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COMPREHENSIVE REVIEW OF DIFFERENT EXTRACTION TECHNIQUES INVOLVING GREEN SOLVENTS FOR OIL EXTRACTION

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ABSTRACT
Petroleum-based solvents are currently heavily regulated universally; there is a mounting need to use eco-friendly and reusable solvents for the extraction, refinement, and development of food and natural products. Non-Volatile Organic Compounds (NVOCs), which have an extremely high flash point and solubility as well as low toxicity and minimal environmental impact, are the appropriate alternative solvents. They must be affordable, renewable, and recyclable. Based on chemistry, green solvents are categorized into five types: super critical fluids, universal solvents, Ionic fluids, Fluorous biphasic solvents, and polymers. The employment of so-called green solvents like ionic liquids and supercritical fluids has been applied as extracting substances for purification, amelioration, or even effluent remediation based on the concepts of green chemistry & engineering. However, the majority of the solvent technology examples that promote sustainability involve the use of tried-and-true solvents. A synopsis of edible oils as solvents augmented with several bioactive substances sourced from natural resources is provided in this paper. It has been discussed how fatty acids and lipids function in vegetable oils.
Keywords: Ionic Liquids, Solvent Extraction, Green Solvents, Vegetable Oils, Nature-Based Education, Micro Extraction.
RASĀYAN *J. Chem.*, Vol. 18, No.4, 2025

INTRODUCTION

Prolonged exposure to solvents harms all living things and damages the majority of their organs. The everyday use of large amounts of dangerous and flammable solvents has raised concerns in the chemical processing industry. One, because of their unique properties, green solvents boost chemical reactions, diminish the requirement for solvents, and decrease the count of processing steps.[1] Ethyl lactate, for instance, is a green solvent produced during the processing of corn.[2]

Categories of Green Solvents

Super Critical Fluids

A material that is in a phase transition between a gas and a liquid at a temperature and pressure greater than their respective critical values is called a supercritical liquid. A material that can exist both above and below its critical temperature and pressure is known as a supercritical fluid, or SCF[3]. Supercritical liquids are the perfect organic solvent alternative for industrial and laboratory processes because of their exceptional solubility in a wide range of polymers. Supercritical pressure of CO_2. There are numerous uses for superheated water extraction, sometimes referred to as pressurized hot water extraction or Subcritical Hot Water Extraction (SHWE). Fig.-1 shows the benefits and drawbacks of super critical fluids.

Application of $ScCO_2$ as a Solvent

$ScCO_2$ is frequently employed during polymerization. Since they are less soluble in organic solvents, monomers containing silicon and fluorine are polymerized in $ScCO_2$.$ScCO_2$ is also used in the free radical polymerization of acrylate monomers with perfluoro-ponytails (with the help of a free radical initiator such as Azobisisobutyronitrile (AIBN)).[5]

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Vol. 18 | No. 4 |1948-1958| October-December | 2025

Universal Solvent

Water functions well as a solvent for most organic processes.[6] Water can undergo a variety of reactions and syntheses, such as multicomponent reactions, radical reactions, and pericyclic reactions. Furthermore, the fact that metal catalysts are insoluble in oxygen in water may work to their advantage.[7] Benefits and drawbacks of the organic process are depicted in Fig.-2.

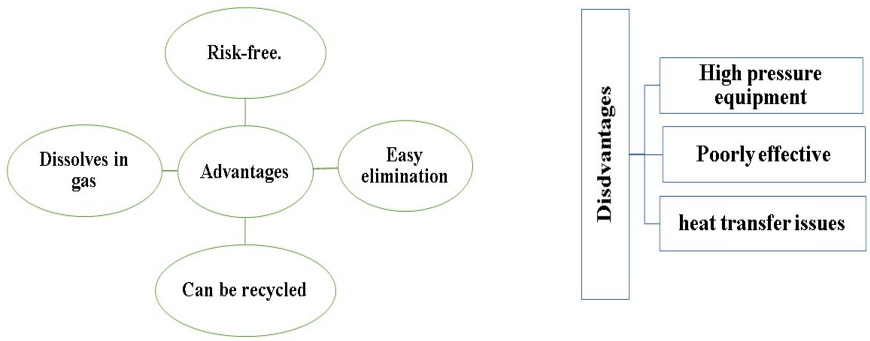

Fig.-1: Benefits and Drawbacks of Supercritical Fluids

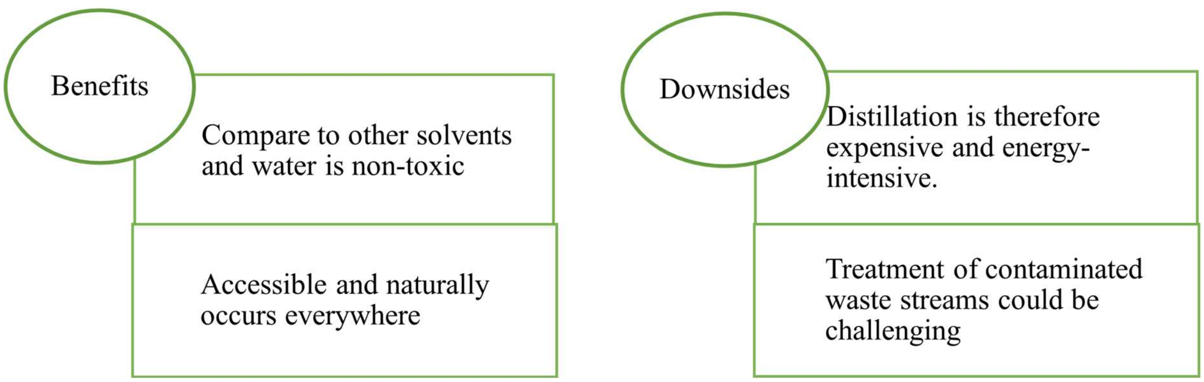

Fig.-2: Benefits and Downsides of the Organic Process

Ionic Fluids

Ionic liquids are defined as either a molten salt in a liquid state or salts with a melting point below a set temperature, like 100ºC. Because of their poor coordination, ions in an ionic liquid are liquid at or even below 100ºC. Ionic liquids do not mix evenly with many organic solvents; instead, they offer two-phase systems an anhydrous polar substitute.[8]

Ionic Liquid Characteristics

- Ionic liquids have a low vapour pressure, are non-ionizing.
- Conduct electricity moderately to poorly, and are highly viscous.
- Ionic liquids can be used as bio catalysis solvents.

Uses of Ionic Liquid

- Ionic liquids can be employed in liquid-liquid extraction operations since they are miscible with water or organic solvents.
- Ionic liquids offer the chance to conduct high-temperature reactions at low pressure since they are stable at temperatures exceeding 300ºC.[9]

Ionic Liquids as Extraction Solvents

The unique physicochemical properties of ionic liquids (ILs) have drawn increased attention in recent years. This may be demonstrated by the increasing use of ILs as laboratory solvents, particularly in numerous sample preparation procedures as a "green" substitute for volatile organic solvents. Ionic liquids have proven to be a successful substitute for traditional organic solvents in a number of analytical chemistry applications, such as mass spectrometry, capillary electrophoresis, chromatography, and sample preparation and extraction methods.[10] Depending on the specifics of individual extraction techniques, the use of ILs can be divided into solvent-based extraction & sorption-based extraction.[11] The basic solvent-based extraction technique is depicted in Fig.-3

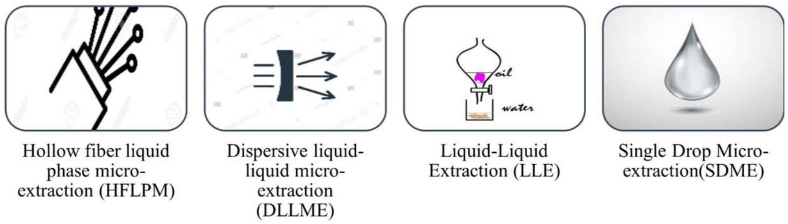

Fig.-3: Basic Methods of Solvent-Based Extraction

Ionic Liquid-Based Single Drop Micro-Extraction

The fundamental idea behind this technique is that the intended compounds are dispersed between the sample and a little drop of extractant that is suspended on the micro syringe's tip. The solvent drop is immersed in the sample solution when using the two-phase direct immersion mode. When utilizing the three-phase headspace SDME mode, the solvent drop that is suspended on the needle tip is submerged in the sample headspace. The droplet containing the sample is drawn into the micro syringe and is immediately available for analysis following the proper amount of equilibration time.[12]

Dispersive Liquid-Liquid Micro-Extraction

DLLME is another broadly used sample preparation micro-extraction technique that was initially described in 2006. To swiftly and affordably extract analytes from aqueous matrices, the DLLME technique employs an organic modifier (dispersive solvent), miscible with both water and the extractant, in addition to a water-immiscible solvent (extraction solvent).[13] The migration of the analytes to the extractant droplets is accelerated by rapid mass transfer brought on by the larger surface area between the extractant and the hydrophilic phase. After that, the extractant is separated, typically by centrifugation.[14]

Hollow Fibre Liquid-Phase Micro Extraction (HF-LPME)

Analytes are shifted from the hydrophilic sample (donor phase) to the organic solvent (acceptor phase), submerged within the sample and trapped inside the hollow fiber lumen, using this technique. Two-phase mode, in which the acceptor solvent used to saturate porous fibre walls fills the hollow fiber lumen. Three-phase mode, in which the acceptor phase and an extra solvent that is miscible with them are immobilised on the fibre walls.

Liquid Phase Micro-extraction (LPME)

Ionic liquids are successfully used in the sample preparation field when using sorbent-based extraction methods, especially solid-phase micro-extraction.[15] Analyte partitioning between the stationary phase and sample, followed by analyte desorption, forms the basis of Solid Phase Micro Extraction (SPME). Metallic or fused Silica Fiber coated with the proper stationary polymeric phase serves as the foundation for the majority of SPME variants. Ionic liquids were utilized to improve selectivity because the coating is one of the major SPME variables. Originally utilized as disposable Fiber coatings, ionic liquid Fibers can now be reused repeatedly thanks to technological advancements.[16]

Fluorous Biphasic Solvent

At room temperature, fully fluorinated hydrocarbon solvents that are immiscible with organic solvents are known as "fluorous biphasic." Reactants and catalysts should dissolve in the fluorous phase during a reaction, but the products should readily separate into a different phase at room temperature.[17] Because of the extremely strong C-F bond, fluorous bi-phase solvents have properties like low flammability, high levels of inertness, high thermal stability, hydrophobicity, and non-toxicity.

Polymers

The polymer, like polyethylene glycol (PEG), can also be degraded by soil and sewage bacteria. Following PEG, the next non-toxic liquid shares many of PEG's properties but is somewhat less

Vol. 18 | No. 4 |1948-1958| October-December | 2025

biodegradable. *Daphnia magna* is harmful to Poly Tetra Hydro Furan (PTHF) in the form of hydrous emulsion, and Poly Dimethyl Siloxane (PDMS) and di-alkyl ether-capped PEG were revealed to be much less biodegradable.[18]

Benefits of PEG as a Solvent

1. PEG is capable of conducting several metal-catalyzed carbon-carbon bond-forming processes.
2. Both thermal and microwave irradiations have great compatibility with reactions.

Green Extraction (Changed to heading font)

The goal of green extraction is to create extraction processes that consume a reduced amount of energy, allow for the practice of renewable natural resources and alternative solvents, and ensure the creation of high-quality, safe extracts and products. Three primary solutions have been established in order to develop and reveal green extraction on a lab and manufacturing scale, and to strive toward optimal solvent consumption, raw material, and energy consumption.

- Improving and streamlining existing procedures.
- Making use of non-specialized tools.
- Innovation in techniques and processes, along with the discovery of alternative solvents.[19]

Six Principles of Green Extraction

Green extraction briefly provides principles. Figure-4 illustrates the green extraction principle.

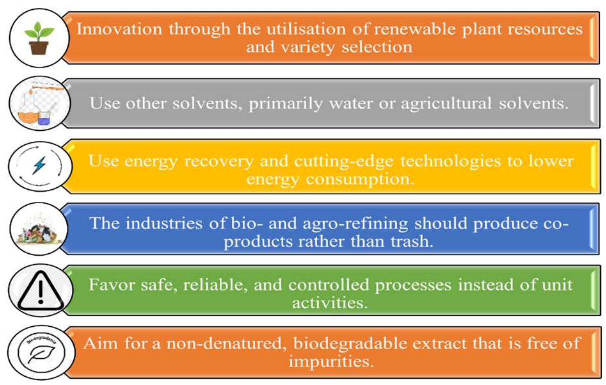

Fig.-4: Principle of Green Extraction

Utilisation of Renewable Plant Resources

Either extensive culture or in vitro plant cell or organism growth must be used to favor fully renewable resources. As a result, numerous research projects have been launched to identify alternatives to removing trees belonging to this endangered species. Therefore, if natural resources are harvested on a large scale without adequate control, there is a danger that the species will become scarce or possibly extinct.[20] The main natural source of α-bisabolol is the *Erymanthus erythropappus*, which is found in the Minas Gerais State, Brazilian rainforest, and is in a vulnerable ecological state. Recently, Symrise decided to cease extracting natural bisabolol from the candeia tree to incorporate conservation into its corporate social responsibility. One recent example is an anti-malarial substance-artemisinin extracted from *Artemisia annua*. A novel method known as "plant milking" has been established to generate and extract useful molecules without endangering the plant. Chemicals are secreted and exuded from the roots of plants growing in a liquid medium in a greenhouse when they are stimulated physically, chemically, or biologically.[21] Following established protocols, the compounds are then removed and purified. This approach focuses more on obtaining active ingredients from rare plants, whose chemical synthesis requires a significant investment of time and resources.[22]

Uses of Other Solvents

The bio or agro solvents among the green solvents are crucial in replacing petrochemical solvents. These bio solvents are non-toxic, biodegradable, and have a high solvent power. The most prevalent bio-solvent is ethanol, which is produced through the fermentation of foods high in sugar, such as sugar beets and grains.[23] Because of its high solvent power and low polarity, D-limonene can be utilized to extract fats and oils. Glycerol, a trans-esterification secondary product of vegetable oils, is widely utilized in the cosmetics industry as a macerating solvent for spices and herbs. Because of their excellent physicochemical stability, solvent power, and non-flammable and non-volatile organic compound (NVOC) qualities, ionic liquids are presently utilized in the environmentally friendly extraction of phyto-chemicals. For example, a simple precipitation process yields pure chemicals when artemisinin is extracted using ionic liquids.[24] Water's polarity makes it suitable as an extractant for both inorganic and naturally occurring water-soluble substances, including organic acids, proteins, and carbohydrates. Pressurized hot water-based extraction in hot (or) sub-critical form, particularly in a dynamic mode, is among the most promising contemporary green extraction methods and technologies.[25] A recent environmentally friendly development in aqueous extraction is the addition of cyclodextrins in solution because of their unique hydrophobic cavity and inclusion complexes forming capacity between bioactive compounds. Resveratrol and other polyphenols were effectively extracted by ultrasonography from the *Polygonum cuspidatum* ground roots in a cyclodextrin-based water solution. Microwave hydro-diffusion and gravity, which is another innovative microwave solvent-free extraction technique, has been used to extract colours, essential oils, and antioxidants. This technique likewise includes putting the crude drug in a microwave reactor in the absence of solvent or water, and it is based on a pretty straightforward premise. Thus, microwave heating releases the essential oil and water *in situ* that penetrate the crude drug, and causes them to fall out of the microwave reactor due to gravity.[26]

Usage of Energy Recovery

Environmental and economic issues that call for a drastic reduction in energy use and waste production have an especially negative impact on extraction. When moderate pressure is used, the distillation duration is reduced by a factor of two or three. As a result, less steam is used, which reduces energy usage. Certain fragrant plants, such as sandalwood, cloves, or the rhizomes of iris, ginger, and khus, need to be distilled in air for more than twenty-four hours, but they can be distilled in less than three hours using pressure steam distillation at 0.5 MPa.[27] It is possible to capture and use the heat generated during vapour condensation to lower energy usage. In the south of France, approximately 55% energy can be recovered using a prototype eco-evaporator for the steam distillation of lavender. Yuzu, a Japanese citrus fruit, has been used to extract its essential oil via ultrasound-assisted hydro distillation. This method produces 44% higher extraction yield than the conventional one. Examples include the use of microwaves, ultrasonic activation, and pulsed electric fields. By installing extraction processes in intricate recurring installations, the restrictions imposed by the process type can be overcome, and even new items could be introduced.[28]

Industrial Production

Waste is any material that needs to be disposed of by industry using an incinerator, landfill, or other type of waste disposal facility. It's erratic and unintentional. By-products such as potato fiber proteins, beet pulp, oil cakes, cereal grains, etc., have economic worth.

Co-product is a substance that is intentionally and ineluctably produced concurrently with the primary product within a single manufacturing process.[29] In contrast to the conventional approach, which is often intended to produce just one product from a given biomass, this innovative green method for transforming agro-products produces a range of target items. This idea employs an integrated approach to using the entire plant. Creating edible oil from oil seeds produces a significant amount of waste. Oil cakes made from pressing seeds and high in protein, are typically fed to livestock.[4,30]

Process Control

To stay competitive, industries that extract natural resources need to combine safer and cleaner extraction methods with process intensification.[31] The industrial growth of intensified processes faces a variety of obstacles, including the need for fewer unit operations and more compact production units, as well as the need to conserve energy and raw materials and control process safety and waste output.[32] The method for extracting β-carotene from carrots is depicted in Fig.-5.

It is possible to reduce a process's chain steps in order to save money and improve energy efficiency. The supercritical fluid extraction process has the merit of utilizing a fresh solvent and creating an extract with the fewest possible steps.[34] After extraction, all solvent residues are removed from the isolated extract, the pressure is released, and CO_2 is expelled and recycled.

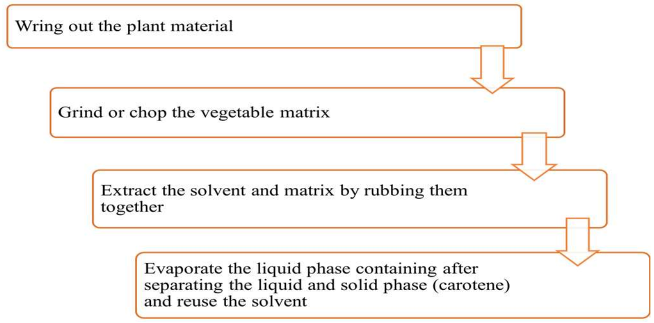

Fig.-5: Procedure of Extracting β-Carotene from Carrots[33]

It has always been detrimental to believe that supercritical fluid extraction is a costlier process than traditional ones.[35] But if we look at the full process flow instead of just the specific activity, we can see that this technique only requires a few separate steps. It also eliminates the need for hazardous solvents and lowers the energy needed to move the solvent from the extraction phase to distillation or recycling. An innovative development in the environmentally friendly extraction of crude products will be the extraction method. Usually, industries that modify raw materials are situated far from the locations where plant species are cultivated.[36] This raw material's transportation to the extraction site has a high carbon cost. The "green" distillation of lavender serves as a case of a "transferable" technique. Fresh lavender is distilled on-site, avoiding drying and shipment to the distillery. All stages of the "green" distillation are streamlined, reducing handling while preserving the essential characteristics of the plant.[37] When evaluating the possible environmental effects of an extraction operation, it is necessary to quantify the system emissions and resource consumption. Therefore, creating a movable, transportable "green extractor" is a means to significantly lessen the environmental impact of the extract or product.[38]

Uncontaminated Biodegradable Extraction

The extract's composition might therefore impact the quality and bioavailability of different phytochemicals due to the existence of natural transporters such as saponins, phospholipids, and others. Nonetheless, the challenges imposed by the law usually call for different preparation instructions.[39] Unlike the more stringent rules found in the food and pharmaceutical sectors, the legislation governing essential oils in cosmetics is rather simple. Using this assessment method across several ecological impact categories enables the measurement of any possible environmental impacts of a system that includes every step involved in producing a good or service, from the acquisition of raw materials to the last stages of processing, which include distribution and consumption. This approach entails keeping track of energy, natural resources, and environmental emissions consumption. Every stage of the life cycle has documentation of the flows of matter entering and leaving.[40] The number of hazardous substances released and resources collected during a life cycle is displayed in this inventory of base flows. The amount that each flow contributed to the different environmental issues under investigation is used to evaluate the environmental effects based on the data gathered.[41] It is preferable to examine the life cycle analysis of the complete supply chain, which includes the production of the plant and its cultivating, the recycling of the secondary products, and the biodegradability of the products obtained, rather than just the product or the isolation as a single unit operation, to gauge the impact of "green extract" on sustainability.[42]

Selection of an Ideal Solvent

Products can be implemented, applied, cleaned, or separated using solvents. Today, solvation is a necessary stage in every production process for fine chemicals, pharmaceuticals, food components, nutraceuticals, perfumes, and cosmetics. Examples of applications for solvents include diluents and additives for paints and inks, cleaning agents, and extraction and synthesising solvents. Macroscopically speaking, a solvent is a continuum with characteristics like density, viscosity, refractive index, surface tension, relative permittivity, boiling and melting points, vapour pressure, and thermal conductivity.[43] The term "green solvents" generally refers to seven types of solvents:

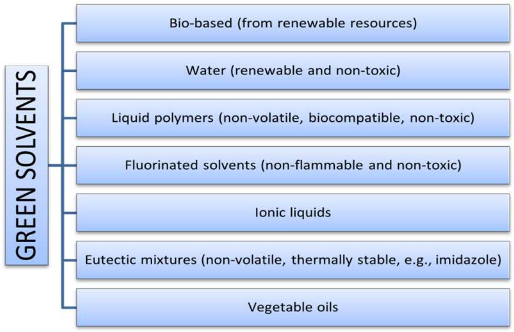

Fig.-6: Seven types of Green Solvents

Whether consumed either in refined, virgin form or through a range of food industrial products, vegetable oils constitute an essential part of contemporary diets. These greasy goods can also be utilized as components or ingredients in a variety of other sectors, such as paints, lubricants, biodiesel, cosmetics, and nutraceuticals.[44] Therefore, the re-examination of vegetable oils as possible 21st-century alternatives to petroleum-based solvents will encourage the development of efficient techniques for applying them in such exciting fields as refinement, formulation, and extraction.[45]

Non-Triglyceride Vegetable Oils

Primary Ingredients in Vegetable Oils

Pressing or solvent extraction from seeds and fruits (such as rapeseed, sunflower, or olive) produces vegetable oils. They are regarded as nonpolar, lipophilic systems with a highly varied and complicated composition depending on their source, calibre, and manufacturing processes. Based on the degree of saturation, fatty acids are classified as saturated, monounsaturated, or polyunsaturated.[46] These fatty acids can have diverse configurations, which can change their chemical and physical characteristics.

Minor Components in Vegetable Oils

Non-glycero lipids, including tocopherols/tocotrienols, free fatty acids, phenolic compounds, pigments, proteins, vitamins, sterols, water, and glycerolipids, including mono- and diglycerides and phospholipids, etc., can be separated into these two categories.[47] The plant sources of different phytoconstituents are displayed in Table-1.

Table-1: Botanical Sources of Various Phytoconstituents

Extracts	Oil types	Materials	Reference
Phenolic substances	Almond oil	Oregano and dried rosemary	48
	Dietary fats	Olive fruits, Fruit fragments, or fruit residue	49
	Sunflower, Olive, and palm oils	leaf of olive (*Olea europaea*)	50
	Filtered sunflower oil, Sunflower oil high in oleic acid	An olive cake, Cakes made of Frozen Olives	51
	Refined oils for food	Olive pits	52
	Canola oil for frying	Hazelnut-green leaves covered in Olive and Hazelnut leaves	53
	Virgin Sicilian Olive oil, Almond oil.	Rubus genus (*Rubus saxatilis*, ideally Rubus fructicosus Schott., from the group Rubusulmifolius)	54
Astaxanthin	Bean oil, palm oil	Creature waste	55
	Olive, Grape seed, Soy, Corn, and other oils	Colossal tiger shrimp (*Panaeusmondon*)	56
	Hempseed oil	Microalgae (*Haematococcus pluvialis)*	57

Antioxidants	Grain oil, Virgin Olive oil	Dry Thyme blooms (*Thymus capitatus*) and Rosemary	58
Other aromatic substances	Virgin olive oil	Aromatic Tunisian plants (Thyme, Rosemary, Lavender, Basil, Lemon zests, White Sage, Garlic, Menthe, and Geranium) as well as dried rosemary, oregano, and spicy pepper	59
Essential oil	Virgin and refined Olive oils combined	Mature Garlic bulbs, Rosemary leaves, and dried Oregano	60
	Filtered corn oil	Peel of a *Citrus aurantium*	61
Carotenoids	Dietary fats	Discarded shrimp (*Panaeusindicus*)	62
	Refined oils for food	Microalgae (*Haematococcus pluvialis)*	63
Oleuropein complex or secoiridoids, which account for 89.4% of the phenolic-rich extract; Hydroxytyrosol, phenyl alcohols (p-coumaric acid, Vanillic acid, and vanillin), and tyrosol, which account for 3.5% of the extract; and flavonoids, which account for 6.0%	Almond oil	Substances derived from the Olive cakes polyphenols	64
Reduced oxidation	Safflower oil	Powdered, air-dried Lamiaceae plants	65
Flavouring substances	Oil of olives	Olives	66

CONCLUSION

Green chemistry is basically made on the principle of an eco-friendly nature. Such an approach can proceed in the absence of solvents in order to prevent hidden toxicity. However, green chemistry does not eliminate the need for a solvent since it contributes to particle size uniformity and bonding strength. Hence, organizing bodies for chemistry suggest the use of green solvents owing to the non in non-flammability, eco-friendly nature. Qualities of various green solvents have been improved by the application of innovative techniques like ultrasonication and the microwave approach. While ionic fluids are widely used in electro synthesis of metal complexes containing zinc and ions, supercritical fluids are not only used for extraction and purification processes but also for dissolving gaseous substances if needed. Further research must be focused on the development of the efficiency of green solvents by improving the reaction rate of the process and the commerciality of the final products.

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CONFLICT OF INTERESTS

There is no conflict of interest, according to the authors.

AUTHOR CONTRIBUTIONS

This review is related to *SDG 12: Responsible Consumption and Production*. All the authors contributed significantly to this manuscript, participated in reviewing/editing and approved the final draft for publication. The research profile of the authors can be verified from their ORCID ids, given below:

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[RJC-9424/2025]