



## Review article

## Recent advances in the extraction methods of essential oils and oleoresins from plant materials and its potential applications: A comprehensive review

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### ABSTRACT

Due to the important role of bioactive secondary metabolites in cosmeceuticals, functional foods, nutraceuticals, therapeutics, and health care, more attention has been paid to the extraction of active components using efficient extraction methods. Efficient, economical, and promising extraction routes are essential for resource recovery without compromising the quality and efficacy of the extracted products for desired applications. The present review emphasizes the recent developments and improvements in extraction methods of essential oils and oleoresins that have been achieved through different and innovative processing techniques and their diverse applicability. Besides, the trends of extraction techniques, the effect on essential oil glands in different extractions, industrial relevance, and optimization design were found important to industry professionals in realizing wider possibilities for selecting suitable extraction methods and developing new sustainable bioactive compounds-based formulations. In addition, this review facilitates the academics to select the effective extraction method in their research as extraction is the first and crucial part of plant-based research. The study revealed that the extraction methods developed using the combinations of novel technologies such as microwave assisted, ultrasound assisted, enzyme assisted, and supercritical fluid extraction were effective and efficient for the extraction of both essential oils and oleoresins. The extracts' applications are widening due to the advance in the delivery systems, such as micro and nanoencapsulation technologies.

**Keywords:** Essential oils; Oleoresins; Extraction; Conventional routes; Modern routes

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## 1. Introduction

The extraction is a crucial and fundamental step in numerous processes, especially in extracting essential oils and oleoresins for applications such as flavours and fragrances. They are further isolated as bioactives used in cosmeceuticals, functional foods, therapeutics, and healthcare. Essential Oils (EOs) and Oleoresins (ORs) are extracted using diverse and various methods by researchers and industrial organizations in botanical extraction. The conventional routes of extraction of EOs and ORs are developed into novel routes for minimizing the cost of production, improving the extraction efficiency and enhancing the quality of the EOs and ORs. Most modern routes are introduced as greener technologies as

they minimize energy consumption, emission of carbon dioxide to the environment, and the usage of toxic solvents. In addition to that, the route of the extraction greatly influences the composition of the EOs and ORs qualitatively and quantitatively (Tavakolpour et al., 2016).

Moreover, the EOs and ORs are highly used in the food and beverage industries as flavours, fragrances, preservatives and other additives. The major constraint of applying natural EOs and ORs is the high price compared to synthetic products. Hence, efficient, economical and promising extraction routes are essential for that particular resource recovery without compromising the quality of the extracted products for desired applications in the academic and industrial fields (Belwal et al., 2018; Bhushan et al., 2019). Due to its significance, many recent reviews focused on the modern and

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conventional routes of extraction from plants in a general way (Naboulsi & Aboulmouhadjir, 2018). Picot-Allain et al. (2021) extensively reviewed the green extraction methods of all extracts without focusing only on the essential oils or oleoresins. Some reviewers had done thorough studying of EOs without considering the nonvolatile part. In our review, the novel and conventional extraction methods of both EOs and ORs are considered as EOs and ORs are related, and the same raw materials are used for extracting volatiles in EOs and non-volatiles in ORs. In addition, this is the first attempt to compare and review the SEM images of the oil glands in different extraction methods.

In this review paper, many different extraction methods are discussed, alone or in combination, that has been exploited to produce the EOs and ORs from plant materials with consideration of the pros and cons and the mechanism of each extraction method. The widely used conventional routes such as hydrodistillation, steam distillation, solvent extraction, enfleurage, cohobation, maceration and some of the novel routes such as ohmic assisted hydrodistillation, microwave assisted hydrodistillation, solvent-free microwave assisted extraction, microwave hydro diffusion and gravity, microwave steam distillation, molecular distillation, fractional distillation, ultrasound-assisted extraction, microwave-assisted extraction, supercritical fluid extraction, enzyme assisted extraction are reviewed. This study contributes to deepening the understanding of the conventional, innovative and modern processing techniques to extract essential oils and oleoresins from different plant materials by summarizing the recent research studies. Moreover, the effect on the oil glands by different extraction conditions was evaluated using Scanning Electron Microscope (SEM) images. In addition, this review further addresses the applications of EOs and ORs, the current status, challenges and future trends of the extraction field.

## 2. Description of the extractives

### 2.1. Secondary metabolites (SMs)

SMs are the products or intermediates which are biosynthesized using one or more primary metabolites in multiple biochemical pathways and are not directly essential for basic life functions such as the growth and development of the plant. Nevertheless, SMs play an imperative role in the fitness of the plant through the interaction of the cell with their environment and ensure continued existence in their ecosystem through the defence system. The SMs, including terpenes (isoprenoids), phenolic compounds (flavonoids and phenylpropanoids) and nitrogen-containing compounds (cyanogenic glycosides, alkaloids, and glucosinolates), are synthesized in different pathways such as shikimic acid pathway, malonic acid pathway, mevalonic acid pathway, methylerythritol 4-phosphate (MEP) pathway using primary metabolites of erythrose-4-phosphate, phosphoenolpyruvate, pyruvate, 3-phospho-glycerate (War et al., 2019; Twajj & Hasan, 2022). SMs contribute to the plant by protecting from pathogenic microorganisms (phytoalexins) by their antibiotic, antifungal, and antiviral bioactivities, defending from other plants by anti-germinative and toxic properties, being safe from harmful leaf damage from the light by making UV absorbent compounds, protecting from herbivores-predators by anti-feeding properties, ensuring plant survival and growth in stressful environments caused by abiotic factors such as soil, temperature stress, light intensity, salinity, drought, chemical stress, and biotic factors such as pathogenic attacks (Mendoza & Silva,

2018; Ashraf et al., 2018). Hence, these supernatural compounds are used as natural pharmaceuticals, nutraceuticals, food preservatives, flavours, fragrances, colours, agrochemicals, biopesticides, cosmeceuticals, insecticides, herbicides, and industrial additives. Furthermore, the various usage in these applications is enhanced further due to the regard of SMs as “Generally Recognized As Safe” (GRAS) compared to synthetic additives and bioactives (Desai & Parikh, 2015).

### 2.2. Essential oils and oleoresins

Amidst the SMs, Essential oils (EOs) and Oleoresins (ORs) are highly significant as they are applied to food, fragrance, flavour, nutraceutical and pharmaceutical industries. EOs are concentrated hydrophobic lipophilic liquids consisting of complex combinations of volatile compounds extracted from aromatic plants and in lower molecular weights. Currently, there are 3000 essential oils, and 300 of them are commercially important products (Bhavaniramy et al., 2019).

Essential oils are used for flavouring agents in food and beverages, fragrances in cosmetics, aromatherapy, and bio-actives in pharmaceuticals. The antioxidant, antimicrobial and anti-inflammatory activities of essential oils are drawn great interest in the food, cosmetic, and pharmaceutical industries due to their potential use as natural additives. They are obtained from plant materials such as flowers, buds, seeds, leaves, twigs, bark, herbs, wood, fruits, and roots. The chemical constituents of EOs can be classified into two major categories: terpenes and phenylpropanoids. The terpenes are mainly divided into hydrocarbons such as monoterpenes, sesquiterpenes, diterpenes, and oxygenated compounds (terpenoids) such as aldehydes, ketones, phenols, acids, esters, and lactones. EOs are biosynthesized from three pathways; the mevalonate pathway (sesquiterpenes), methylerythritol pathway (monoterpene, diterpenes), and the shikimic acid pathway (phenylpropenes) (Moghaddam & Mehdizadeh, 2017).

Oleoresins are liquid or semi-solid viscous resin-like materials obtained by the extraction of spices. They are the true essence of the spices and are 5-20 times stronger compared to crude spices. They mainly consist of a small amount of volatile EOs, and mostly nonvolatile compounds such as fixed oils, pigments, resins, and pungent constituents. They contribute to the spice's aroma, flavour profile, and bioactive properties.

### 2.3. The extraction of EOs and ORs

Various EOs and ORs extraction methods have been used for many years. For instance, distillation for obtaining EOs was done in the East (Egypt, Persia, and India), and the written documents were reported in the 12<sup>th</sup> century by Villanova; Catalan Physician (Guenther & Althausen, 1948). These extracts were obtained by solid-liquid extraction from the plant matrix using conventional routes such as maceration, cohobation, hydro distillation, steam distillation and solvent extraction. Gradually, the conventional routes of extraction methods were developed and improved to increase the extraction yield, efficiency, and quality of the extract through lowering thermal and hydrolytic effects. In addition, it is expected to minimize the degradation of unsaturated or ester compounds, lower the residues of the toxic solvents, reduce the extraction time, and minimize the loss of the volatiles (Jeyaratnam et al., 2016).

Furthermore, operational cost reduction is a primary objective that can be achieved by minimizing energy consumption with continuously rising energy prices. Researches on innovative technological extraction methods are highly significant in environmental aspects since they lead to the reduction of CO<sub>2</sub> emissions. Other than the extraction method, the factors which

influence the extraction are the part of the plant, the time of maturity of the plant part, the method of obtaining the plant part, the particle size of the plant materials, and some pre-treatment methods such as removing outer layers, drying for appropriate moisture levels, pulverizing, crushing, grinding, powdering and soaking of the plant materials (Belwal et al., 2018).

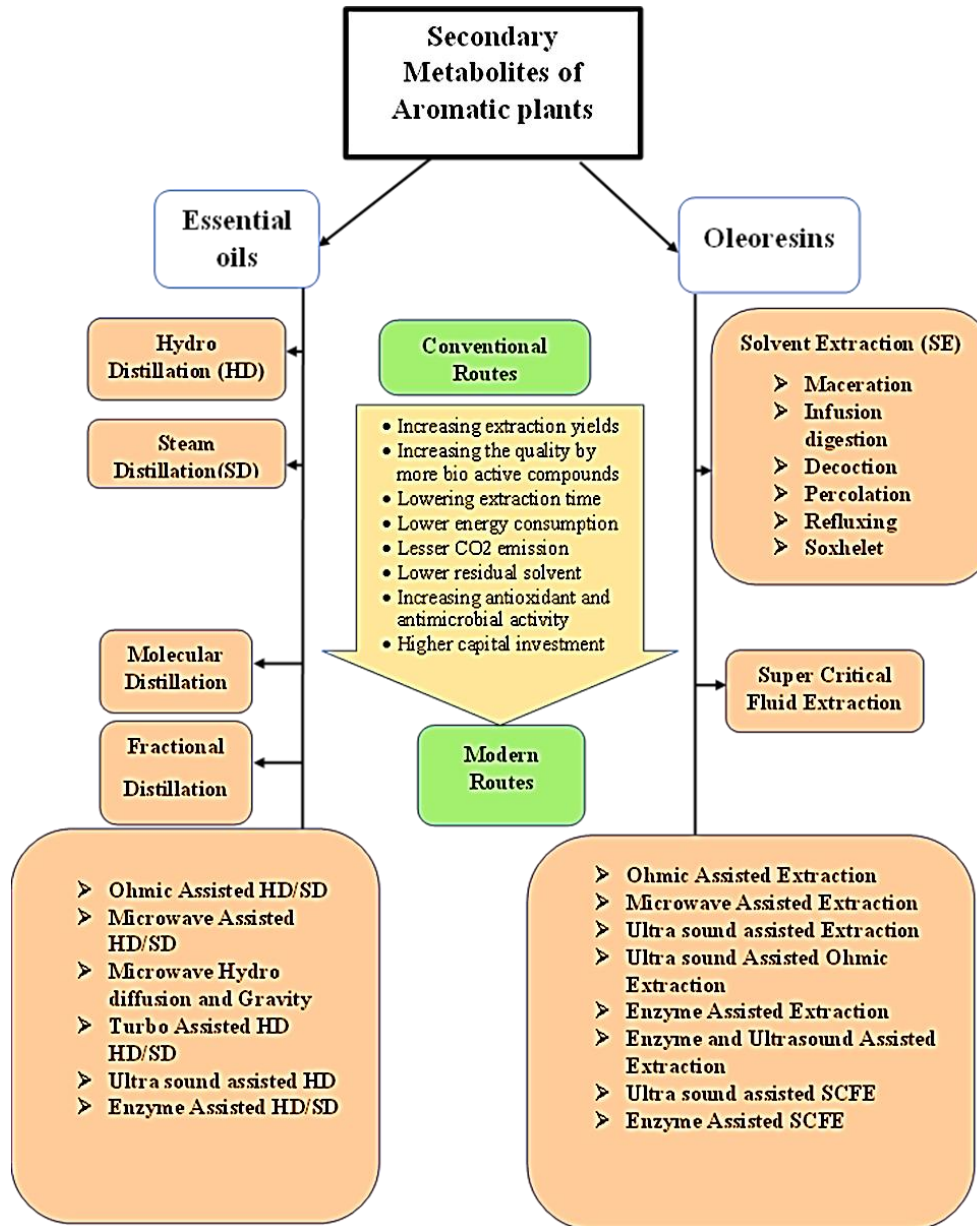


Fig. 1. Schematic Diagram of extraction methods of Essential oils and oleoresin.

Table 1. Comparative studies of modern extraction routes with conventional extraction routes.

Modern Extraction method	Conventional Extraction method	Plant	Results of comparison of the modern extraction methods and conventional extraction methods	References
UAHD	HD	<i>Cinnamomum cassia</i> bark	Higher extraction yield (27%) in UAHD (2.14%) compared to HD (1.68%). Lower extraction time of UAHD (60 min) compared to HD (120 min), High trans-cinnamaldehyde content (81.9%) in UAHD compared to HD (78.3%) Lower energy consumption for UAHD (0.79 kWh per 1g of EO) compared to HD (1.49 kWh per 1g of EO) Lesser carbon dioxide emission in UAHD (0.63 kg) compared to HD (1.19 kg)	(Chen et al., 2021)
UAE	Maceration, SXE	<i>Curcuma domestica</i> , <i>Curcuma xanthorrhiza</i>	Higher extraction yield in SXE for <i>C. domestica</i> ( $18.89 \pm 0.68\%$ ) and <i>C. xanthorrhiza</i> ( $14.70 \pm 0.62\%$ ). Highest curcumin yield in maceration for <i>C. domestica</i> ( $49.52 \pm 0.63\%$ ), <i>C. xanthorrhiza</i> ( $15.82 \pm 0.27\%$ ) Lowest residual solvent in UAE for both <i>C. domestica</i> ( $16.00 \pm 4.67\%$ ) and <i>C. xanthorrhiza</i> ( $15.66 \pm 3.33\%$ ) <i>C. xanthorrhiza</i> oleoresin obtained from maceration tends to show the best colour, while the best colour of <i>C. domestica</i> oleoresin is obtained from UAE.	(Nurhadi et al., 2020)
MAHD	HD	<i>Coriandrum sativum</i> L (Coriander)	Extraction yields were recorded as $0.325 \pm 0.021$ in MAHD compared to $0.31 \pm 0.014\%$ in HD EO extracted by MAHD showed better antimicrobial activity, higher phenols yield and antioxidant activity compared to HD	(Ghazanfari et al., 2020)
HD after RF, HD after RF with enzymes, HD after RF with UltraSound	HD	<i>Salvia officinalis</i> L (Sage), <i>Rosmarinus officinalis</i> L (rosemary), <i>Laurus nobilis</i> L (bay laurel)	Extraction yield increase (40–64%) was recorded in all modern methods compared to HD Significant yield increase HD after RF (Soaking the ground plant material at 40°C for 1 hour) significantly increased the yield)	(Miljanović et al., 2020)
MAHD, SE, MAE, UAE, SCFE	HD	<i>Mentha piperita</i> L. (peppermint)	MAHD is an alternative for HD in EO recovery, SCFE is better as a green method of producing terpenoids and other lipophilic bioactives-rich extracts	(Pavlič et al., 2021)
MAHD	HD	<i>Cinnamomum cassia</i> bark	MAHD produced more oxygenated compounds (9% higher) compared to HD, Reduction of carbon dioxide emission in MAHD (by 59%), EO of MAHD showed a lower lethal concentration 50 (LC50) value of 51.2 mg/L compared to HD (68.9 mg/L)	(Jeyaratnam et al., 2016)
OE, UAHD, UAOE	HD	<i>Thymus daenensis</i>	Extraction time was reduced in UAOE (31 min) compared to HD (95 min), Extraction efficiency was highest in UAHD and lowest in OE, The extraction yield increased in UAOE compared to HD	(Tavakolpour et al., 2016)
EAHD, EAE	HD, SE	<i>Laurus nobilis</i> L (Bay Leaves)	An increase of 243% in EO yield, higher content of monoterpenes, and higher antioxidant activity were recorded in enzyme pretreated samples	(Boulila et al., 2015)
MAE (Using MAE unit)	HD	lemongrass	A higher extraction yield of 1.85% was recorded in MAE compared to 1.8% HD The extraction time was reduced to 45 min in MAE compared to 90 min in HD Loading capacity was doubled in MAE than HD due to no inclusion of water Improved antimicrobial activity and antioxidant activity in EOs of MAE compared to HD	(Desai & Parikh, 2015)
MAHD	HD, SD, SE	<i>Melissa officinalis</i> L	MAHD reduced the extraction time to 20 min with 0.3% extraction yield, while SE increased the yield to 0.56% in 9 hours compared to HD (0.24% extraction yield in 180 min) and SD-0.42% (120 min)	(Abdellatif & Hassani, 2015)
MAHD	HD	<i>Navel Navelateoran</i>	Extraction Yield increased in MAHD ( $1.8 \pm 0.1\%$ ) compared to HD ( $1.7 \pm 0.1\%$ )	(Bustamante et al., 2016)

		ges(Fruit Peels)		
MAHD	HD	<i>Tanacetum polyccephalum</i> and <i>Artemisia chamaemelifolia</i>	Extraction time is shorter in MAHD(45 min) than in HD (150 min), No significant differences in extraction yield physical properties, antioxidant activities, and antifungal activities in both MAHD and HD	(Eblaghi et al., 2016)
MAHD	HD	<i>Rosmarinus officinalis</i>	Similar Extraction yields in both MAHD and HD, Reduced extraction time in MAHD (20 min) compared to HD (180 min), Lower electric consumption in MAHD (0.23 kWh) than HD (225kWh), Lesser Carbon dioxide releasing in MAHD(184g) than in HD(1800g) The essential oil contains a high amount of oxygenated compounds in MAHD	(Elyemni et al., 2019)
Coaxial MAHD	HD	<i>Lavandula angustifolia</i> (lavender), <i>Salvia Officinalis</i> L.(sage) and <i>Rosmarinus Officinalis</i> (Rosemary), <i>Foeniculum vulgare</i> (fennel seeds) <i>Syzygium aromaticum</i> L.(clove buds)	More energy and time saving, higher extraction of Monoterpenes in MAHD than in HD	(González-Rivera et al., 2016)
MAHD (SFME)	HD	<i>Citrus limon</i> (Lisbon variety) peel	Higher extraction yield in SFME (1.36%) compared to HD (0.22%) and MAHD (1.18%) Lesser extraction time duration in SFME (12 min) compared to HD (97 min) and MAHD (11.5 min) No significant differences in the physical parameters (refractive index, specific gravity, visual appearance, and colour) and the same compositions.	(Golmakani & Moayyedi, 2015)
MAE	HD, SD	<i>Anastaticahie rochuntica</i> (L)	Higher extraction yield in MAE (0.08%) compared to HD (0.05%) and SD (0.06%) Lesser Extraction time duration in MAE (30 min) compared to HD (4.5h) and SD (3h) More oxygenated compounds and lower monoterpenes and sesquiterpenes in MAE	(Abd El-Gaber et al., 2018)
MAHD	HD	Sandalwood	The oil yield is higher in MAHD than in HD Faster extraction in MAHD than in HD	(Kusuma & Mahfud, 2016)
MSD	SD	<i>Citrus auranticum</i> L(Orange Peel)	Higher extraction yield in MSD (0.65%-0.75%) compared to SD (0.59%-0.7%) Lower Extraction time duration in MSD (140 min) compared to SD (7 h) No significant variation of essential oil composition between MSD and HD and no adverse effects on the composition	(Kusuma et al., 2016)
MSD	SD	<i>Origanum glandulosum</i>	Higher extraction yield MSD (1.86±0.20%) compared to SD=1.84±0.25% Lower Extraction time duration in MSD (6min) and SD (30 min) More antioxidant activity and insecticidal activity in EO obtained by MSD than in SD	(Sahraoui et al., 2017)
MSD	SD	<i>Thymus pallelescens</i>	Higher extraction yield in MSD (2.45±0.1%) compared to SD (2.35±0.21%) Lower Extraction time duration in MSD (5min) compared to SD=20 min More antioxidant activity in EO is obtained by MSD than by SD Chemical composition is qualitatively comparable in MSD and SD	(Sahraoui & Boutekdjiret, 2015)
MAHD	HD	Lemongrass	Lower Extraction time duration in MAHD (90 min) compared to HD =360 min Higher extraction yield in MAHD (0.35%) compared to HD (0.15%)	(Hien Tran et al., 2019)
MAHD	HD	<i>Pogostemon cablin</i> (Patchouli leaves)	Lower Extraction time duration in MAHD (126 min) compared to HD =417 min Higher extraction yield in MAHD (2.72%) compared to HD=2.61% Higher in Total Oxygenated Compounds MAHD (38.15%) compared to HD (28.33%)	(Hashmi & Kim, 2015)
MAHD	HD	<i>Piper betle</i>	Lower Extraction time duration MAHD (50 min) compared to HD (210	(Amaresh et al.,

		<i>L.</i> (Betel leaf)	min) Extraction yield is higher, and the colour is lighter (Colorless to pale yellow) in MAHD compared to HD and No Significance difference between the composition and physical properties	2017)
OAHD	HD	<i>Mentha piperita</i> L.(Peppermint)	Extraction time was reduced to 20 min in OAHD compared to 56 min in HD.	(Gavahian et al., 2012)
OAHD	HD	<i>Thymus vulgaris</i> L.(Thyme)	Extraction time reduced to 24.75 min in OAHD compared to about 1 h for HD	(Gavahian et al., 2012)
OAHD	HD	<i>Myrtus Communis</i> (Myrtle)	Extraction time of 26.11 minutes while this value was more than 1 hour for HD for approximately the same amount of EO recovery (0.7% v/w)	(Taylor et al., 2013)
OAHD	HD	<i>Zataria multiflora</i> Boiss.(Shirazi thyme)	Lower Extraction time duration in OAHD (32.21 ± 2.59 min) compared to HD (57.21 ± 2.33 min)	(Gavahian et al., 2011)
Advanced MAHD	HD	<i>Thymus vulgaris</i> L.(Common thyme)	MAHD was superior in terms of saving energy and extraction time (75 min, compared to 4 h in HD)	(Golmakani & Rezaei, 2008)
SFMAE	HD	<i>Ocimum basilicum</i> L. (Basil), <i>Mentha crispata</i> L.(Garden mint), <i>Thymus vulgaris</i> L.(Thyme)	30 min for SFME method against 4.5 h for HD and less CO <sub>2</sub> rejected in the atmosphere	(Lucchesi et al., 2004)
UAE, ISMGHD, MSD, MHG, MSDF	HD, SD, THD	<i>Lavandin Grosso</i> (Lavandula intermedia var. Grosso)	The best extraction method is MHG, with a 5.4% yield 15 min extraction time (120 min for SD) and 1.3 kWh (against 8.06 kWh for SD) energy consumption.	(Périno-issartier et al., 2013)
SCFE	HD	<i>Ocimum basilicum</i> L. (Basil)	Extraction yield is higher in SCFE (0.45 %) compared to HD (0.82 %) Insignificant variation in physical parameters such as colour, solubility, density and refractive index Antioxidant activity, antibacterial activity(against <i>P. Multocida</i> ), and antifungal action (against <i>A. Flavusas</i> ) is higher in SCFE than in HD	(Abbas et al., 2017)
SFE	SE, HD	<i>loxophleba ssp.</i> Lissophloia(leaves of <i>Eucalyptus</i> )	Extraction yield is higher in Soxhlet Extraction (7.9%) compared to SFE (4.78%) and HD (3.77%)	(Zhao & Zhang, 2014)
SCFE	HD, SD	<i>Boswellia serrata</i>	Essential oil yield is higher in HD(8.18%) compared to SCFE (0.31%) Higher antioxidant activity in SCFE followed by SD compared to HD Higher antimicrobial activity in SD followed by HD compared to SCFE	(Ayub et al., 2018)
SCFE	HD, SD	Rosemary	Extraction yield is higher in SD (2.35%) compared to HD (0.35%) Antioxidant activity is 14 times better than HD and SD	(Conde-Hernández et al., 2017)
SCFE	SE	Thyme and rosemary	IR spectra reveal that the peaks intensities, positions, and shapes of the main specific bands in the spectra are the same.	(Topala & Tataru, 2016)
USAE	SE	<i>Zingiber officinale</i> R.(Ginger)	USAE is 1.75 times more rapid than SE	(Dani et al., 2011)
UAE	Maceration, SE	<i>C. domestica</i> and <i>C. xanthorrhiza</i>	Highest oleoresin yield in SE, highest curcumin yield in maceration method, lowest residual solvent and better colour in UAE	(Nurhadi et al., 2020)
UAHD	HD	<i>Cinnamomum cassia</i> bark	Lower Extraction time duration in UAHD (35 min) compared to HD (180 min) Extraction yield is higher in UAHD (3.17%) compared to HD (2.98%) Electric Consumption is lower in UAHD (0.291 kWh) in HD (1.14 kWh)	(Jadhav et al., 2020)

EASD	SD	<i>Foeniculum vulgare</i> (Fennel)	Increase oil yield in EASD (11 – 22.5%) as compared to SD Same volatile oil composition	(Baby & Ranganathan, 2016b)
MAE UAE EAE	SE	<i>Curcuma longa</i> <i>L</i> (Turmeric))	Higher Oleoresin Yield in Soxhlet Extraction (8.29%) followed by EAE (6.27%), MAE (5.19%), UAE (5.72%) Higher curcumin yield SE (6.90%) followed by (EAE4.1%), MAE(3.72%), UAE=3.92%	(Sahne et al., 2016)
EASD	SD	Cardamom seeds	Increase yield in EASD (7.23–7.83%) compared to SD (6.73%) 1, 8 cineol and terpinyl acetate increased from 34.3 to 37.6% and 40.8–42.3%, respectively, with enzyme pre-treatment	(Baby & Ranganathan, 2016a)
EAE	SE	<i>Capsicum annuum</i> <i>L</i> (Green chilli)	Higher extraction yield of oleoresin (5.1- 5.9%) compared to SE	(Baby & Ranganathan, 2016c)

Note: Abbreviations; Hydro Distillation (HD), Steam Distillation (SD), Solvent Extraction (SE), Refluxing (Rf), Soxhlet Extraction (SXE), Microwave Assisted Hydro Distillation (MAHD), Ohmic Extraction (OE), Ultrasound Assisted Hydro Distillation (UAHD), Ultrasound Assisted Ohmic Extraction (UAOE), Microwave Hydro Diffusion and Gravity (MHG), Microwave steam diffusion (MSDF), Solvent Free Microwave Extraction (SFME), Microwave Steam Distillation (MSD), Turbohydrodistillation (THD), In Situ Microwave Generated Hydro Distillation (ISMGHD), Ultra Sound Assisted Extraction (UAE), Ultra Sound Assisted Hydro Distillation (UAHD), Enzyme Assisted Hydro Distillation (EAHD), Enzyme Assisted Steam Distillation (EASD), Enzyme Assisted Solvent Extraction (EAE).

### 3. Extraction methods

#### 3.1. Developments of distillation

Distillation is based on heating the plant matrix to recover volatile compounds of plant materials and azeotropic distillation. The basis of the distillation is that evaporation happens when the sum of the vapours of water or steam and vapours of volatiles equals the ambient pressure close to 100°C. However, the volatile compounds consisted of higher boiling points. There are two types of EOs which have a higher density than water and a lower density than water. Therefore, after condensing water and essential oils, they can be separated using density differences. The conventional methods of distillation are hydrodistillation, steam distillation, and cohobation. There are new trends of distillation; altering the hydrodistillation techniques such as ohmic assisted distillation, ultrasound assisted distillations, enzyme assisted distillations, and various types of microwave assisted distillation to overcome the limitations of the conventional distillation (Fig. 1). At the industrial level, steam distillation and hydrodistillation are the most widely used methods for extracting essential oils.

In these methods, using water as the green extraction solvent has many advantages, such as non-toxicity, inexpensiveness, and environmental friendliness. Processing becomes convenient compared to organic solvents since the purification of products is facilitated due to the insolubility in the water at ambient temperature. The impairment of solubility is affected due to the polarity, density, and viscosity of the water which can be conquered by altering the physical conditions and the parameters of the extraction process (Filly et al., 2016). Managing the ionic dissociation of salts and the polarity depends on the macroscopic dielectric constant of the solvent. At high temperatures and pressures, the water has a lesser dielectric constant, which facilitates an increase in the water diffusivity and solubilizes more non-polar constituents. Therefore, some methods have been developed using that theory. Another approach is based on the hydrolysis of cell walls to facilitate the release of essential oils by using enzymes.

#### 3.1.1. Hydrodistillation based methods

##### 3.1.1.1. Hydro distillation (HD)

Hydrodistillation was used in the past due to the convenience and low requirement of specific conditions. In hydrodistillation, the material is dipped in the water, and due to the heat, the water vapour and the vapours of the volatiles form a heterogeneous mixture that gets its boiling temperature close to water boiling temperature, although volatile molecules have a higher boiling point (Asbahani et al., 2015). The mixture of vapours liquefies in the condenser, and the oils can be separated according to the density differences. It is required to filter the oil for further purification after adding dry anhydrous sodium sulphate.

The main advantages of hydrodistillation are convenience, lower set-up, and operational cost. Besides, compared to steam distillation, hydrodistillation leads to a higher extraction of the volatiles due to better penetration in plant materials since steam distillation forms lumps which are partially penetrable to the steam (Dilworth et al., 2017). There are limitations of hydrodistillation such as higher energy consumption, prolonged distillation time, loss of volatiles, degradation of thermo-labile compounds due to the elevated temperatures, high CO<sub>2</sub> emissions, decreasing antioxidant and antimicrobial properties and odour deterioration (Gavahian et al., 2012; Gavahian & Farahnaky, 2018) There are more shreds of evidence for the demerits of the hydro distillation in Table 1.

##### 3.1.1.2. Ohmic assisted hydrodistillation (OAHD)-modern route

The OAHD had been developed to overcome the limitations of hydrodistillation, such as higher processing time and energy consumption. The principle of this technique is based on ohmic heating (electro heating/ electroconductive heating/ Joule heating) and the traditional distillation process as described in hydrodistillation. OAHD can be considered a green technology since it uses less energy. Electrodes are used to form ohmic heating, and the apparatus consists of the same as hydrodistillation.

Furthermore, the joule effect facilitates internal energy transformation from electric energy to thermal energy inside the plant material since it acts as a resistor. The heating rate depends on electric conductivity, and a continuous water system is required for ionic mobility (Gavahian et al., 2015).

Sudden ruptured essential oil glands can be seen in SEM images of the OAHD, which accelerate the extraction process. The essential oil composition is the same in both extracted EO (Gavahian et al., 2011; 2012; 2015). As ohmic heating leads to a straightforward and rapid process, OAHD is not only a quick method of extraction of essential oils but also reduces the loss of volatiles due to the prolonged heating, saving energy, better process control, and further minimizes the operational cost (Gavahian et al., 2020). Nevertheless, the study by Gavahian and Farahnaky (2018) reported that the application of OAHD includes negative aspects such as electrical conductivity, operational safety issues, and the requirement for higher capital investment.

### 3.1.1.3. Microwave assisted hydrodistillation

The MAHD was developed by using the dielectric heating of the microwave for effective and selective heating and minimizing the thermal gradients. Hence, it facilitates a faster, environmentally friendly and more efficient extraction, enhancement of purity and lower degradation of volatile compounds (Hien Tran et al., 2019; Jeyaratnam et al., 2016; Liu et al., 2012). The modified microwave oven must be fitted to the hydrodistillation Clevenger apparatus in lab-scale distillation.

The composition of extracted EOs in MAHD is similar to essential oils in HD, although it is more environmentally friendly (Jeyaratnam et al., 2016). The principle is based on the dielectric heating of the microwave. The morphological analysis of MAHD extracted plant materials reveals the sudden and mild rupture of the oil glands, which leads to an efficient extraction (Golmakani & Rezaei, 2008; Jeyaratnam et al., 2016). This rupturing happens because of the conversion of the microwave energy to the heat energy of the water due to the high dielectric properties. Thus, the heat is transferred to the plant materials and distributed to release the EOs without damaging or rupturing the EO glands (Desai et al., 2010). The extraction process is faster in MAHD because the synergistic combination of heat and mass transport phenomena happens in the same direction, and the heat is dispersed inside the irradiated medium. Hence, the pressure difference between inside and outside the cells facilitates the mass transfer due to the easy release of the compounds into the medium (Kusuma & Mahfud, 2017; Hashmi & Kim, 2015).

Mostly, the extractions are done using microwave oven-type devices at the laboratory level. However, the scaling up of that approach is more expensive. Therefore, some novel technologies have been developed using a coaxial dipole antenna where electromagnetic energy is applied to the extraction medium. It was revealed that extraction using coaxial MAHD is inexpensive, convenient for scaling up, and safe (González-Rivera et al., 2016).

### 3.1.1.4. Some other modern technical modifications for hydro distillation

- Turbo hydrodistillation: The mixture of the water and plant materials is constantly stirred at a certain rpm for a certain period. This method is mostly used for hard plant materials such as wood and seeds (Filly et al., 2016; Périno-issartier et al., 2013).

- Salt-assisted extraction followed by conventional hydrodistillation (Salt-HD): Raw materials are mixed with water, and a certain amount of NaCl and HD is carried out.
- Enzyme-assisted extraction followed by conventional hydrodistillation (Enzyme-HD): Raw materials are mixed with water and specific enzyme at a specific concentration. The mixture is incubated for a certain time and at a specific temperature with stirring.
- Micelle-mediated extraction followed by conventional hydrodistillation (Micelle-HD): Raw materials is mixed with an aqueous surfactant solution containing 10% Tween 40.

### 3.1.1.5. Solvent free microwave assisted extraction (SFMAE)

Solvent-free microwave extraction combines dry distillation and microwave heating as MAHD. In this technique, the kinetics of the extraction is further increased, and the solvents are eliminated. The solvent-free microwave-assisted extraction does not facilitate the hydrolysis of essential oils due to using too much water as the medium because the SFMAE model operates without water (Bhushan et al., 2019).

In this method, plant materials are placed without solvent or water, neither microwave-assisted extraction (organic solvent usage) nor microwave-assisted hydrodistillation (water usage). The in-situ water inside the plant material is internally heated, and the distending of plant cells affects to rupture of EO glands and oleiferous receptacles. The distillate is cooled and condensed, and the excess water is refluxed to the extraction vessel to reinstate the in-situ water.

### 3.1.1.6. Microwave hydrodiffusion and gravity (MHG)

There are two routes of MHG; solvent-free microwave-assisted extraction mode working against gravity (similar to microwave-assisted hydrodistillation apparatus, and the main difference is the addition of water to the flask) and solvent-free microwave-assisted extraction mode in favour of gravity. The key difference is that the EOs are not required to evaporate to be condensed; however, after rupturing the oil glands, the extracts are drained due to gravity (Bhushan et al., 2019). MHG is very efficient compared to HD since heat transfer happens from outside boiling water to the inside of the plant material, while mass transfer occurs from inside to outside (Kala et al., 2017). In the case of MHG, natural water molecules in plant materials absorb microwaves and heat. This thermal stress leads to both phenomena of mass transfer and heat transfer from inside to outside, which will boost the extraction process.

### 3.1.2. Steam distillation-based techniques

#### 3.1.2.1. Steam distillation (SD)-conventional route

Steam distillation is a widely used method in the essential oil industry due to its convenience of controllability. In steam distillation, steam is generated and supplied to the vessel or apparatus of the plant material at a specific pressure. Then, the steam flows through the plant materials and the volatile molecules are released due to the exposure of materials to pores. The decomposition of the organic compounds can be controlled in



steam distillation until it reaches below the decomposition temperature of the constituents by controlling the temperature (Azmin et al., 2016). The limitations can be mentioned as degradation, and structural alterations of chemical compounds due to the high temperature especially compounds such as monoterpenes.

### 3.1.2.2. Microwave steam distillation

Although SD is widely used, it consumes more extraction time and energy. In addition, chemical compounds have structural alterations due to high temperature. To overcome the drawbacks of SD, microwave steam distillation has been introduced. The microwave oven is connected to the reactor (where the raw materials are placed) or cartridge of the general steam distillation apparatus. It is subjected to microwave heating (Sahraoui & Boutekedjiret, 2015). The saturated steam generated by the steam generator goes through the raw materials while continuously heated in the microwave zone. The release of the EOs from oil glands is facilitated by the saturated steam and direct contact with microwaves. The main advantages of this method are effective heating, selective extraction, increased extraction efficiency, and minimization of energy consumption (Kusuma et al., 2016).

### 3.1.3. Other modern techniques

#### 3.1.3.1. Molecular distillation

Molecular distillation is appropriate for extraction, separation, and purification of thermally sensitive compounds as it decreases the substance's boiling point due to the higher vacuum condition. MD is based on the molecular mean free path difference and is undertaken by a high vacuum, lower pressure, and lower temperature (Yang et al., 2017). Limited information was found in molecular distillation for getting purified essential oil fractions (Borgarello et al., 2015). In some studies, enrichment of some specific compounds from essential oil can be done using molecular distillation. For instance, thymol in residue fraction was higher than the subsequent molecular distillation for oregano essential oil (Borgarello et al., 2015).

#### 3.1.3.2. Fractional distillation

Fractional distillation is a process of separating two or more constituents using the difference in volatile properties of the constituents. Fractional distillation depends on temperature, pressure, and the physical and chemical properties of the constituents that are required to be separated. In some cases, fractional distillation is done parallel to the hydro or steam distillation, and some undertake separated fractional distillation under vacuum conditions using a column (Warsito et al., 2018). In some cases, batch distillation methodologies have developed (Almeida et al., 2018; Pires et al., 2019). As a lower temperature is used in fractional vacuum distillation, there is a minimum degradation of chemical constituents. The boiling point of the constituents is the major determinant for fractionation, and the fractions are obtained at different temperatures under lesser and controlled pressure (Nadeem et al., 2017).

## 3.2. Development of extractions

As oleoresin contains nonvolatile compounds and little amount of volatile, the extraction methods are based on dissolving extractives in a solvent. This solvent may be an organic solvent or supercritical fluid, called solvent extraction or supercritical fluid extraction. The modern routes were developed using novel technologies, as displayed in Fig. 1.

### 3.2.1. Solvent extraction

Solvent extraction (Solid-liquid extraction) is an old method which evolved in the 11<sup>th</sup> century by differentiating the conditions while mixing with a solvent. Those methods are maceration, infusion digestion, decoction, and percolation. Soxhlet extraction was developed in the 18<sup>th</sup> century. Powdered plant materials are mixed with a solvent in maceration; if the solvent is cold or boiling water, it is an infusion. During the digestion, the powdered plant materials are mixed with a solvent as maceration but with gentle heating. The decoction is a method of extracting by mixing powdered plant materials with water and boiling them. In percolation, the powdered plant materials are mixed with a solvent in a percolator (Belwal et al., 2018).

Solvent extraction is vital in extracting oleoresins as some specific bio-actives (such as piperine) are insoluble in water. Hence, their extraction is not feasible using the methods such as hydrodistillation (Dutta & Bhattacharjee, 2015). The solvent extraction process consists of several steps; penetration of the solvent to the matrix, dissolving the solute in the solvents and diffusing the solute from the matrix. After the extraction, the solvent is evaporated to get the extract by using an evaporator under vacuum conditions.

In addition to the extraction method, the extraction yield and the quality of the extract depend on the appropriate solvent type, the ratio of the solvent mixture, the temperature, the time duration, the number of extraction cycles, the counter-current extraction ways, the design of the extraction vessel and particle size of the raw materials. The most commonly used solvents are ethanol, acetone, hexane, ethyl acetate, methanol, isopropanol, and dichloromethane alone or in combinations with different ratios. The solvent selection must be made considering the solubility, polarity, selectivity, safety, and cost of the solvent. Solvents with a polarity value near the solute's polarity enhance the extraction according to the law of similarity and inter-miscibility. A higher solvent-to-solid ratio increases the extraction by raising the extraction yield.

Nevertheless, a more solvent-to-solid ratio leads to excessive extraction and higher cost due to the extension of the time for solvent evaporation. Higher temperatures lead to high solvent loss, degradation of thermo-labile components, and unwanted impurities. The higher extraction time duration boosts extraction efficiency until it gets the solute equilibrium inside and outside the plant materials. Therefore, increasing the number of extraction cycles and developing counter-current extraction steps are used on an industrial scale. The particle size is another crucial factor; in general, smaller particles facilitate extraction efficiency by escalating the penetration of solvents and diffusion of the solutes. The main drawback is that the finest particles will cause the excessive absorption of the solute in solid and difficulty in later filtration (Zhang et al., 2018).

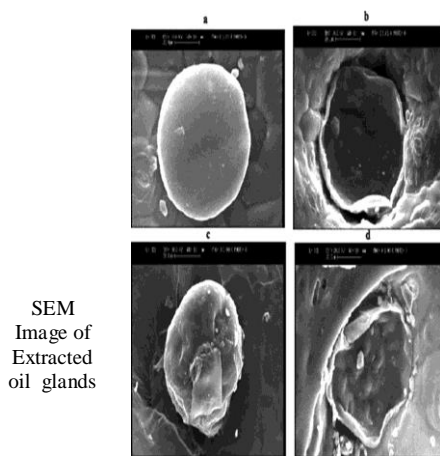
Table 2. SEM Images of plant materials after different extractions.

Plant Material	<i>Thymus daenensis</i> leaves	<i>Cinnamomum cassia</i>	Cardamom seeds
Extraction Methods	Hydrodistillation (HD) Ohmic Extraction (OE), Ultrasound-assisted hydrodistillation (UAHD) Ultrasound-assisted Ohmic Extraction (UAOE)	Microwave-assisted hydrodistillation (MAHD) Hydro distillation (HD)	Enzyme Assisted Steam Distillation (Viscozyme L, Pectinex Ultra SP-L, Celluclast 1.5L, Protease) and Steam Distillation (Control) and Cold and hot water treated steam distillation.

Affecting to the oil glands  
 In the OE method, oil glands are disjointed in every way, and many openings result across the whole wall of the oil glands compared to the HD. In Ultrasound Assisted extractions, slight damage and shrinkage in the cell wall of oil glands are taken place.

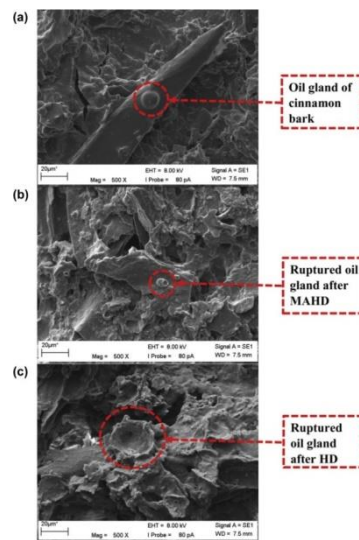
In MAHD, oil glands are slightly disrupted because of the thermal distribution. Effective and easy release happens due to the unnecessary rupturing of the oil glands compared to HD since HD causes aggressive and explosive rupturing of the oil glands.

The lower yield has resulted from cardamom since cardamom consists of a hard seed coat. According to the Enzyme assisted extraction results in rupturing of the cells and the destruction of cell walls. SEM images of the control sample show lower cell wall rupturing, and the outer coat looks like a fibrous and lumpy structure. Water-treated samples show a physical rupturing of cell walls, and greater damage has resulted from hot water than cold water. The enzyme-treated samples resulted in decomposition and openings of the cell walls. This may be because the enzyme hydrolyses the cell wall materials, resulting in an efficient and effective extraction process.

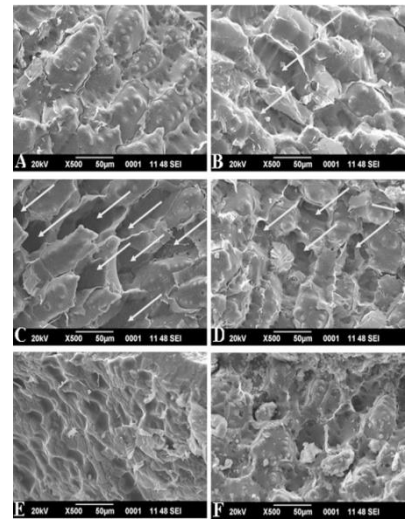


(a)- Microstructure of intact  
 (b)- Hydrodistilled  
 (c)- Ultrasound treated  
 (d)- Ohmic treated

(Tavakolpour et al., 2016)



(Jeyaratnam et al., 2016)



(A) -Control without any treatment  
 (B) -Pretreated with pectinex;  
 (C) -Pretreated with Viscozyme  
 (D) -Pretreated with celluclast  
 (E) -Soaked in cold water  
 (F) -Soaked in hot water.

(Baby & Ranganathan, 2016a)

Soxhlet Extraction is a solvent extraction method where powdered plant materials are mixed with the solvent and heated inside the apparatus for a precise time. Soxhlet extraction apparatus consists of a distillation flask, sample holder (thimble), siphon, and condenser. The drawbacks of solvent extraction include thermal

decomposition of the components, higher time and energy consumption, and the remaining residual solvents in the final extracts. These limitations and the strict regulations for solvent usage lead to experiments on the modern routes for extracting plant materials (Dutta & Bhattacharjee, 2015).

### 3.2.2. Ultrasound assisted extraction

Ultrasound assisted extraction (UAE) method is a method that has a higher yield in the extraction in lower time duration at a lower temperature. Because it facilitates the higher mass transfer efficiency and lower usage of solvents. This method is highly effective in the extraction of heat-sensitive constituents as this method involves low extraction temperatures (Ananingsih et al., 2020).

Extreme pressures, temperature gradients, and shear forces generated by high shock waves cause chemical compounds to dissolve in the solvent, exclusive of high temperature and mass transfer. The high-speed solvent jets (generated due to ultrasonic waves) facilitate the enlargement of pores in cell walls, accelerating the penetration of the solvents and increasing the mass transfer. Furthermore, the cavitation process causes to rupture of cell walls and membranes. Therefore, the cellular mass causes the flow to the solvent, and selective compounds are dissolved according to the solvent medium (Tavakolpour et al., 2016). For UAE, the ultrasonic bath is used at the laboratory level and ultrasonic heads at the pilot level. There is a remarkable difference between the composition of the extracts extracted by UAE and conventional methods due to the release of the compounds from secretary cells and changes in structures of unstable chemical compounds. In oleoresins, the residual solvent is critical, and the study done by (Nurhadi et al., 2020) revealed that there is lesser residual solvent in ORs, which were extracted using UAE.

### 3.2.3. Microwave assisted extraction (MAE)

Microwave-assisted extraction (MAE) uses microwave radiation for microwave heating. Mostly, it is combined with other conventional extraction techniques for its significant benefits such as high extraction efficiency with lesser time, lower solvent consumption, eco-friendliness, energy conservation, efficient selective and heating, decreased recovery time, a quick thermal transfer (Desai & Parikh, 2015; Mejri et al., 2018). If it is combined with solvent extraction, it decreases solvent usage and time. The principle of MAE is transferring the energy by electrical fields to take place dipole rotation by interacting the dipoles with polar compounds and causing the dipoles to realign with the applied field. The heat is generated due to the movements of molecules. In addition, when the electromagnetic field is applied, the movements of the charged ions inside the solvents occur, and heat is generated due to the friction within the solution. The microwave heat evaporates the moisture inside the cell wall, and extreme pressure affects the rupturing of the cell wall and the release of the active compounds inside the cells (Ekezie et al., 2017). The extraction depends on the polarity of the solvents, temperature, extraction time duration, microwave power, and solid-liquid ratio.

Recently, diverse extractants have been used, such as two-phase solutions, and hydrotropic liquids in MAE, although organic solvents or water are used in traditional extraction. The principle of microwave-assisted extraction instruments is based on two techniques; a commonly used technique is done under controlled pressure and temperature in a closed vessel. The second technique is based on extraction in an open vessel under atmospheric pressure, and the solvent boiling point was set as the maximum temperature of it. A modified microwave oven is used at the laboratory level for MAE. The extraction efficiency is higher in

MAE due to the higher thermal energy through frictional resistance to the ion flow and continuous dielectric heating by the electromagnetic radiation (Belwal et al., 2018).

### 3.2.4. Super critical fluid extraction

Super critical fluid extraction is a technique of separating the extract from the plant matrix using supercritical fluids as the extraction solvent. Super critical fluid (SCF) technology was reported in the 1970s, and super critical fluid extraction (SCFE) emerged to minimize the drawbacks of other extraction processes. Furthermore, the SCFE requires lesser operational cost and enhances the yield, flavour, aroma and storage abilities (Chen & Huang, 2016). The other key benefit of supercritical fluid is environmental friendliness. In addition, the extraction efficiency is higher in SCFE since the supercritical fluids have higher diffusivity, higher density, and lower viscosity which facilitate the speedy and selective extraction, solute fractionation, and convenient concentration of extracts by depressurizing without further separation of the solvents. The compounds have lower thermal degradation as it operates at a lower temperature.

The supercritical fluids get the temperature and pressure above their critical values in the supercritical state. Hence, supercritical fluids exhibit gas-like characteristics by diffusing through solids like a gas and liquid-like characteristics by dissolving compounds like a liquid. In addition, the liquid-like densities direct the high loading of solutes and enhance solvating capacities. The gas-like low viscosities, high molecular diffusivities, and low surface tensions lead to better penetration to plant matrices, convenient and effective mass transfer, and faster and selective extraction of target compounds (Dutta & Bhattacharjee, 2015).

The density of the SCF can be changed by altering pressure, temperature, or both. Changing density influences the solvating power of the extracts since solvation depends on the intermolecular forces due to packing the solvent molecules around the solute molecules. Therefore, the solubility can be changed by modifying the supercritical solvent, and the selectivity can be increased by changing the density. In addition, the fractionation of extracts is possible by gradually decreasing density.

The SCFE is greatly influenced by temperature and pressure. Solvating power can be increased by enhancing the fluid density due to increasing pressure. Temperature directly affects the extraction yield because it depends on the fluid density and solute vapour pressure. Higher temperatures reduce the density of the SCF and influence the dissolving of the compounds. Although high temperatures increase the solubility, it affects the volatility of the compounds.

The SCFE includes offline and online techniques (Yousefi et al., 2019), and this can be done as both continuous or batch processes using high-pressure equipment. The supercritical fluid is sent to the vessel where the raw materials are placed and are contacted with the materials. After the supercritical fluid gets saturated from the extract, it is subjected to reduced pressure (atmospheric conditions) in the collection vessel compared to the extractor. The solubility of the extract, which depends on the density, is decreased due to the reduced pressure. The target extract is separated, and the recovered supercritical fluid is sent for recycling. When the supercritical solvent is passed into a vessel at a lower pressure than the extraction vessel, supercritical fluids' density and dissolving power vary sharply with pressure. As the lower density CO<sub>2</sub> solubility is much lower, the material is precipitated for collection. Therefore, the general SCFE system

contains a pump (for pumping supercritical fluids), pressure vessel with heating abilities (for getting critical conditions to SCF), pressure maintenance (for maintaining the pressure in the system), collection vessels (for separation of the extracts), cooling (for maintaining liquid conditions) and heating after pressurization.

The most common supercritical fluids are carbon dioxide, and other solvents include water, ethanol, methanol, hexane, propane, and butane. The modifiers and co-solvents facilitate the extraction processes.

Supercritical carbon dioxide includes the below characteristics,

- Eco-friendly;
- Inert, Non-toxic, non-inflammable, non-explosive, non-corrosive;
- Readily available and widely used;
- Odourless, tasteless, colourless;
- Generally recognized as safe (GRAS) by the Food and Drug Administration (FDA);
- Non-reactive with extraction materials and equipment;
- Carbon dioxide is produced as a by-product of other industries (Environmentally friendly), and it is less expensive than other common solvents;
- In a supercritical state, the critical temperature is 31.1°C and 73.8 bar;
- The critical conditions are easy to reach, and high diffusivity;
- Convenience in gasifying and separation from solute due to low vaporization enthalpy;
- Volatile and heat liable matters can be conveniently separated;
- Appropriate for extraction of lipophilic compounds as the polarity of carbon dioxide is the same as liquid pentane at a supercritical state

(Dassoff & Li, 2019; Dutta & Bhattacharjee, 2015).

Although the water (H<sub>2</sub>O) is used in SCFE, it has limitations such as the requirement of high temperature (more than 374°C) and pressure (more than 221 bar) and corrosiveness under these conditions.

Recent developments of SCFE involve polar modifications with co-solvents, ultrasonic assistance, and some pre-treatment methods, such as enzymatic pre-treatment. Another remarkable merit of this method is that SCFE can also do fractionation by either way of stepwise extractions, such as the first extraction happening at a lower density than the second extraction or one stage extraction with many separation steps (Essien et al., 2020). This operation can be done using several pressure-reducing steps and a series of collection vessels. Solvent flow rate, extraction time, temperature, pressure, particle size, and pre-treatment of the plant materials are the main factors that affect the extraction process. It is revealed that the extracts which are taken from supercritical carbon dioxide extraction have more antioxidant, antibacterial, and anti-proliferative compared to the solvent extraction (Zizovic et al., 2018)

The limitations of supercritical carbon dioxide extraction are the risk of retention of carbon dioxide in the blood of operators and difficulties in high-pressure conditions at the industrial level (Essien et al., 2020).

### 3.2.5. Enzyme-assisted extraction method

Enzyme assisted extraction (EAE) has been developed to enhance the efficiency of extraction, increasing the yields in an eco-friendly way by using enzymes and reducing the use of solvents, extraction time, energy inputs, and environmental issues. In EAE, hydrolytic enzymes such as alpha-amylase, cellulase,

pectinase, and protease enzymes or blends hydrolyze the cell wall components such as cellulose, hemicelluloses, lignin, and pectin (Boulila et al., 2015). It will disrupt the structural complexity of the cell wall and easy release the target compounds by increasing the permeability of the cell wall or cell plasmatic membrane (Belwal et al., 2018). The EAE depends on the enzyme concentration, pH, temperature, particle size of the substrate, and extraction time (Puri, 2017).

The enzymes can be used as a pre-treatment of raw materials prior to the distillation of essential oils too. Research that used enzymes as a pre-treatment of raw materials resulted in a higher yield than the control (Baby & Ranganathan, 2016c, 2016a; Boulila et al., 2015).

### 3.2.6. Subcritical water extraction

Subcritical, superheated, or pressurized hot water is in its liquid state at a temperature between 100°C and its critical point of 374°C under adequate pressure. These specific conditions decrease the dielectric constant and act as a solvent for hydrophobic matters. The main advantages of this method are reducing the extraction time, eco-friendliness, selective extraction, and the use of nontoxic solvents (Essien et al., 2020). The main drawback of this method is the degradation of the thermolabile compounds due to the usage of higher temperatures.

### 3.3. Development of combined distillation and extraction methods

Recently, the extraction methods have shifted towards modifications based on combinations of different extraction routes for the advantage of different methods.

#### 3.3.1. Ultrasound-assisted ohmic extraction

In this method, ultrasound and ohmic techniques are coupled to increase extraction efficiency due to fast and homogenous heat transferring and increasing the surface area for extraction due to the sudden rupture of essential oil glands after the onset of high temperature. The plant materials are sonicated using an ultrasonic device in a brine (NaCl) solution. Then the whole content was extracted in a chamber of the ohmic extractor after onset boiling, followed by a constant voltage (Tavakolpour et al., 2016).

#### 3.3.2. Enzyme-assisted supercritical carbon dioxide extraction

Currently, supercritical extraction is done using enzymes. The lyophilized enzyme is mixed with raw material powders in optimum ratio and subjected to supercritical extraction, which can be done in both batch and continuous modes. The main disadvantage is the lower yield in continuous mode due to the short incubation period.

As spice coatings consist of a significant amount of carbohydrates, treating with enzymes will enhance the hydrolysis of starch to release the oleoresins, increasing the extraction yield. In this method, the raw materials are mixed with the lyophilized enzyme, and supercritical fluid extraction is done. A study done to extract black pepper oleoresin reveals that using alpha-amylase followed by supercritical extraction resulted in a significantly

enhanced yield and higher piperine content in black pepper oleoresin (Dutta & Bhattacharjee, 2015).

### 3.3.3. Ultrasound assisted super critical extraction methods

This method had been developed to increase yields, especially in high-valued but low-volume products (Dassoff & Li, 2019).

### 3.3.4. Enzyme-assisted ultrasound extraction

Enzyme-assisted extraction can be combined with ultrasound extraction at high frequency and low intensity to enhance the extraction efficiency, higher recovery and reduce solvent usage since the ultrasound technique improves the enzyme capability and even distribution of the enzyme (Cheng et al., 2015).

## 4. Comparative studies of modern extraction routes and conventional extraction routes

Many research studies have been done to compare the modern extraction routes and conventional extraction routes in terms of the extraction yields, the composition of the extract, the antioxidant activity and antimicrobial activity of the extract, the carbon dioxide emission to the environment, and energy consumption. Table 1 demonstrates the comparison of the modern routes and the conventional routes.

## 5. Plant matrices and oil glands in extractions

Fresh or dried roots, barks, leaves, flowers, and seeds are used as plant materials to extract essential oils and oleoresins. The extraction of these botanicals depends on the plant materials, such as moisture content, type of plant matrices, and cell structure. Some phytochemicals are met in specific cell structures found inside the plant tissues or on the plant's surface. The EOs biosynthesized in the special cell structures such as osmophores, glandular trichomes, conical-papillate cells, cavities, ducts and rarely non-specialized cells and released to the environment via two different secretion mechanisms of granuloocrine and eccrine. Mostly the glandular trichomes are present in the families such as *Solanaceae*, *Lamiaceae* and *Asteraceae*.

The key purpose of these compounds is to defend the plants' diverse organs. Thus, there are two types of glands; short term glands (capitate glands), in which fast secretion happens for defending the young organs and long-term glands (peltate hairs), in which gradual accumulation happens in subcuticular space and leads to defending mature organs (Hazzoumi et al., 2020). Some studies revealed that some nonvolatile constituents occur in foliar glands, and foliar glands are loaded with a wide range of flavonoids related bioactive compounds (Goodger et al., 2016). The recent research studies on the extraction methods are explained using Scanning Electron Microscopes (SEM) images of oil glands for better understanding (Table 2). According to the SEM images of Tavakolpour et al. (2016), the split oil glands for different directions and many holes can be seen throughout the wall in the OE method due to the higher evaporation rate and heat transfer rate.

In contrast, longer time heat exposure in HD resulted in shrivelled tubers. Rapid and efficient crumbling and rupturing were observed in MAHD, which causes slight damage and shrinkage to

the structure in the wall of the oil gland (Golmakani & Rezaei, 2008). The study by Desai and Parikh (2015) revealed that rupture of the cell wall could not be observed in hydrodistilled materials, and MAE's tremendous structural damage to the cell wall was observed. The reason for that is the absorption of microwave radiation by the polar compounds and sudden energy transfer.

## 6. Applications of extracts and future trends

The EOs, ORs, and bioactives are extracted using different extraction methods are used for food and beverages, cosmeceuticals, perfumery, pharmaceuticals, nutraceuticals, sanitary and agronomy (Swamy et al., 2016). Essential oils and oleoresins can be used to preserve foods due to microbial spoilage and autoxidation. For instance, EOs such as ginger oil can be used not only for flavouring but also to improve the shelf life of food products. Many studies have been done to determine the antimicrobial activity of secondary metabolites. The chemical compounds of aromatic plants can hinder the growth of a diverse range of pathogens, such as bacteria, fungi, and viruses. Essential oils are effective against many foodborne pathogens such as *Salmonella typhimurium*, *Escherichia coli*, *Staphylococcus aureus*, *Listeria monocytogenes*, *Campylobacter*, *C. Jejuni*, *L. Monocytogenes*, *Salmonella enteric*, *Pseudomonas aeruginosa*, *Proteus mirabilis*, *Laurus nobilis*, *Myrtus communis*, *Bacillus cereus*, *Bacillus subtilis*, *Enterococcus faecalis*, *Klebsiella pneumoniae*, *Acinetobacter*, *Pseudomonas aeruginosa* (Chouhan et al., 2017). Because of its antimicrobial properties, the EO can be applied in the cheese-making industry as a natural preservative (Khorshidian et al., 2018) and preserve meat products against spoilage and pathogenic microbes (Pateiro et al., 2021). There is a trend of application of EOs in the meat industry as a possible replacement for synthetic antioxidants (Pateiro et al., 2018). In addition, there is a potential for applying EO vapours in a system used to prevent microbial spoilage and preserve foods (Reyes-Jurado et al., 2020). The nutmeg oleoresin consists of antifungal properties. Therefore, EO formulations will be developed as a safe replacement for synthetic preservatives, which will add value because of green consumerism, sustainable food security, cost-effectiveness, renewability and biodegradability (Pandey et al., 2017).

There are many studies done on applying EOs for incorporation into active food packaging due to their antibacterial and antioxidant properties. Mostly the trend is using them for edible and biodegradable films for food packaging for different types of foods such as fruits, meat, fish, and cooked and raw food because of environmental concerns. In addition, the increasing drug resistance matter of currently used antimicrobials and developing antimicrobials using EOs are highly significant. There are many research studies and reviews done to find out the microbial activity of the EO against various human pathogenic bacteria such as *Staphylococcus aureus*, *Salmonella typhimurium*, *Clostridium perfringens*, *Staphylococcus epidermidis*, *Escherichia coli*, *Bacillus cereus*, *B. Subtilis*, *Clostridium botulinum*, *Streptococcus mutans*, *Enterobacteriaceae*, *Streptococcus pyogenes*, *Klebsiella pneumoniae*, against pathogenic fungi such as *Candida albicans*, *Aspergillus niger*, *Fusarium oxysporum*, and against pathogenic virus such as ORF virus (a parapox virus), Herpes simplex virus type 1 (HSV-1), Avian influenza A virus (H5N1), HSV-1, HSV-2, avian influenza virus (AIV) subtype H9N2, Influenza A (H2N2) virus, Japanese encephalitis virus (JEV) (Swamy et al., 2016). Because of their antimicrobial and antioxidant activity, essential

oils can be used for medicines. For example, Origanum can treat chronic diseases such as Alzheimer's and diabetes mellitus. Significantly, EOs can be used for new botanical larvicides. Especially mosquito larvae are sensitive to the sesquiterpenes of the EOs. There is a potential for using neuroprotective and anti-ageing properties. Therefore they can be used as multi-potent agents against neurological disorders such as depression, anxiety, cognitive hypofunction epilepsy, convulsions and neurodegenerative disorders such as dementia and Alzheimer's disease (Ayaz et al., 2017).

Many studies focusing on the novel extraction routes for extracting EOs and ORs are based on comparing the novel method with the traditional method in terms of extraction yield, extraction time, and the chemical composition of the EOs or ORs. The effect of the novel extraction on the plant matrices was mostly observed using SEM images and will lead researchers to visually observe the impact of the treatment on oil glands. In addition, many studies focused on evaluating the antioxidant and antimicrobial activity using *in-vitro* assays to determine the best extraction method.

Moreover, there is a remarkable increase in recent research which is based on the application of extracted the EOs, oleoresins and bio-actives with an advanced delivery system such as microencapsulation, nanoencapsulation for enhancing the intended functionalities and applications (Granata et al., 2022; Hemmatkhal et al., 2020; Fakhari et al., 2020; Burgos et al., 2017; Jayanudin et al., 2019). For instance, the study by Ansarifard and Moradinezhad (2022) revealed the application of thyme essential oil for strawberry preservation by incorporating it into electrospun zein fibre mats. Encapsulation facilitates the controlled and sustained release of the extractives, enhancing the bio-availability, efficacy of bio-actives and water solubility, preserving active ingredients by increasing the thermal and oxidative stability and lowering the degradation, higher efficacy for multi-resistant pathogens, decreasing the adverse effects of organoleptic properties, increasing antimicrobial efficacy of the food systems (Kapustová et al., 2021; Madhumitha et al., 2021; Rai et al., 2017; Saifullah et al., 2019). Along with this, a trend can be recognized in recent research which focuses more *in vitro* and *in vivo* assays in nano-based encapsulations of EOs and ORs (Amiri et al., 2021; de Alcantara Lemos et al., 2021; Kapustová et al., 2021).

## 7. Conclusion

The secondary metabolites of aromatic plants are extracted using different extraction methods. Essential oils and oleoresins are obtained using conventional routes such as maceration, cohobation, hydrodistillation, steam distillation, and solvent extraction. Novel approaches are developed to increase the extraction yield, efficiency, and quality of the extract by lowering thermal and hydrolytic effects, lowering the residues of the toxic solvents, reducing the extraction time, minimizing the loss of the volatiles and minimizing the energy consumption. Novel extractions include essential oil extraction methods such as ohmic assisted hydrodistillation (OAH), microwave assisted hydrodistillation (MAHD), solvent free microwave assisted extraction (SFMAE), microwave hydrodiffusion and gravity (MHG), microwave steam distillation (MSD), molecular distillation (MD), fractional distillation (FD) and oleoresin extraction methods such as ultrasound assisted extraction (UAE), microwave-assisted extraction (MAE), supercritical fluid extraction (SFE), enzyme assisted extraction (EAE).

The essential oils and oleoresins, extracted using different methods, are used in industries such as food and beverages, cosmeceuticals, perfumery, pharmaceuticals, nutraceuticals, sanitary and agronomy because of their bioactivity. The emerging encapsulation technologies enhance the applicability of the extracts.

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## Conflict of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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