optimum conditions of 43% ethanol, 60 °C extraction temperature, and 33 min extraction time. Gallocatechin and epigallocatechin were the major phenolic compounds of TFPB. In another study, Kim et al. [116] evaluated the optimization of the TPC, antioxidant activity, and EGCG of green tea leaves at different ethanol concentrations (0–100%), extraction times (3–15 min), and extraction temperatures (10–70 °C). The maximum antioxidant activity (88.4%) was obtained using 57.7% ethanol at 70 °C for 15 min. Zielinski et al. [117] optimized the extraction process of phenolic compounds from white tea, and optimum conditions were 10 min extraction time at 66 °C and using 30% ethanol solution. Hung et al. [51] evaluated the effect of using water and ethanol at different concentrations (10, 50, and 95%) on the extraction of catechin and the antioxidant capacity of different *C. sinensis* twig. The results showed that the 50% ethanol extract had the highest level of phenolic (161.3 mg GAE /L) and flavonoid (278.9 mg quercetin equivalents/L) contents.

Although the conventional extraction of bioactive compounds from tea is easy and convenient, applying these methods implies adverse thermal effects on the extraction yield and quality with a significant expenditure of solvents and energy. Thus, the potential of using innovative extraction methods such as ultrasound-assisted extraction (UAE), microwave-assisted extraction (MAE), pulsed electric field (PEF), supercritical fluid extraction (SFE), and accelerated solvent extraction (ASE)/pressurized liquid extraction (PLE) has been reported to be an excellent alternative to produce tea extracts at industrial scale with an optimal expenditure of energy and chemicals. The application of some of these techniques for extracting bioactive compounds from tea is discussed in the subsequent sections.

Innovative Methods of Extraction

Ultrasound-Assisted Extraction

Ultrasound-assisted extraction (UAE) is one of the most emerging, efficient, and eco-friendly methods to disrupt cells to extract intracellular compounds from the cell matrix. UAE works on a simple principle of cavitation phenomenon where micro channels are formed in the sample by increasing the rate of diffusion of the solvent into the matrix [118–121] (Fig. 1). It is also referred to as the mechanical waves that can increase with pressure leading to the formation of cavities [106, 122]. With the increase in pressure, the cavities reach a specific limit beyond which they cannot absorb more energy and lead to the collapse of the bubbles when they reach maximum volume, thereby aiding in the disruption of the cells [122–124]. UAE can enhance the mass transfer rate of bioactive compounds during the extraction process from the plant tissues [125]. The application of this method reduces the extraction time and energy consumption and provides higher extraction yield [126–130]. Mixing the solvent using UAE will also increase the surface area under contact between the solvent and the cell matrix [118, 131]. Due to the reduced extraction times, UAE can decrease the thermal degradation of heat-sensitive bioactive compounds such as polyphenols [132].

UAE has been in use for a long time to extract bioactive compounds from various tea leaves, as referred to in Table 1. Most of the research work was on green tea leaves [121, 134–143] and Black tea [31, 136–138, 143]. In addition, research has been carried out to extract bioactive from tea infusions [39], matte leaves [144, 145], tea solids [146-148], tea seeds [131], yellow tea [122], white tea [136, 137, 149], and Oolong tea [136, 137]. Solvents used for the extraction process were dependent on the targeted bioactive compounds for the process. Water remained the most used solvent for extraction [31, 121, 122, 138, 142, 143, 146-148] followed by methanol [138, 143, 144, 148, 149], ethanol [122, 138, 142, 147, 149], hexane [131, 144], and acetone [147]. Sometimes solvents like water/acetone and methanol/ethanol were used in combination for better yields. The solvent used for the extraction process should also be appropriately selected based on its selectivity for the target bioactive compound. For example, methanol served as the best extraction solvent for tannic acid, with a 19% higher yield [148].

UAE experiments were designed to understand the effect of various processing parameters like temperature, sonication time, power, and solid solvent ratio on the extraction efficiency of tea [39, 122, 131, 134, 140, 141, 144, 146, 147]. UAE was explored as an extraction method to extract

Fig. 1 Graphical representation of the cavitation formation and bubbles collapse accelerating the release of bioactive compounds from the plant cells (retrieved from Roohinejad et al. [133])



Sample	Extraction conditions*	Extracted compounds	Keynote (s)	Reference
Tea	60 °C, 10 min, 20 kHz, water	Tea solids	 Higher extraction efficiency up to 40% after 10 min of sonication Improved extraction efficiency at lower temperatures 	Mason and Zhao [146]
Tea infusions	60 °C, 40 min, 40 kHz, 250 W, water	Polyphenols, amino acid and caffeine	 Better extraction yield of the chemical compound's aroma compounds and other glycosidic precursors at lower temperature Better sensory quality attributes of the UAE- extracted tea compared to the conventional extraction method Extraction method Extraction yield of polyphenols were at 22.67% (wt.), amino acids at 1.81% and caffeine at 2.94% 	Xia et al. [39]
Matte tea	75 °C, 180 min, 40 kHz, 90 W, ethanol	Caffeine, phytol, and palmitic and stearic acids	- No significant differences in quality of the extracts obtained from the different extraction methods	Jacques et al. [144]
Green tea	28 °C, 30 min, 25.1 kHz, water	Catechins	 Effective method for increasing the extraction yield of catechin from green teas at low temperatures Ultrasonic power applied was the most main parameter affecting on the catechins extraction Effective for catechin extraction at ~15 mg with UAE 	Koiwai and Masuzawa [134]
Green tea Green tea, and Black tea	90 min, water, acetone and ethanol 40 °C, 10 min, 35 kHz, methanol, water, acetonitrile	Catechins and caffeine Catechins (EGC, + C, EC, ECGC, and ECG), and caffeine	 Significantly improved extraction yield Dynamic UAE: increased the extraction efficiency, decreased extraction time A significant reduction in the rate of oxidation and hydrolysis of the analysts Extraction yield of caffeine was 34 mg/g and catechins at 209 mg/g with UAE 	Saito et al. [147] Gu et al. [143]
Tea	50 °C, 20 s, 20 kHz, water	Tannic acid	 Best extraction solvent was methanol The highest extraction yields 	Sonawane and Patil [148]
Tea seeds	30 °C, 30 min, 24 kHz, 50 W, <i>n</i> -hexane	Oil	 - A shorter time for the oil extraction with the minimal solvent usage - A substantial increase in oil extraction yield (46.23–85.21%) with an increase of the ultrasonic power (10–50 W) and a decrease of the temperature 	Shalmashi [131]
Green tea	45 °C, 60 min, 37 kHz, 95 W, ethanol	Flavonoids	The high process repeatability with achieving the highest extraction polyphenols	Naşcu-Briciu et al. [141]

Table 1 Summary of extraction of bioactive compounds obtained from tea under various UAE conditions

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Sample	Extraction conditions*	Extracted compounds	Keynote (s)	Reference
Yellow tea	38 °C, 30 min, 20 kHz, 200 W, ethanol	Antioxidants (e.g., flavonoids, non- flavonoids, Polyphenolics, and methylxanthines)	-The maximum extraction yield of polyphenols and methylxanthines from yellow tea using the ultrasound probe in presence of ethanol (75%) as solvent -Extraction yield of caffeine was 27.88 mg/L after UAE	Horžić et al. [122]
Black tea, green tea, Oolong tea and White tea	32 °C, 21 min, methanol	42 volatile compounds	Better release of volatiles from the plant matrix at lower temperatures	Sereshti et al. [137]
Green tea infusions	60 °C, 15 min, water	Catechin (EGCG)	Increased extraction yield of EGCG by 15% with the highest oxidative stability	Lante & Friso [135]
Black tea, green tea, Oolong tea and White tea	3 min, methanol and water	Theophylline, theobromine, and caffeine	 A simple, low-cost, and eco-friendly procedure to isolate polar and hydrophilic molecular species from the aqueous solutions The high desorption of analysts with low volume of the organic solvent ~ 97 to 108% recovery of theobromine, 92 - 110% recovery of theophylline and 88 - 106% recovery of caffeine was observed 	Sereshti et al. [136]
Green tea	40 °C, 120 min, ethanol	Caffeine, and catechins	 The high recovery of catechins in presence of ethyl acetate/ dichloromethane for isolation Higher extraction efficiency than room temperature extraction and reflex extraction based on the process time and the productivity rate Low temperature decreased the process time and prevented the epimerization of catechins caused by extraction at high temperatures Better recovery of catechins from the green tea extracts by organic solvents 	Choung et al. [142]
Matte tea	16 °C, 47 kHz, water	Soluble matter	- The enhanced extraction yield (> \sim 31%) and efficiency \sim 74% with reduced extraction times	Kotovicz et al. [145]
Black tea	40 °C, 1440 min, 25 kHz, 150 W, methanol and water	Polyphenols	 Higher quasi equilibrium concentrations in the liquid phase by the ultrasonic intensification process Increasing the amount of polyphenols extracted by 15% Increased polyphenols content by 30–35% 	Both et al. [150]
Java tea	30 min, 20 kHz, 300 W	Bioactive compounds (phenolic, flavonoids)	Higher extraction yield (86–95%) for bioactive compounds compared to the conventional Soxhlet method	Lam et al. [151]

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Table 1 (continued)				
Sample	Extraction conditions*	Extracted compounds	Keynote (s)	Reference
Green tea	65 °C, 57 min, 28 kHz, 150 W, water	Caffeine and catechins	An increased extraction efficiency (85%) for catechins, phenolics (96 ± 6 mg gallic acid/g of DW), and antioxidant activity (EC ₅₀ value for DPPH inhibition = 66 mg/g) the bioactives obtained from green tea	Ghasemzadeh-Mohammadi et al. [121]
Green tea	80 °C, 30 min, 500 W, water	Catechins	 Improved extraction efficiency of catechins from green tea in the presence of BGG-4 (betaine, glycerol and D (+) glucose) More stability of catechins in DES (deep eutectic solvents) extracts compared to the other solvents used 	Jeong et al. [139]
Sage herbal byproducts of filter tea factory	75.4 °C, 80 min, 40 kHz, 42.5 W, ethanol	Phenolics and flavonoids	 The most important extraction parameters were the temperature and the ethanol concentration Total phenolics extraction was 9.8 GAE/ 100 g DW and total flavonoids extraction was 6.9 GAE/ 100 g DW with UAE as extraction method 	Zeković et al. [152]
Green tea	21 min, 461.5 W, DES choline chloride glycerol	Polyphenols	-Increased extraction yield of total phenolics (243 GAE/g), catechins, and antioxidant activity	Luo et al. [140]
White tea	(5, 10, 15 min), intensity (40, 70, and 100%) methanol, ethanol, and methanol/ ethanol	Phenolic compounds	-The extraction efficiency varied with the sonication time, power and solvent type -The maximum amount of polyphenols (68.38 mg GA/g) and antioxidant activity (77.65%) at 15 min time, 70% intensity with methanol as solvent	Ahmadi et al. [149]
Black and green tea	60 °C, 20 min, Ethanol, methanol, and water	Phenolic compounds, Isolation of catechins and theaflavins	-UAE was more effective than the conventional (757.33 vs. 741.66 mg/100 g GAE) for the isolation of bioactives - The highest extraction efficiency with ethanol	Imran et al. [138]
Black tea	4 °C, 30 min, 40 Hz, water	Phenolic compounds	-UAE maximized the efficiency of extraction under cold brewing conditions -The total phenolic content was 4 times higher comparison to hot brewing	Raghunath and Mallikarjunan [31]
*1- Temperature (°C), 2- E	ctraction time (min), 3- Frequency (kH	(z), 4- Power (W), 5- Solvent		

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compounds like catechins [134, 135, 138, 139, 142, 143], caffeine [136, 142, 143], tannic acid [148], oil [131], flavonoids [141], volatile compounds [137], theophylline and theobromine [136], theaflavins [138], antioxidant polyphenols [31, 140], total phenolic content [149], and antioxidant capacity [138] of tea extracts were analyzed with UAE. Among these, the catechins were found to be more stable in deep eutectic solvent (DES) extracts than in any other solvents (e.g., water, methanol, and ethanol) used for the extraction process [139]. In addition, UAE has been studied for increasing the total phenolic content and antioxidant capacity in cold-brewed black tea [31].

UAE effectively increased the extraction efficiency of catechin and decreased the extraction time and solvent consumption [134]. The application of dynamic UAE also reduced the oxidation and hydrolysis of the analytes because the system was airtight [143]. Using 50% acetone with sonication provided the highest extraction yield of 36% and 17% for EGCG [147] compared to conventional hot brewing at 33% and 12%. The highest extraction of total flavonoids with UAE (2,957.73 mg/L) was using ethanol as a solvent in comparison to conventional water extraction at ~ 500 mg/L [122]. Using water as a solvent, the extraction yield for green tea was 85% [121]. The solvent BGG-4 (betaine, glycerol, and D (+) glucose) for UAE improved the extraction efficiency of the catechins (217 mg g^{-1}) vs. UAE with water as solvent led to 100 mg g^{-1} for total catechins for green tea [139]. The optimum conditions for temperature included a range of 28 - 60 °C [39, 131, 134, 137, 141, 142, 146–148], power in the range 50–461W [39, 131, 140] and frequency in the range of 20 kHz—40 kHz [39, 122, 134, 148]. Sonication time for optimized conditions varied from as low as 10 s to as high as 120 min [39, 122, 131, 134, 137, 140–142, 146–149]. UAE is efficient at lower temperatures (60 °C), thereby minimizing thermal damage and maintaining the organoleptic characteristics of tea [39]. Application of UAE at high pressure, such as 90 to 338 kPa, increased the extraction yield to 200% and reduced extraction time and efficiency of extraction to 74% [145]. UAE significantly reduced the cold brewing time from 16 h to 30 min providing an energy-efficient method of extraction from black tea leaf matrix [31]. The application of UAE enhanced the extraction yield of EGCG to 15%, and the highest oxidative stability was observed in the nanoemulsion sample prepared with green tea with peanut oil [135]. The UAE with deep eutectic solvents resulted in an increase in the extraction yield of TPC, the total quantity of four main catechins, and the antioxidant activity of green tea polyphenolic extract [140]. Increasing the ultrasonic power from 10 to 50 W increased the extraction yield from 46.23 to 85.21%. The yield decreased by increasing the temperature [131].

The major advantages of using UAE compared to the conventional extraction methods include faster energy transfer, adequate mixing, faster response to process control systems, lesser energy consumption with average extraction time, reduction in thermal degradation, greater purity of the finished product, and fast return on investment [123]. The extraction efficiency usually differs from one tea to another due to the differences in the structure and the chemical composition of the leaf. Other factors, such as the turbidity of the plant tissue (tea leaves) and the starch granules, can also be influenced by the ultrasound energy applied and, thus, the effectiveness of the extraction [153]. UAE can be considered for shorter extraction time with minimal solvent usage for extraction of bioactives [131].

Microwave-Assisted Extraction

Microwave-assisted extraction (MAE) has received extensive attention as an alternative method for extracting bioactive compounds from plant-based food matrices [27, 125, 153–155]. The principle of MAE involves the propagation of non-ionizing electromagnetic waves between X-rays and infrared rays in the electromagnetic spectrum. These waves can penetrate the sample, interact with the compounds, especially polar compounds, and generate heat, which subsequently leads to changes in the structure of the cells [123, 125, 153, 156] (Fig. 2). The synergistic combination of heat and mass transfer, working in the same direction, is the primary cause of the process acceleration and the increased extraction yield [153, 157, 158].

MAE is a sequential process. The solvent initially penetrates the solid matrix of tea, followed by the structural breakdown. This aids in transporting the solutes rich in bioactive compounds out of the matrix. The solute migrates from the external solid surface to the bulk solution leading to the separation and the discharge of the extract containing bioactive compounds [153, 160, 161]. Then, the solvent interacts with the free water molecules present in the plant cells resulting in the rupture of the cell wall and aiding in the release of bioactive compounds from the cells to the solvent [162, 162, 163].

MAE was used to optimize better extraction efficiency of the bioactive compounds in various tea varieties which, includes green tea [3, 30, 37, 154, 155, 164, 165], black tea [27, 155, 164, 165], oolong [155, 164], decaffeinated green tea [166], mulberry tea [167], tea flower (*C. morifolium*) [151], decaffeinated Iranian green tea leaves [121], and tea blends consisting of white tea, green tea, mint, and peppermint [38]. MAE was suitable for producing green tea extracts rich in polyphenols [30].

MAE was used for the extraction of polyphenols [3, 30, 37, 164, 166, 168], tea flavonoids [11], antioxidant and anti-diabetic properties of tea extracts [151], and total phenols recovery [27, 94, 95, 165, 169]. The bioactive compounds extracted using MAE include catechin and epicatechin [38, 154], caffeine [38, 155, 170], and tea saponins

from the oil—tea camellia seed cake [171]. The optimized temperature conditions used for MAE were in the range of 80-200 °C [3, 30, 94, 95, 151, 152, 164, 169]. The temperature was considered the primary variable [3, 121] for caffeine and catechin extraction using MAE [152]. However, even though a higher temperature was used, MAE was only carried out for a short period ranging from a minimum of 30 s to a maximum of 60 min [11, 27, 30, 37, 38, 94, 95, 151, 152, 154, 155, 166-169, 171] and microwave power in the range of 150–900 W [11, 27, 30, 38, 94, 95, 121, 151, 152, 155, 165–168, 170]. The solvent composition, solid-tosolvent ratio, extraction temperature and time, microwave power, stirring speed, pressure, and surface area of contact are the critical parameters affecting the extraction of bioactive compounds in an MAE process [88, 153, 156]. The order of influence of these parameters was deduced to be irradiation time > intensity > tea to the solvent ratio [166].

The solvent used for the extraction depends on the solvent's penetration and interaction with the targeted bioactive compounds [153, 172]. Some of the solvents used for the extraction process were water [30, 37, 121, 151, 169], ethanol [37, 38, 94, 95, 152, 168, 169], and acetone and methanol [37, 169]. The solvent should also have the capacity to absorb the microwave energy, but absorption depends on the boiling point, microwave energy dissipation, and dielectric properties of the solvent. Depending on the solvent volume,

the heating time would be altered [27]. The ethanol concentration was the most influential parameter for the extraction of polyphenols. The enhancement of product recovery in MAE is due to the heating effect of the microwave [168]. MAE can be considered one of the fastest extraction methods for polyphenols and caffeine [37]. The caffeine recovery in the tea samples was found to be in the range of 88.2% to 99.3% [170] using dynamic MAE. MAE's extraction efficiency was more than 95% [11, 94, 95, 121] for the recovery of catechins and derivatives within a short time [3, 11, 154]. It also had a good recovery process compared to conventional methods [152]. MAE-assisted green tea extracts were more concentrated in total polyphenols (26%) compared to black tea (16%) [165]. The yield and efficiency of the extraction increase proportionally to the time and the microwave applied power and decrease beyond a specific limit mainly due to the increase in temperature. Microwave power is a crucial parameter to optimize the extraction process. The mixing process is an essential consideration in the optimization process since it is directly related to the mass transfer in the solvent. The surface area under contact also enhances the efficiency of the extraction process. The finely powdered samples usually have a large surface area aiding in a better contact surface between the plant matrix and the solvent, thereby deepening the penetration of the microwaves [153, 172]. The extraction time and the irradiation power were the



Fig. 2 Comparative illustration of conventional (A), ultrasound (B), and multi-mode microwave (C) applicator used to extract bioactive compounds from tea tissues (retrieved from Barba et al. [159])

parameters that should be reduced to decrease the degradation of polyphenols [168].

Many researchers have reported that the MAE is more effective than the conventional extraction methods (e.g., solid-liquid extraction, extraction at room temperature, maceration, reflux extraction) in extracting bioactive compounds from green teas at ambient temperature [11, 37, 173]. The results of the bioactive extraction from tea leaves using different operating conditions of MAE are summarized in Table 2. MAE can be used for industrial applications with minor modifications in the sample size and solvent-to-solid ratio [168]. MAE was the fastest extraction method, involving a rapid extraction of phenols [164]. The extraction time was reduced to more than eight times when compared with HRE (hot reflux extraction), two times when compared to UAE (ultrasound-assisted extraction), and five times when compared to SFE (supercritical fluid extraction). The extraction yield was increased by 17.5% compared to HRE. The energy consumption was 1/4 when compared to UAE, with a 40% increase in the total phenolics. The study concluded that MAE only required a shorter time and lesser energy consumption and provided higher extraction selectivity and extraction yield [94].

MAE is an easy, convenient [167], fast, and reliable method for the recovery of catechins and epicatechins [154]. It can be considered one of the effective methods of extraction [166, 169] due to its targeted extraction of bioactives and thereby enhancing the antioxidant quality of the extracts [168]. Compared to the UAE, the extraction efficiency of bioactives was higher with MAE [121] and more efficient at higher temperatures. MAE also led to a higher recovery of total phenolic compounds, compared with the standard brewing techniques, without affecting the antioxidant potential of the tea [27]. MAE reduced the extraction time, energy consumption, and environmental burden [30], making it an alternative technology for the extraction of bioactives from tea.

Pulsed Electric Field

Pulsed electric field (PEF) is a non-thermal technology that extracts targeted bioactive compounds from tea. This method works on the simple principle of cell wall disruption, increasing the cell matrix's permeability for efficient extraction. PEF is one of the emerging technologies in the process industries and is considered competitive compared to the other processes concerning cost-effectiveness [174]. For the extraction process, the tea sample is placed between two electrodes, followed by the application of high electric field pulses for a concise duration (ns to μ s) [34, 35, 175–180] (Fig. 3). Currently, high-intensity PEF (> 10 kV/ cm) is used as a preservation method for the inactivation of microorganisms due to the breakage of cell membranes [181, 34, 35, 182, 183] and in turn increases the shelf life of the food [34, 35, 179, 184]. The size and formation of pores can be reversible or irreversible, depending on the pulse intensity, the electric field strength, the number of pulses, and the treatment time [33]. The effect of various operating parameters involved in PEF technology on the extraction efficiency and functionality of tea's bioactive compounds are summarized in Table 3.

PEF treatment induced pores on the tea matrix's surface and facilitated the solvent's penetration and the polyphenols' migration [187]. Electroporation efficiency is also controlled by the electric field intensity [180]. The degree of disintegration of the cell matrix also depends on treatment time, and intensity of the electric field applied. Lower intensities take a longer time for the electroporation and vice versa [180]. Thus, increasing the intensity of electric fields increases the extraction yield proportionately [33, 177]. For the solvents used for PEF extraction, the conductivity and solubility of targeted compounds in the solvent are critical. The increase in conductivity of the solvent will increase the extraction efficiency due to enhanced electroporation [177]. In addition, He et al. [171] reported that increasing the solvent concentration can reduce the concentration of the bioactive compounds bound to the sample, making the dissolution process easier. Other parameters for optimization include the pulse duration, the number of pulses, and the pause between the pulses [180]. The longer pulse duration for the extraction was more effective compared to a short duration. The longer pulse duration increases the permeation of the cells but negatively leads to the decomposition of the extracts. The effect of the pulse width varies depending on the electric field strength, types, quality, and contact parameters, such as the geometry and the size of the samples [180]. The extraction kinetics strongly depends on the interval between the pulses and higher field strengths [180].

By using PEF for extraction, tea's aromatic compounds, polyphenols are protected from damage [34, 35, 188], and its color, taste, and aroma remain unchanged [34, 35, 39, 179, 186, 188]. Traditionally, PEF has been used in the extraction and retention of polyphenols from green tea [179, 187], black tea [186], Puer tea [174], and green tea infusions [33-35]. PEF retained more bioactive compounds and color than conventional heat treatment [34, 35]. PEF is mainly used to extract polyphenols, catechins, and free amino acids [35]. The extraction yields for polyphenols ranged from 22 to 32% [179, 180, 186] and the extraction rate was observed to increase ~ 2 times without degrading or altering the phenolic profiles [187]. Using PEF beyond a specific limit, had a loss of < 10% in volatiles and but it increased amino acid content by 7.5% [34, 35]. On the contrary, it reduced the loss of aromatic compounds due to the volatilization in instant tea [186]. Additionally, PEF treatment has been known to improve the

Table 2 Summary on the MAE condit	tions to extract bioactive compounds fr	rom tea		
Samples	Extraction conditions*	Extracted compounds	Keynote (s)	Reference
Green tea	Ethanol, 4 min, 700 W, 20:1, 90 °C	Polyphenols, and caffeine	 Faster extraction of polyphenols (30%) and caffeine (4%) with higher extraction yield than other conventional extraction methods like RT, UAE, Heat Reflux 	Pan et al. [37]
Tea	Water, 65 min, 150 W, 100 °C	Flavonoids TEC (tea epicatechins), TCD (total catechins derivatives), and TTP (total tea polyphenols)	 Maximum extraction yield within a short period of time Extraction of polyphenols at~ 10% Extraction of epicatechins using MAE in green tea and black tea were~65 mg/g and~25 mg/g 	Sultana et al. [11]
Green tea	Water, 60 min, 600 W, 20:1, 80 °C	Polyphenols	 Suitable for producing tea extracts rich in antioxidants, flavanols and polyphenols, with the highest concentration of EGCG (epigallocatechin gallate) and antioxidant activity 97.46 mg catechin / g of tea and 77.14 mg CE/g with MAE Shorter extraction time with a notable reduction in the energy consumption 	Nkhili et al. [30]
Black tea	Water, 600 W, 100:1	Polyphenols	Higher recovery rates of phenolic compounds compared to the normal brewing techniques without any negative effect on the tea's antioxidant potential	Spigno and De Faveri [27]
Green tea, Oolong tea, and Black tea	Water, 2 min,1000 W, 20:1, 230 °C	Phenolics (e.g., pyrogallol, catechol, dihydroconiferyl alcohol, and vanillin)	 The high extraction yield of green tea with 24.6% pyrogallol The good extraction yield of oolong tea extract-with 10.3% dihydroconiferyl alcohol and 8.1% of vanillin A rapid extraction for tea phenols only within 2 min 	Tsubaki, Sakamoto, and Azuma [164]
Black tea, and Green tea	Water, 450 W, 30:1, 70 °C	Polyphenols	-Higher concentration of total polyphenols (26%) in green tea than that of black tea (16%) - MAE: an effective low-energy, and time-saving method for obtaining extracts rich in phenolic compounds with strong free-radical scavenging activities. from both teas	Nshimiyimana and He [165]

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Table 2 (continued)				
Samples	Extraction conditions*	Extracted compounds	Keynote (s)	Reference
Green tea	Water, 1 min, 400 W, 50:1	Catechins, and epicatechins	<ul> <li>A good recovery procedure for catechins (118%) and epicatechin (120%)</li> <li>A simple, faster, and reliable technique for the catechin extraction from green tea</li> </ul>	Li et al. [154]
Green tea (decaffeinated)	Water, 3 min, 600 W, 20:1, 68 °C	Polyphenols	- The extraction efficiency and polyphenols content were highly affected by the microwave irradiation time - Extraction yield using MAE was ~ 14–15% and EGCG was at ~ 22–25%	Li and Jiang [166]
Tea	Ethanol, 10 min, 600 W, 12:1, 80 °C	Polyphenols	<ul> <li>- 96.5% extraction yield of polyphenols</li> <li>- The extraction time was saved &gt; 8 and 5 times compared with HRE (Heat reflux extraction) and UAE (Ultrasound assisted extraction)</li> <li>- Lower energy consumption and higher extraction selectivity</li> </ul>	Wang et al. [94]
Tea	Ethanol, 4 min, 70 W, 100:1, 80 °C	Caffeine	<ul> <li>High recovery rate of caffeine in the tea samples (88.2–99.3%)</li> <li>The caffeine yield using DMAE (dynamic microwave-assisted extraction) (47 mg/g) was higher than SAME (static microwave- assisted extraction) (37 mg/g) with a reduced volume of organic solvent and reduced time required for the preparation</li> </ul>	Wang et al. [170]
Green tea, black tea, and Oolong tea	Water, ethanol, acetonitrile, 6 min, 600 W, 50:1, 80 °C	8 catechins monomers, and caffeine	<ul> <li>Higher extraction of caffeine and catechins (mainly EGCG) in black and green teas</li> <li>The increased extraction yield of catechins and caffeine with increasing the irradiation time by 6 min</li> </ul>	Rahim et al. [155]

Samples	Extraction conditions*	Extracted compounds	Keynote (s)	Reference
Tea camellia seed cake	Ethanol, 4 min, 400 W, 10:1, 60 °C	Saponins	<ul> <li>Notable reduction of extraction time from 6 h to 4 min</li> <li>Enhanced extraction yield by 14% with a significant decrease (up to 50%) in the consumption of organic solvent</li> </ul>	He et al. [171]
Mulberry tea	Water, 11.41 min, 602.28 W, 80:1	1-deoxynojitimycin	MAE was more convenient than the extraction method by hot water immersion	Liu et al. [167]
Tea	Ethanol, 3 min, 500 W, 100:1, 80 $^\circ\mathrm{C}$	Polyphenols	Best method for the extraction of tea polyphenols	Bekdeşer et al. [169]
Tea (C. morifolium)	Water, 5 min, 400 W, 20:1, 80 °C	13 major bioactive compounds	- Reliable method to prepare samples for the extraction processes	Lam et al. [151]
By product of filter tea factory- wild apple extract	Ethanol, 18.7 min, 600 W, 20:1	Polyphenols	-Ethanol concentration was the most influential parameter to extract phenols -Reduction to the extraction time and irradiation power to decrease the degradation of polyphenols - Extraction yield of total solids was at 16.09 mg/mL, phenolics at 5.29 mg GAE/ ml and flavonoids at 3.37 mg CE/ mL using MAE	Pavlić et al. [168]
Tea (Iranian green tea)	Ethanol, 7.8 min, 190 W, 40:1, 110 °C	Caffeine and catechins	<ul> <li>High extraction efficiency (95%) with a high TPC (125±5 g of gallic acid/g DW)</li> <li>Temperature played an important role in the extraction of caffeine and catechins</li> <li>The best technique for the extraction of polyphenols with a yield of 90%</li> </ul>	Ghasemzadeh-Mohammadi et al. [121]
Sage herbal dust from filter tea factory	Ethanol, 18.7 min, 600 W, 40:1	Phenols and flavonoids	<ul> <li>- A better recovery process for sage polyphenols compared to the conventional traditional methods</li> <li>- Extraction of total phenols at 10.3 GAE/ 100 g DW and flavonoids at 7.5 g CE/ 100 g DW using MAE</li> <li>- Ethanol concentration played a critical role in the efficiency of extraction process</li> </ul>	Zeković et al. [152]

Samples	Extraction conditions*	Extracted compounds	Keynote (s)	Reference
Tea blends (white tea, green tea, mint, and peppermint)	Ethanol, 3 min, 18.1	Bioactive compounds	-Use of hydroalcoholic solutions to extraction bioactives -High yields of antioxidants, total phenols (2164 mg/100 g) and flavonoids (12.71 mg /100 g) -Shorter processing time	Rehder et al. [38]
Green tea	Water, 250-300 s, 15:1, 140-220 °C	Bioactive compounds	<ul> <li>A significantly shorter time in the MAE than autohydrolysis</li> <li>The critical role of temperature for antioxidant activity and TPC, reducing in the MAE extraction process</li> </ul>	Sanz et al. [3]

Table 2 (continued)

^{*}1- Solvent, 2- Extraction time (minutes), 3- Power (W), 4- Solvent to solid ratio, 5- Temperature (°C)

taste and aroma of tea samples and characteristics like the natural aging of tea. The high voltage of PEF assisted in artificial aging, thus improving the taste with an accelerated aging process. It can also be used to rapidly age unfermented Pu Erh tea [174]. Overall, this extraction technique provided the maximum extraction yield of polyphenols without destroying their activities [179] compared to hot brewing methods. Moreover, PEF has been investigated as an alternative to drying processing before extracting polyphenols [187].

A longer shelf life of infusions was achieved (about 90 days) when storage temperature at 4 °C was considered along with PEF treatments [34, 35]. A slight temperature increase was recorded during the treatment at  $\leq 10$  °C [33]. Effective log reductions were observed for *Escherichia coli* and *Staphylococcus aureus*, at 38 kV/cm and less treatment time 16–200 µs [34, 35]. PEF is a time and energy-efficient extraction method. The advantages of using PEF include minimal energy consumption, uniform transmission, and quicker processing [174, 189]. PEF has not been extensively researched compared to other technologies reviewed in the study due to economic limitations. More validation and further research into the extraction using PEF should be studied and analyzed to understand the underlying effects of PEF on tea.

# **Supercritical Fluid Extraction**

Supercritical fluid extraction (SFE) is one of the alternative technologies used to extract bioactive compounds from tea leaves [88, 106, 190, 191]. SFE works based on the principle of the supercritical properties of fluids like  $CO_2$  and water. SFE is a two-step process, extraction, and separation. The system consists of an extraction chamber, wherein the tea samples are placed inside the supercritical fluid. The samples are then subjected to a specific temperature and pressure to extract the bioactive compounds. After the extraction process, the mixture containing the bioactive compound passes through the separator for the separation process (Fig. 4) [191].

SFE was used in the extraction of theophylline, theobromine, and caffeine [193–197], safrole and allylbenzenes [198], monoterpenes [199], triterpenic acids [200], alkaloids [195], phenolics [196], volatile compounds [201], and oil [202]. Japanese tea [203], Sassafras tea [198], Mate (*IIex paraguariensis*) leaves [193, 197, 199], *Malaleuca alternifolia* Cheel leaves [204], herbal tea [200], *Hedyotis diffusa* and *Hedyotis corymbose*, Assam tea—seeds (*C. sinensis var. assamica*) [202], Korean tea [205], green tea [194, 196, 201, 205–207] were all studied for the extraction of various bioactive compounds using SFE from tea leaves.

The temperature and pressure can be adjusted based on the dissolving power of the target bioactive compounds

Table 3 Summ	ary of extraction of bioactive compounds obtained fr	om tea under various PEF conditions	
Sample	Extraction conditions*	þ	Keynote (s)
Green tea	38.4 kV/cm, 20 °C, 160 µs, 667 pps, 2 µs	Polyphenols and free amino acids	- A promising technology to maintain

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Sample	Extraction conditions*	р	Keynote (s)	Reference	
Green tea	38.4 kV/cm, 20 °C, 160 µs, 667 pps, 2 µs	Polyphenols and free amino acids	<ul> <li>A promising technology to maintain the quality of the bioactive compounds and color</li> <li>The inactivation of microorganisms (e.g., <i>Escherichia coli and Staphylococcus aureus</i>)</li> <li>Synergistic effect in the reduction of the microorganisms by the low-temperature storage and antibacterial property of polyphenols extracted</li> <li>No significant effect on polyphenol extraction with PEF (1.82 mg/mL (PEF) vs 1.82 mg/ml (untreated)) and 36% in amino acids compared to conventional methods</li> </ul>	Zhao et al. [185]	
Green tea	20–40 kV/cm, 121 °C, 50–200 μs, 667 pps, 2 μs	Polyphenols, catechins, and free amino acids	High retention of color and bioactive compounds	Wang et al. [183]	
Green tea infusions	38.4 kV/cm, 37 °C, 200 μs, 667 Hz, 2 μs	NR	<ul> <li>Shelf-life period more than 90 days (at 37 °C) for the infusions with PEF</li> </ul>	Zhao et al. [34]	
Green tea infusions	20 to 40 kV/cm, 5–15 °C, 200 μs, 667 pps, 2 μs	Catechins, polyphenols, and free amino acids	- Increased content of amino acids (specifically theanine) by 7.5% at 40 kV/cm with the loss of volatiles ( $\approx 10\%$ ) - Efficient retention of the bioactive compounds and color	Zhao et al. [35]	
Tea	0.9 kV/cm, 5 °C, 5 × $10^5$ µs, 5 × $10^4$ µs	Polyphenols	<ul> <li>27% maximum extraction yield for polyphenols</li> <li>Significant effects of the pulse and strength of electric field on the extraction yield</li> </ul>	Zderic et al. [180]	
Black tea	20 kV/cm, 95.83 µs, 125 Hz, 2 µs	Total solids, polyphenols, and amino acids	<ul> <li>- A 22.7% extraction yield for instant black tea powder</li> <li>- Improved the tea solubility in cold water along with reducing the tea cream</li> <li>- Total polyphenols (40.73 mg/g), amino acids (20.79 mg/g) and total solids (221.19 mg/g) were extracted using PEF</li> </ul>	Ye et al. [186]	
Unfermented Pu'er tea	18 kV/cm, 60 min (extraction time), 200 pps, 120 Hz, 0.3 µs	Polyphenolics, and theine	<ul> <li>Improved the taste, aroma and other sensory parameters in tea samples compared with the effect of natural aging on teas</li> <li>Effective in the artificial aging as it improved the content of tea extract and its taste</li> <li>Shorter and quicker method for aging of unfermented Pu'er tea</li> <li>New method to enhance the tea quality and safety</li> <li>Total polyphenols extraction was~30% and theanine~4% and did not show significant difference compared to conventional method</li> </ul>	Chen et al. [174]	

Table 3 (continued)				
Sample	Extraction conditions*	þ	Keynote (s)	Reference
Tea	0.9 kV/cm, 10 °C, 3×10 ⁶ μs	Polyphenols	<ul> <li>27% maximum extraction yield for polyphenols</li> <li>Direct and positive relationship between the time of pulse and the treatment time</li> </ul>	Zderic and Zondervan [33]
Tea	1.1 kV/cm, 100 μs, 0.1×10 ⁻³ s, 50 pulses	Polyphenols	Maximum extraction yield (32.5%) of polyphenols with the PEF without destroying bioactive compounds compared to the conventional hot-brewing method	Zderic and Zondervan [179]
*1- Field strength (kV,	/cm), 2- Temperature (°C), 3- Pulse duration (μ	is), 4- Pulses per second (pps)/Frequency (Hz), 5	5- Pulse width (µs), NR not reported	

[106, 208], and they serve as critical factors for the extraction system [197, 209]. This is because the density of the solvent and solubilization of the solute will depend on the pressure applied to the system. However, increasing the pressure beyond a specific limit will negatively impact the extraction process as it might reduce the diffusion rate of the solvent. This will hinder the extraction resulting in lower yields [209]. Pressure is also a critical parameter in the SFE extraction [190], and most pressure conditions ranged from 7.4 to 88 MPa. Different flow rates for CO₂ were employed, and there is no guiding explanation involved why the flow rates were different for each sample; one detail can be attributed to the difference in the system used, and the flow rate ranges from 0.9 g to 1250 g / min [190, 195–197, 201–206]. Extraction curves further helped prove that the extraction rate increased with pressure [210]. Diffusivity and apparent volume of solvent increase with temperature, while density decreases with temperature. The extraction process ranged from 10 to 540 min, with temperatures ranging from 40 to 100 °C [190, 193-198, 201-207]. Depending on the sample matrix, varying time-temperature combinations were employed under the SFE extraction process. A decrease in the temperature conversely increases the density and the solvation of the solutes. This is called

a "crossover effect," where the high temperatures result in lower yields, and lower temperatures provide higher extraction yields. Thus, the author states that the increase in kinetic energy (due to temperature) will be directly proportional to the diffusion rate of  $CO_2$  [209]. However, Natolino et al. [210] state that temperature had a negligible effect on

extraction kinetics. SFE has increased the extraction efficiency and preserved organoleptic characteristics of the bioactive extracts. The supercritical fluid commonly used in SFE is CO₂ (with high purity) as it is non-toxic and non-flammable under low critical pressure, is cost-effective, and allows easy removal of the supercritical fluid from the extracts [123, 207]. Generally, the co-solvent used in the extraction process has intermediate volatility between the supercritical fluid and the extracted bioactive compounds. This aids in enhancing the solubility of the bioactive compound in the supercritical fluid. The type of supercritical fluids is the rate-determining step of the entire SFE process. The bioactive compounds (solutes) of interest in tea, such as polyphenols and alkaloids, are less soluble in carbon dioxide, and hence increasing the pressure increases the solubility of the solutes. In addition, cosolvents (isopropyl alcohol, ethanol, n-hexane, pentane, heptane, toluene, methanol, acetone) [203] can also be used to increase the solubility of polar compounds during the extraction process. For the optimization and strong interaction of the matrix and the bioactive compounds [209] the co-solvent type and concentration, like ethanol and water, during the initial pre-treatment used were the critical parameters for



Fig. 3 a-d Schematic illustration of the electroporation mechanism in the cell membrane exposed to an electric field (retrieved from Roohinejad et al. [133])



the extraction process of green tea [207]. Table 4 provides a summary of the extraction process of SFE in the extraction of targeted bioactives in tea over the years.

Ethanol served as a suitable solvent for extracting methvlxanthines [197]. Solvents such as ethanol [196, 197, 200, 206, 207], methanol [193, 203], hexane [204], and water [195, 205, 206] were used as the solvent phase in the extraction process. However, water was experimentally proved to be the best solvent for the selective extraction of caffeine from green teas with a selectivity of 0.88 compared to the selectivity of ethanol (0.24) [195]. SFE uses a lesser solvent for extraction [197]. The extraction success depends on selecting the conditions that can enhance the extraction of the desirable compounds by regulating the solvation power, avoiding the influence of other materials, and reducing the co-extraction of impurities [213]. The sample matrix of the leaves played a fundamental role in the extraction process for SFE [204]. The solvent removal from the system also depends on several factors, such as the solubility of the solute, the interactions of the solute-solid matrix, the localization of the solute in the matrix, and its porosity.

The extractability with SFE was found to be 57%, 68%, and 94% for theophylline, theobromine, and caffeine from mate tea [197], and a recovery of > 96% for safrole and other alkylbenzenes in Sassafras teas [198]. SFE has been

employed to remove caffeine in green tea by avoiding the extraction of antioxidants from the tea matrix [214]. For green tea, SFE showed a maximum removal of caffeine (91.5%) and retention of 80.8% of catechins [206]. The caffeine content in green tea was minimized to 2.6%, and 37.8% of ECGC was lost in the process [207].

Most of the process optimization was based on the maximum extraction of caffeine (54%) and EGCG (21%) [195]. The extraction rate was 36.1% for caffeine and 40.6% for catechins in green tea [196]. The increased amount of caffeine extracted using SFE was accompanied by decreased volatiles present in green tea [201]. The solubility of the extracted caffeine was found to be lower in green tea [194] as compared with pure caffeine (61 times higher). The combination of UAE and SFE extraction methods also resulted in higher extraction yields (15-16%), decreasing the extraction time to 95 min in comparison to 180 min of heat reflux extraction and 135 min of UAE, and aiding in minimizing the amount of solvent used (43 mL) [200]. SFE was efficient for extracting caffeine and theobromine compared to extraction rates of other polyphenolics from mate tea [193]. Chlorophyll was sometimes co-extracted with caffeine, and further processing was required to recover the chlorophyll to improve the quality of tea [196]. Additionally, antioxidant activity improved with the highest extraction

Samples	Extraction conditions*	Extracted compounds	Keynote (s)	Reference
Roasted Japanese tea	80°C, 40.5 MPa, methanol, 10 min, 450–500 g/min	Mesitylene (1,3,5-trimethylbenzene), 1-ethyl-3-methylbenzene, 1-ethyl-4-methylbenzene, 2-propiophenone, tetrahydro-2- furanmethanol, dihydro-2-furanone, benzenedicarboxylic acid bis(2- methoxyethyl) ester, nonacosane, and caffeine	- Quick alternative method to the liquid solvent extraction - Silylating agents were complexed with the sample and served as both polar modifier and derivatizing reagent -Higher extraction yield and efficiency with SFDE (simultaneous supercritical fluid derivatization and extraction) method compared to the SFE one	Hills et al. [203]
Sassafras tea (unbrewed)	80°C, 69.0 MPa, methanol, 15 min, 2 g/min	Safrole and allylbenzene	-96–101% recovery respectively for safrole and allylbenzenes - Accurate and better results within a short time with the SFE in comparison to the steam distillation	Heikes [198]
Mate tea	70 °C, 25.5 MPa, organic solvents, 420 min, 0.9–1.2 g/min	Caffeine, theobromine, and theophylline	- The extractability rate of theophylline, theobromine and caffeine was 57, 68 and 94%, respectively—Higher selectivity of caffeine compared to theobromine, and theophylline towards $CO_2$ - Retrograde behavior for caffeine with the temperature was recorded while theobromine and theophylline had a normal behavior	Saldaña et al. [199]
Tea tree (Melaleuca alternifolia Cheel)	100 °C, 7.4 MPa, hexane (rinse solvent), 10 min, 0.25 g/mL	Monoterpenes	The sample matrix has a fundamental role mainly in the SFE process	Wong et al. [204]
Mate tea	70 °C, 40 MPa, ethanol (co-solvent), 400 min, 5.7 g/min	Caffeine and methylxanthines	<ul> <li>- 98% extraction rate for caffeine</li> <li>- High efficiency of extraction process of methylxanthines by ethanol at low amounts</li> <li>- Applied temperature and pressure were critical for the extraction of bioactive compounds</li> <li>- A short time period for the caffeine extraction using the SFE</li> </ul>	Saldaña et al. [197]
Tea seeds	60–80 °C, 30–40 MPa, hexane, 20–30 min, 1 g/min	Tea seed oil	-Ethanol (modifier) and pressure were critical parameters -Extraction efficiency is 54% with MAE - The best method for obtaining tea seed oil without using the organic solvent	Rajaei et al. [211]

 Table 4
 Summary of extraction of bioactive compounds obtained from tea under various SFE conditions

Table 4 (continued)					
Samples	Extraction conditions*	Extracted compounds	Keynote (s)	Reference	
Korean tea	50 °C, 40 MPa, water, 60 min, 468 g/min	Caffeine	<ul> <li>- 66% extraction rate for caffeine</li> <li>- Extracting the catechins along with the caffeine at a higher temperature than 49 °C</li> </ul>	Kim et al. [205]	
Green tea	60°C, 30 MPa, ethanol or water, 10 min, 12 g/min	Caffeine and catechins	<ul> <li>Maximum removal (91.5%) of caffeine and the high retention of catechins (80.8%)</li> <li>Critical parameters were pressure, temperature, and ratio of CO₂ to tea</li> </ul>	Huang et al. [206]	
Tea	70 °C, 30 MPa, ethanol, 120 min, 8.5 g/min	Caffeine and catechins	<ul> <li>Critical role for the type and concentration of co-solvent (ethanol/ water) used in SFE process</li> <li>A significant decrease in the content of caffeine extracted using the SFE method (2.6%)</li> <li>A 37.8% reduction in ECGC by the SFE process (loss)</li> </ul>	Park et al. [207]	
Green tea	70 °C, 30 MPa, ethanol, 51 min, 1250 g/min	Volatile compounds and caffeine	<ul> <li>Higher caffeine and lower volatiles in tea extracts obtained by the SFE</li> <li>SFE is an efficient technique to decaffeinate green teas</li> </ul>	Lee et al. [201]	
Tea	40 °C, 40 MPa, water, 300 min, 468 g/min	Caffeine and EGCG	<ul> <li>-Maximum extraction yield of caffeine (54%) and EGCG (21%) by the SFE</li> <li>- Water as the best solvent for the selective extraction of the caffeine</li> <li>- The selectivity was found to be 0.88 for water compared to 0.24 for ethanol</li> </ul>	Kim et al. [195]	
Mate tea	50 °C, 15 MPa, methanol	Caffeine, theobromine, and polyphenolics	-Suitable method only for the extraction of caffeine and theobromine and not for the other polyphenolics from tea - Maximum extraction of caffeine was 33.6%(w/w) and theobromine at 0.2% (w/w)	Cassel et al. [193]	
Green tea	63 °C. 23 MPa, ethanol, 120 min, 8.5 g/min	Caffeine and catechins	<ul> <li>Extraction rate by SFE was 96.06% for caffeine and 40.61% for catechins</li> <li>Simultaneous extraction of chlorophyll caffeine</li> </ul>	[196]	

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Table 4 (continued)				
Samples	Extraction conditions*	Extracted compounds	Keynote (s)	Reference
Tea	50 °C, 30 MPa, no solvent, 10 min for static and 90 min for dynamic, 2000 g/min	Volatile compounds	Identification of 59 bioactive compounds using GC–MS in the essential oil of tea flowers	[190]
Tea	50 °C, 18.8 MPa, ethanol, 60 min, 2.94 g/min	Total phenols, flavonoids and Tannic Acid	- The most effective parameters: pressure and co-solvent used - High phenolic (131.24 mg GAE/100 m1),flavonoid contents (194.60 mg QE/100 m1) and tannic acid (49.99 mg TAE/100 m1)	Maran et al. [212]
Green tea	50 °C, 25 MPa, ethanol, 540 min	Caffeine	<ul> <li>The solubility ranged from 44.19–149.55×10⁻⁶ within a wide range of temperature and pressure</li> <li>Less solubility of the extracted caffeine compared to its pure form (61 times higher)</li> </ul>	Gadkari and Balaraman [194]
Tea (Hedyotis diffusa and Hedyotis corymbosa)	40 °C, 15 min, 40 kHz, 185 W, water	Triterpenic acids (e.g., oleanolic, and ursolic acids)	USC-CO ₂ (Ultrasound assisted supercritical carbon dioxide extraction) was higher than SCCO ₂ due to the higher extraction yield (up to $15-16\%$ ), and lower extraction time (95 min), with the minimum of solvent	Wei and Yang [200]
Tea seeds	50 °C, 220 Bar,4 h, 110–170 L/h	Tea seed oil	-The maximum of phenolic compounds: (15.89 mg GAE/100 g), the DPPH (IC50) (15.35 g/L), and ABTS (IC50) (87.23 g/L) -An efficient method for the extraction of oil from Assam tea seeds	Muangrat and Jirarattanarangsri [202]
Tea seeds	40 °C and 300 bar	Tea seed oil	-The extraction rate was increased at higher pressure -The temperature had less effect on the extraction kinetics -The maximum extraction efficiency (50.03 ± 0.68% w/w)	Natolino et al. [210]
*1- Temperature (°C), 2- Pressure (b	ar), 3- Solvent, 4- Time (min), 5- Flow r	ate		

yield of 50% (Natolino, Da Porto, and Scarlet 2022) in tea after SFE. The maximum TPC, TFC, tannin content, and total antioxidant activity was 131.24 mg GAE/100 mL, 194.60 mg QE/100 mL, 49.99 mg TAE/100 mL, and 262.23 µmol TEAC/100 mL for *Camellia sinensis* [190]. Supercritical CO₂ extraction efficiently extracted oil from Assam tea seeds with a maximum of TPC (15.89 mg GAE/100 g) and scavenging activities (IC50=15.35 g/L for DPPH and 87.23 g/L for ABTS) [202]. SFE can achieve higher extraction efficiency and yield [203] within a shorter extraction time [197, 198]. The selectivity of CO₂ for caffeine extraction was much higher [197, 199] than tea's other bioactive compounds.

#### **Pressurized Liquid Extraction**

The pressurized liquid extraction (PLE) process uses elevated temperature and pressure to extract bioactive compounds from tea [215]. Due to the high temperature used, the structural bonds in the bioactive compound weaken, resulting in the rapid extraction of the selective bioactive compounds from the tea matrix [41]. The critical factors contributing to the extraction process include the high solubility of the bioactive compounds in the solvent and the high diffusion rate caused due to the weakening of the bonds [208, 216]. This extraction process has been explained in detail elsewhere [217-219]. The solvent forms a layer around the tea to enhance the desorption of the bioactive compounds from the matrix site of the tea leaves [220]. Furthermore, the bioactive compounds diffuse into the organic solvent and finally get distributed into the extraction phase. The efficiency of the extraction depends on the nature of the matrix and the compound, the location of the targeted bioactive compound inside the matrix [218, 220], solubility, and the diffusion rates [218, 221, 222]. Pressurized solvent extraction (PSE), pressurized hot-water extraction (PHWE), subcritical water extraction (SCWE), accelerated solvent extraction (ASE) technology, and superheated water extraction (SHWE) are different forms of PLE used for the extraction [123, 218]. PLE is typically used to extract and isolate caffeine and catechins from tea. The PLE had the highest recovery rates of target bioactives but also ensured a maximum degree of accuracy due to automation [215].

Temperature and pressure are major parameters of concern for PLE. The temperature in the process affects the efficiency and sensitivity of detecting the targeted bioactive compounds. Higher temperatures improve extraction efficiency by disrupting the bonds, eliminating interactions, and lowering the activation energy required for the desorption process. It also decreases surface tension by altering the sample's wettability and solubility [218]. However, increasing the temperature might impact negatively by extracting non-targeted bioactive compounds, resulting in decreased selectivity. On the other hand, elevated pressure in the process affects the solvent's boiling point, causes cell disruption, and enhances the mass transfer rate. The elevated pressure also assists in controlling the challenges related to the bubbles formed within the matrix that hinder the solvent from reaching the bioactive compounds. This boosts the bioactive compounds' solubility and desorption rate [218].

Several experimental procedures to extract catechins [28, 29, 41, 223], epicatechins [41], caffeine [28, 29, 224–226], phenolics, ligands, carotenoids, oils, lipids, essential oil, and nutraceuticals [218] using PLE from tea has been reported. Various types of tea products like green tea [223, 225, 226], black tea [41], fermented and non-fermented teas [41], mate leaves [144, 216], herbal tea [215, 218], and eagle tea [215] were studied. Solvents such as water [41], methanol [41, 144], ethanol [29, 41], ethyl acetate [41, 144], ethyl lactate [28, 29], n-hexane, toluene, dichloromethane, acetone have been also carried out with PLE [144].

For the extraction processes, the temperature range was between 275 °C [28, 29, 41, 144, 215, 216, 226] to a lowest at 40 °C [225] and some maintaining high pressure to reach 275 °C [224]. The best solvents that can be used for PLE include methanol [41, 144, 224], water [28, 226], ethyl lactate [28, 29], n–hexane [227] for enhanced extraction of bioactive compounds. When a combination of water and ethyl lactate was used as extraction solvents, the yields were 3.5 times and 1.5 times higher than using them separately with water and ethyl lactate. PLE also aided in 26–36% removal of catechins from the tea leaves [28] with 53–76% caffeine recovery. In terms of recovery rates, there was 3.21% for catechin and 2.96% for epicatechin from black tea [41].

Most of the extraction time ranged from a lowest of 5 min [215, 227] to a highest of 20 min [28, 29, 41, 144, 216, 225, 226]. However, a study with micro pressurized extractions where the treatment required only 20 s (5–100 mg sample) for rapid solid sample analysis [224]. The optimized pressure also had a range between 3 and 10 MPa [29, 41, 144, 215, 216, 224–227].

PLE was demonstrated to be an alternative method for the extraction of caffeine [225] but not great for tea because it squeezes out the soft leaf matrix making the diffusion of caffeine and hindering the penetration of the solvent into the matrix. Ethyl lactate was a suitable solvent for isolating caffeine from green tea leaves [28, 29]. PLE is considered a selective method of extraction since it reduces the co-extraction of catechins during the selective extraction of caffeine [28]. The use of high temperature enhanced the extraction of polar compounds [144, 216], and the polarity of the solvent also had a higher effect on the extraction process [216]

The extraction process of PLE is analogous to the liquid– liquid extraction process [226]. Some studies showed no significant changes in palmitic acid, phytol, stearic acid, squalene, and vitamin E extraction from tea [144]. A significant advantage of PLE is that it prevents tea leaves from being damaged by oxygen and light during the process, and it is highly automated to ensure precision [215] and rapid extraction results for complex samples. PLE could also be used for the regular detection and extraction of pesticide residues faster and simpler [227]. PLE is better and faster with a lower sample and solvent technique [224]. Although it has many advantages, one of the significant drawbacks of PLE is thermal degradation due to elevated temperature [215]. The results of PLE application under different processing conditions to extract-specific bioactive compounds from various species of tea are shown in Table 5.

# Innovative Processing Technologies: Advantages and Drawbacks

Summarizing all the technologies, UAE has been proven to have higher extraction efficiency at lower temperatures. The most critical parameters affecting extraction in UAE include sonication power, frequency, solvent-to-solid ratio, temperature, and sonication time [146, 152]. One of the limitations of using UAE is when the sample is exposed to UAE for a longer time, it can significantly affect the process since it generates heat, which leads to the decomposition of the thermo-sensitive compounds. Thus, selecting an appropriate sonication time plays an important role in the extraction process [228]. Conversely, MAE is considered a rapid alternative method that couples microwave heating with chemical extraction techniques. MAE is most employed for extraction and pre-treatment, wherein time, temperature, and pressure play the most influential role in the process. The major advantage of using MAE is to extract healthyfunctional constituents from tea, resulting in higher extraction yield with a shorter time, lower energy consumption, and higher extraction selectivity.

PEF serves as an effective method that is more specific for the extraction of intracellular compounds without the use of heat and pressure. In addition, it can be used as a method for the inactivation of the microorganisms present in the tea, depending on the intensity of the electric field strength applied. The effectiveness of the process depends on factors such as the electric field intensity, pulse wave shape, pulse frequency, polarity, solvent selection, the ratio of solute to solvent, pulse duration, and the treatment temperature and time. The electroporation mechanism assisted the extraction to a greater extent with the optimized conditions. The extraction efficiency of PEF falls in the range of 27 to 32%. PEF can be a promising method for large-scale extractions and for extending tea extracts' shelf life. In addition, PEF can help preserve the color of the product and its bioactive properties without affecting the properties.

Comparatively, SFE was more specific for the extraction of caffeine from tea. The critical parameters in SFE include the temperature and pressure, the flow rate of  $CO_2$ , and the matrix composition of tea leaves. This method assisted in the highest retention of catechins in the cells rather than their extraction. SFE is regarded as a method for extracting phytochemicals, and the recovery and extraction rate are found to be more than 90% with a longer extraction time. This method of extraction is found to be better with the use of organic solvents. SFE might be an expensive extraction method compared to UAE, PEF, PLE, and MAE.

Furthermore, PLE can rapidly extract the targeted bioactive compounds, as there is a higher chance of improved wetting of the molecules present inside the tea leaf matrix by the organic solvent. The temperature and the pressure play an influential role in the extraction process. The higher temperature and pressure improved the solubility of the targeted bioactive compound during the extraction process. The method uses a high temperature, thus making it unsuitable for thermo-sensitive compounds. It helps in the faster extraction of bioactive foods like caffeine and catechins. However, reagents and pressure preparations are expensive to be used on a large scale in the industry; hence, further research is needed for determining alternative cheap reagents.

From the review, it can be concluded that SFE is most widely used for the selective removal or extraction of caffeine and has an extraction efficiency of ~> 80%. Moreover, when catechins and caffeine need to be extracted with higher recovery rates, PLE is commonly preferred since it has a recovery range of 50 to 90% for catechins and 26 to 95% for caffeine. MAE and PEF were employed for maximum extraction yields for total phenolic compounds (~ 35%). In addition, PEF was also used for improving the extraction of free amino acids. UAE provided favorable conditions for the extraction of catechins and flavonoids, including theobromine, theophylline, and theaflavins.

Considering the review, each method of extraction has advantages in specific ways. The mass transfer is increased due to the bond breakage between matrix and bioactive compounds for all the extraction methods. The extraction yield SFE ranks higher than other methods discussed but with the disadvantage of longer extraction time. MAE and UAE are the two best extraction methods with water as a solvent and high extraction efficiencies. Both methods have lower extraction time for phytochemicals; however, based on temperature, MAE could be used as an extraction method if a higher temperature is preferred, and UAE can be used if a lower temperature is preferred. PEF, on the other hand, could be opted to increase the extraction efficiency and retain the bioactives with an increased color retention and shelf life. SFE and PLE could be effective methods for selectively extracting caffeine. For industrial-scale applications, PEF, PLE, and SFE can be used as a continuous process

Table 5 Summary of $\epsilon$	extraction of bioactive compounds obtained fi	rom tea under various PLE conditions		
Sample	Extraction conditions*	Extracted compounds	Keynote (s)	Reference
Tea (non-fermented and fermented teas and black teas)	100–200 °C, 10.1 MPa (100 atm), methanol, 10 min	Catechins and caffeine	<ul> <li>No significant effect of high temperature on the stability of catechins</li> <li>Methanol: the best used pure solvent</li> <li>Reduced recovery level by 95% for catechins and epicatechin at 130 °C</li> <li>The highest recovery rate with a relative standard deviation of 3.21% and 2.96% respectively for catechin and epicatechin</li> </ul>	Piñeiro et al. [41]
Sambucus nigra L. flowers, green tea, black teas, and coffee beans	100 °C, 6.07 MPa (60 atm), water, 10 min, 0.5 g	Rutin and caffeine	<ul> <li>Single-step PLE: Successful method to save time instead of a multi-step PLE in various ratios of the solid to solvent -Extraction yield of Rutin and Caffeine was 29.03 mg/g and ~ 35 mg/g</li> </ul>	Dawidowicz and Wianowska [226]
Green tea	70 °C, 4.05 MPa (40 atm), water, 10 min, 0.5 g	Caffeine	-Squeezing the soft matrix in tea at combinations of high pressure and temperature makes it difficult to extract caffeine from tea -16.24 mg/g of caffeine extraction using PLE in comparison to 34.5 mg/g for conventional methods	Dawidowicz and Wianowska [225]
Mate tea	100 °C, 10.3 MPa (102 atm), methanol, 10 min, 7.5 g	Caffeine, palmitic acid, phytol, stearic acid, squalene, and vitamin E	<ul> <li>Substantial amounts of caffeine and palmitic acid in the obtained extracts hainmal extraction time with the highest yield compared to the other methods (e.g., UAE, MAE)</li> <li>Extraction of more polar compounds at elevated temperatures</li> <li>Extraction yield of caffeine was 848.9 mg/kg of sample and mass yield was 23.1 g/100 g using PLE</li> <li>Methanol was the best solvent used for extraction</li> </ul>	Jacques et al. [144]
Mate tea	100 °C, 10.3 MPa (102 atm), methanol, 10 min, 7.5 g, 100 °C, 102 atm, hexane, 10 min, 2.5 g	Caffeine, phytol, squalene, vitamin E, caffeine, palmitic acid, and 37 other chemical compounds	<ul> <li>Solvent polarity, the sample amount, and extraction temperature had the highest effect on the quality and quantity parameters</li> <li>Significant difference in the extraction yield between methanol (13.83%) and hexane (1.67%)</li> </ul>	Jacques et al. [216]

Table 5 (continued)				
Sample	Extraction conditions*	Extracted compounds	Keynote (s)	Reference
Green tea	100 °C, 10.3 MPa (102 atm), <i>n</i> -hexane, 5 min	Flufenoxuron, fenitrothion, chlorfluazuron, chlorpyrifos, hexythiazox, methidathion, chlorfenapyr, tebuconazole, O-Ethyl- o-4 nitrophenyl phenulphosphotionate, bifenthrin, cyhalothrin, spirodiclofen, difenoconazole, and azoxystrobin	<ul> <li>Bifenthrin was the only pesticide identified in the tea samples</li> <li>Faster and simpler extraction method for bioactive compounds from tea samples</li> </ul>	Cho et al., [227]
Green tea	200 °C, 3.57–20.2 MPa (35–200 atm), simple alcohols, 5–15 min	Phenolic compounds, ligands, carotenoids, oils and lipids, essential oils, and other nutraceuticals	Promising method to extract carotenoids from tea	Mustafa and Turner [218]
Herbal tea	50–200 °C, 3.44—20.67 MPa (34– 204 atm), organic solvents, 5–10 min	Flavonoids, catechins, chlorogenic acid, and epicatechin	-Quicker and precise photochemical analysis for extracts obtained with the fast technique of PLE (5 min) compared with other extraction ones (8 h) - PLE: best extraction method with high repeatability for the maximum yield at lower extraction time and solvent consumption	Zhao et al. [215]
Green tea	99.85–199.85 °C, 9.92 MPa (98 atm), ethyl lactate and water, 20 min,1 g	Caffeine and catechins	<ul> <li>Higher solubility of caffeine in mixtures of ethyl lactate and water at the optimum of pressure and temperature values</li> <li>Extraction yields using the combined solvent of water-ethyl lactate were 3.5 and 1.5 times more than when water and ethyl lactate</li> <li>Higher recovery rate for caffeine (53–76%) compared to that of catechins (26–36%)</li> </ul>	Bermejo et al. [28]
Green tea	100 °C, 9.92 MPa (98 atm), ethyl lactate and ethanol, 20 min, l g	Caffeine, catechins, and other phenolics	<ul> <li>Ethyl lactate was the best solvent for the decaffeination process</li> <li>Precipitates obtained by ethyl lactate solvent were 2.3 times more than those of ethanol</li> <li>93% reduction in caffeine content</li> <li>Percentage recovery of main catechins (EGCG) was in the range of 46–74%</li> </ul>	Bermejo et al. [29]
Green tea	275 °C, 15.19 MPa (150 atm), methanol, 20 s, 0.005–0.1 g	Caffeine	- Higher recovery of caffeine at higher temperatures -Micro-PLE method: fast extraction method with low sample and solvent compared with the conventional methods	Alkhateeb and Thurbide [224]

for extraction, whereas UAE and MAE can only be used as a batch process. In addition, SFE, PLE, and PEF also require high initial capital investments. Comparing all the technologies, both UAE and MAE are two technologies that can have a broader spectrum of commercialization in terms of acquiring process equipment and, at the same time, attaining higher extraction efficiency.

The review overall had a broader view of the novel methods used to extract bioactive compounds from tea. With all the conventional techniques used for the extraction process from tea, it was evident that the novel processing technologies provide better results owing to technological advancements. However, each method has different advantages, which are specific to the method and have fewer limitations for the extraction process. However, the study is insufficient to conclude one extraction process, as many variables, such as the type of the sample, the experimental conditions, and many other process parameters, play a major role in selecting an extraction process. More extensive studies should be directed toward the extraction techniques, and comparative studies with other novel extraction technologies need to be carried out from the quantity and quality viewpoints.

## **Conclusions and Future Directions**

Over the last two decades, novel innovative processing technologies (e.g., UAE MAE, PEF, SFE, and PLE) have been used as an alternative technology to replace conventional extraction methods due to their high efficiency and effectiveness in extracting bioactive compounds from tea. Several studies discussed in the present review have highlighted the application of these novel technologies to various teas. Applying these technologies over conventional solvent extraction techniques improves extraction time and temperature, the number of used solvents, and extraction efficiencies. Moreover, food-processing industries are taking sustainable initiatives to fully utilize the possibility of environmental-friendly novel technologies for extraction.

The application of novel technologies for the extraction of bioactive compounds from tea would provide a sustainable solution for tea industries and generate value-added functional ingredients that have commercial value. Additionally, novel-processing technologies might be used to tailor foods with added or enhanced functional and nutritional values, lowering the carbon footprint and substantially reducing the water volumes used in industrial heat transfer processes.

The biggest drawback to applying novel technologies could be consumer acceptance, capital investments, and reproducibility. During the extraction process, the food matrices are subjected to various combinations of pressure, time, and temperature as the main parameters involved in the extraction technique. Improper application of process

Table 5 (continued)				
Sample	Extraction conditions*	Extracted compounds	Keynote (s)	Reference
Tea leaves wastes	140 and 200 °C	Bioactive compounds	-Sequential combination of (microwave hydro-diffusion and gravity, UAE, and autohydrolysis) technologies represents an excellent method -Recovery of total phenolic compounds was ~ 50 mg GAE/g extract. The total antioxidant level ranged between 0.3 to 0.9 g Trolox/g extract	Sanz et al. [2]
Green tea	80 °C, 100 bar, Water, Ethanol: water, Ethanol, Methanol, Ethyl acetate, Acetone: water, 3–10 min	Catechins	<ul> <li>Total amount of catechins and gallic acid was 8.02–505.47 mg/200 mL</li> <li>Highest recovery of catechins</li> </ul>	Koch et al. [223]
*1- Temperature (°C),	2- Pressure (MPa/atm), 3- Solvent, 4- Time (	min), 5- Sample size (g)		

parameters can strongly initiate Maillard reactions, leading to the formation of carcinogenic substances. Hence, every food sample needs to be studied uniquely, and the process variables should be optimized. Also, the functionality of the bioactive compounds extracted using various novel techniques must be examined before commercial approval. In brief, novel technologies can produce high-quality bioactive compounds with minimal environmental impact. Even though high investments are generally required to carry out-tailor made research by industries on these novel technologies, the results from fundamental research are very promising.

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### Declarations

Conflict of Interest The authors declare no competing interests.

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