



Research Article

Green Solvents in Organic Synthesis: Sustainable Alternatives for Eco-Friendly Chemical Transformations

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Organic solvents are widely employed in organic synthesis but raise significant concerns due to their high flammability, volatility, toxicity, and hazardous nature. According to the principles of green chemistry, the use of solvents should be avoided or minimized whenever possible. However, in cases where their use is unavoidable, it is essential to prioritize greener, less toxic alternatives to reduce environmental and health impacts. These challenges make it difficult to identify a compound or class of compounds that can be truly recognized as a green solvent. This chapter focuses on unconventional solvents, such as ionic liquids, supercritical fluids like water and carbon dioxide, deep-eutectic solvent and water, biosolvent, which serve as alternative reaction media to overcome the challenges linked to traditional organic solvents.

Keywords: Green Solvent, Extraction, Flammable, Volatile, ionic liquids, deep-eutectic solvent, bio solvent

INTRODUCTION

In practically all chemistry, biology, chemical engineering, biotechnology, and other multidisciplinary scientific and technological processes, solvents are utilized to help transmit mass and heat as well as to make separations and purifications easier. In addition, solvents are extensively utilized for the purification of chemical compounds via extractions, separations, chromatography, or crystallizations. (Curzons et al., 1999; Henderson et al., 2011). Some of them have been used in organic synthesis for long time for instance benzene, petroleum ether (a low boiling mixture of aliphatic hydrocarbon), diethyl ether, dichloromethane, 1,2-dichloroethane, chloroform, n-hexane, diethyl ether, Di isopropyl ether, 1,2-dimethoxy ethane, 1,4 dioxane and chloroform are categorized as toxic harmful substances (Sheldon, 2019). To minimize the ecological impacts caused by solvents and to reduce the cost associated with waste disposal and environmental remediation, it is important to

mindfully select solvents that conform to the principles of green chemistry. (Slater & Savelski, 2007). Green chemistry is the use of a series of principles that will reduce the use or generation of hazardous substances during the design, production and application of chemical products. In 1998, Anastas and Warner explicitly explained the fifth principle of green chemistry- “safer solvents and auxiliaries”- “the use of auxiliary substance (e.g., solvents, separations, etc.) should be made unnecessary whenever possible and, innocuous when used. “The key aspects that specify green solvents are low toxicity, functionally safe to use (Incombustible, nonexplosive), naturally abundant and easily available, cost-efficient, and ready separation from the reaction mixture. In this chapter include Water, Supercritical fluids (supercritical carbon dioxide (CO₂) and supercritical water), Ionic liquids (ILs), Deep eutectic solvent (DES), Bio solvent and other organic green solvents are discussed. The fundamental properties of these solvents are given in Table 1

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TABLE: COMPARISON OF DIFFERENT GREEN SOLVENTS USED IN ORGANIC SYNTHESIS

Properties	Water	Deep Eutectic Solvents (DES)	Ionic Liquids (ILs)	Bio-solvents	Supercritical Fluids
Example	Water	Binary or ternary mixtures comprising at least one Hydrogen Bond Donor (HBD) and at least one Hydrogen Bond Acceptor (HBA). Example: Choline chloride and ethylene glycol and other combinations.	“Designer solvents”; combination of IL cations are organic, such as imidazolium, pyridinium, and both inorganic and organic anions.	D-limonene, pinene, p-cymene, glycerol, 2-methyltetrahydrofuran (MeTHF)	CO ₂ , Water
Toxicity	Low toxicity	Very low toxicity	Wide range of different toxicity levels depending on ionic structure	Low toxicity	Non-toxic
Flammability	Non-flammable; non-volatile	Non-flammable	Typically, non-flammable and non-volatile	Relatively low flammability	Low flammability
Polarity	Polar	Polar (e.g., glycerol and ethylene glycol-based DES)	Polar	Low polarity	Supercritical CO ₂ : non-polar; water: polar
Applications	Production of biofuels, bioproducts from algal biomass, oil extraction for biodiesel production, extraction of bioactive compounds such as phenolic compounds.	Dissolution and extraction of polysaccharides; used as solvents for biological macromolecules mainly phospholipids, proteins, nucleic acids, and polysaccharides.	Conversion and characterization of biomass; applications in catalysis, biocatalysis, analytical applications, and electrochemical applications.	Used in many industrial sectors including paints, coatings, pharmaceuticals, printing inks, cosmetics, and household care products.	Extraction of natural compounds from food products, extraction of essential oils, and drug extraction from natural resources.

Abbreviations:

- **DES** = Deep Eutectic Solvent
- **HBD** = Hydrogen Bond Donor
- **HBA** = Hydrogen Bond Acceptor
- **ILs** = Ionic Liquids
- **CO₂** = Carbon Dioxide

(HBD)-hydrogen-bond donor, (HBA)-hydrogen bond acceptor, Sc- Supercritical

Table -1. The fundamental properties of green solvents(Claux et al., 2021; Janicka et al., 2022; Zhou et al., 2019)

Water

Green chemistry faces the challenge of rendering chemical technology more environmentally sustainable. To achieve this, it takes into account various factors, including the quantity of waste generated, levels of toxicity, and the energy and electricity consumed. The use of water has minimal environmental repercussions, and it is inexpensive to produce, transport, and dispose of. Consequently, it is wise to explore the replacement of harmful organic solvents with water, which is regarded as one of the most environmentally friendly solvents, ensuring a reduction in environmental impact and health risks. Water has various distinctive physical characteristics, including a significant dielectric constant and substantial heat capacity. These chemical and physical attributes provide it with distinct benefits as a "green solvent" for organic reactions. Additionally, other key traits of water involve hydrophobic interactions and hydrogen bonding, which lead to the exclusion of nonpolar substances from the water layer. Because of its hydrophobic interactions, water can not only speed up the rate of organic transformations that have a negative activation energy change but also enhance both chemoselectivity and regioselectivity (Zhou et al., 2019). For example, Rideout and Breslow discovered in 1980 that using water as a solvent could accelerate Diels-Alder reactions (Zhou et al., 2019). They later demonstrated that the selectivity of certain Diels-Alder reactions improved when performed in aqueous solution and suspension (Breslow et al., 1983)

Subcritical water extraction (SWE)

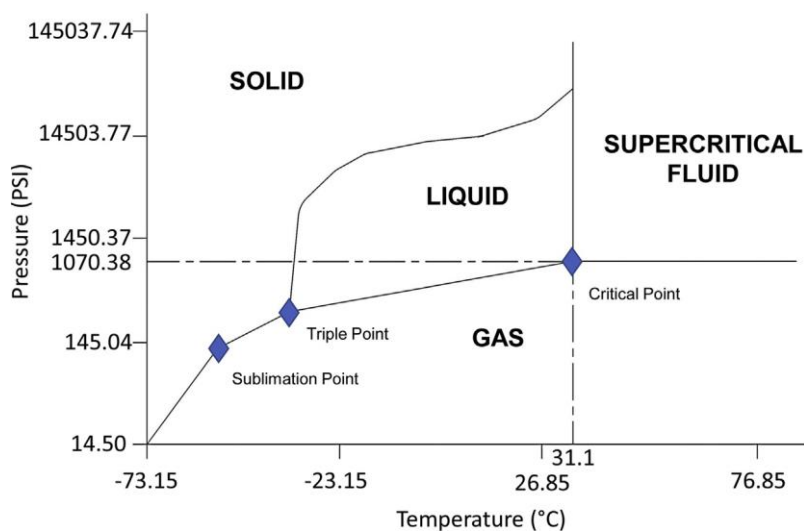
Water is solvent that can adjust its properties by simply changing the temperature. Subcritical water extraction (SWE), also known as pressurized hot water extraction (PHWE), is recognized as a "green" extraction method. To perform an extraction using SWE, it is essential to utilize high temperatures and pressure to maintain water in its liquid form. These conditions alter its physicochemical properties. In SWE there are various parameters like temperature, pressure, extraction time, and flow rate. Among these, temperature stands out as the critical factor impacting extraction efficiency and selectivity. As the temperature rises under subcritical water conditions, it produces notable effects on the extraction process.

The dielectric constant of water, (ϵ), decreases with changes in temperature, indicating that the polarizability of solvent water can be adjusted accordingly. Moreover, at higher temperatures, liquid water acts as a solvent with reduced polarizability and polarity. SWE boasts a significant benefit from the outset due to its sustainable development approach, as it is a process that does not require organic solvents. Moreover, phenolic compounds can be hydrolysed in SWE at extraction temperatures of 196 °C. In general, the extraction of terpenoids was carried out at high temperatures (180-200 °C) and long extraction times (27-120 min), whereas the extraction of polysaccharides was achieved using a broad range of temperatures (from 110 to 185 °C) and times (from 5 to 92 min). Moreover, SWE at temperatures over 200 °C may be used for the production of biochemical compounds used for biofuel production and the extraction of environmental samples (Castro-Puyana et al., 2017).

Supercritical Carbon dioxide (SCCO₂)

"Supercritical fluid" designates a type of substance that remains in a single-phase state and does not condense when maintained above its critical temperature (T_c) and critical pressure (P_c). These critical points represent the maximum temperature and pressure at which a supercritical fluid can coexist as both vapor and liquid. A supercritical fluid demonstrates characteristics of both gases and liquids at the same time. Supercritical carbon dioxide (SCCO₂) is a singular phase where the boundaries between liquid and gas phases are indistinct when it reaches or exceeds the critical temperature

(31.1°C) and critical pressure (72.8 atm) of carbon dioxide, which confers it with excellent diffusivity, viscosity, and solvation abilities(Razzak, 2021).



Phase Diagram of Carbon dioxide(Topuz et al., 2020)

Among the many supercritical fluids available, SCCO₂ is widely favoured because of its advantageous qualities, including being inert, non-combustible, non-harmful, affordable, and easily accessible in high purity (Topuz et al., 2020). These attributes allow scCO₂ to serve as a substitute for organic solvents in numerous applications, promoting safer, more energy-efficient, and environmentally sustainable industrial methods(Sajal & Sutar, 2025) . Its capability to dissolve various substances, which can be altered by changing pressure and temperature, enhances its effectiveness in extracting non-polar and moderately polar bioactive compounds from intricate mixtures(Manna & Banchero, 2018). This versatility is beneficial across several industries, especially within food and nutraceutical sectors, where scCO₂ is utilized for processes like decaffeinating coffee beans and extracting essential oils, flavours, pigments, and fatty acids(Kayathi et al., 2020). Its capacity to operate under moderate temperatures allows thermolabile substances to maintain their biological activity and structural stability(Lyu et al., 2021) . The endorsement from regulatory bodies has enhanced its market potential; for example, scCO₂-derived ingredients have received approval from the FDA and EFSA for safe consumption, highlighting its safety (Yu et al., 2015). The distinctive physical

and chemical characteristics of scCO₂—especially its adjustable density, minimal surface tension, and elevated diffusivity—allow it to act as both a solvent and a co-reactant in various catalytic processes. These attributes promote better interactions among reagents and catalysts, which speeds up reaction times and enhances selectivity. This trait has been utilized in many industrial applications, particularly in drug manufacturing, polymer alteration, and the production(Sajal & Sutar, 2025). Different uses of supercritical CO₂ have been documented, including altering polymers, creating polymer mixtures, blending polymers, producing microcellular foams, generating particles, and facilitating polymerization, where CO₂ functions as a plasticizer, solvent, or antisolvent(Nalawade et al., 2006).

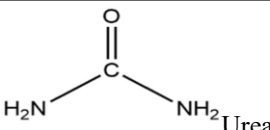
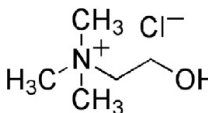
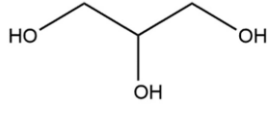
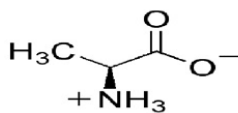
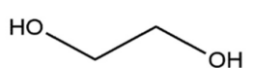
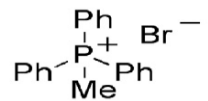
Deep Eutectic Solvent(DES)

Deep Eutectic Solvent (DES) was first introduced by Abbot in 2003 to refer to a new kind of sustainable solvent that, when mixed in the proper molar proportions, had a melting point significantly lower than that of its individual components(Abbott et al., 2003). Deep Eutectic Solvent can be defined as mixtures of two or three safe components that are able of associating with each other through hydrogen bond interactions (Zhang et al., 2012).. Most DESs are binary or ternary mixtures comprising at least

one hydrogen-bond donor (HBD) and at least one hydrogen bond acceptor (HBA) (such as a quaternary ammonium salt), which are strongly

associated with each other via hydrogen bond interactions.

Table -2 Structure of Hydrogen bond donor and Hydrogen bond acceptor for DES preparation

	Hydrogen bond donar	Hydrogen bond acceptor
1	 Urea	 Choline Chloride
2	 Glycerol	 L-Alanine
3	 Ethylene Glycol	 Methyltriphenylphosphonium bromide

This contributes to decrease the lattice energy of the system and thus the melting point. First introduced by Abbott and co-workers, choline chloride and urea are the most studied DES constituents (Abbott et al., 2003). Moreover, the usual components of DES are derived from renewable materials (such as choline chloride, urea, glycerol, lactic acid, carbohydrates, polyalcohols, amino acids, and vitamins). As a result, they possess an exceptional ability for biodegradation, and their toxicity is either absent or extremely low. Considering their minimal environmental impact, affordability of their ingredients, adjustable physical and chemical properties, and simple creation process. DESs are increasingly and effectively substituting the often harmful and volatile organic compounds (VOCs) across various scientific disciplines (Paiva et al., 2014). DES can isolate enzymes (e.g. lipases, dehydrogenases, laccases, peroxidases and ~lyases) and whole-cell microorganisms are active in DESs. Thus, DESs can act as substitute reaction media for water in biochemical activities that replicate cellular metabolites and lipids, while simultaneously maintaining the integrity of protein structures. (El Achkar et al., 2019). DES is also use in polar organometallic chemistry and metal

catalysed reaction and the catalytic system could be efficiently recycled.

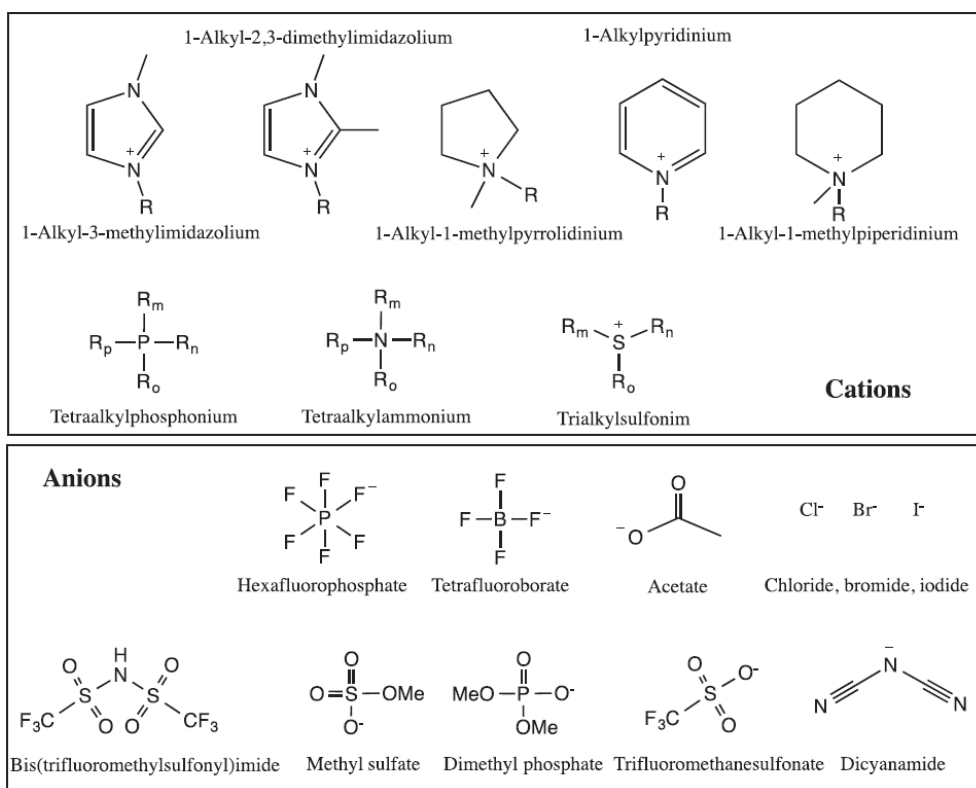
Ionic liquids (ILs)

Ionic liquids (ILs) have been explored as a practical alternative to traditional volatile organic solvents in recent decades. An Ionic liquid (ILs) is characterized as a liquid made up solely of ions that remain liquid at relatively low temperatures, ranging from room temperature to 100 °C (Zhao et al., 2005). The intense electrostatic interactions among ions in ionic liquids (ILs) contribute to their low tendency to volatile or flammable, as well as their adjustable miscibility, polarity, strong chemical and electrochemical stability. ILs are regarded as "green solvents" due to their minimal vapor pressure. Furthermore, ILs possess unique attributes as "designer solvents," enabling modification of their qualities through specific combinations of cations and anions. Factors like melting point, viscosity, density, and hydrophobicity nature can be altered by changing the ionic structure. For instance, the melting point temperature of 1-alkyl-3-methylimidazolium tetrafluoroborates and hexafluorophosphates depend on the length of the 1-alkyl chain, leading to the formation of liquid crystalline phases when the alkyl chains exceed 12

carbon atoms₂(Earle & Seddon, 2000). While the cations are usually organic, such as imidazolium, pyridinium, aliphatic ammonium, and both alkylated phosphonium and sulfonium ions, IL anions can be derived from both inorganic and organic sources. Ionic liquids are formed from salts that include one or both ions of considerable size, with the cation exhibiting low degree of symmetry. These characteristics typically lead to a decrease in the lattice energy of the solid state of the salt, resulting in a reduced melting point. Ionic liquids can be classified into two primary types: first is simple salts, which consist of a single anion and cation, and second is binary ionic liquids, which involve multiple ionic species in a state of equilibrium. For instance, $[\text{EtNH}_3][\text{NO}_3]$ represents a simple salt, while combinations of aluminium (III) chloride and 1,3-dialkylimidazolium chlorides form a binary ionic liquid system, featuring various ionic components, and their melting point and properties are influenced by the relative amounts of aluminium

(III) chloride and 1,3-dialkylimidazolium chloride(Earle & Seddon, 2000).

ILs find use in diverse fields, including catalysis, biocatalysis, various analytical processes, electrochemical applications, inorganic synthesis, polymerization, metal extraction, and other uses. With the increasing focus on lignocellulosic biomass, the role of ILs has been broadened to include the conversion and analysis of this biomass. Using ILs Lignocellulosic biomass can be characterised and recognized as a valuable resource due to its abundance and sustainability. Its structure is complex and rigid, primarily comprising cellulose, hemicellulose, and lignin. These structural properties contribute to the resistance of biomass to breakdown; hence, effective methods are essential(Yoo et al., 2017). Given figure illustrates several common cations and anions found in ILs.

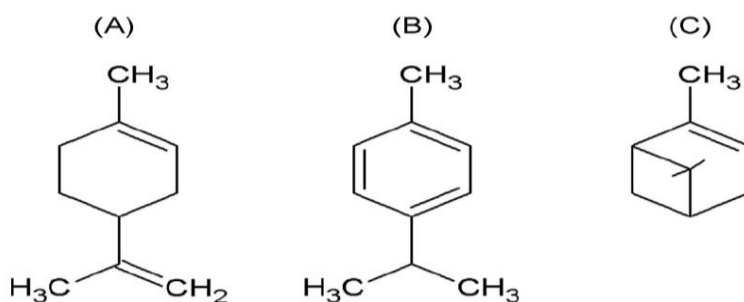


Common anions and cations used in Ionic liquids (Yoo et al., 2017)

Bio Based Solvent

Bio-based solvents refer to solvents made from renewable biomass materials like energy crops (like corn), timber products (such as wood), aquatic sources (like microalgae), and waste substances (for instance, urban waste). These solvents are generated in a biorefinery, which integrates processes and machinery to convert biomass into fuels, energy, and various chemicals (Li et al., 2016). These methods yield a variety of substances, such as alcohols, esters, glycerol, terpenes, furfurals, and furans. Many of these have the ability to be utilized in eco-friendly extraction methods because they have low solubility in water, higher boiling points, better stability compared to other solvents, along with being non-toxic, biodegradable under typical environmental conditions, and capable of being recycled. (Janicka et al., 2022). These bio-based solvents could serve as alternatives to the fossil fuel-derived solvents that are presently utilized across various industries, including paint and coating, pharmaceuticals, adhesives, printing inks, cosmetics, and personal care products (Clark et al., 2015). Ethyl lactate as bio-based and benign solvent was applied for the determination of total petroleum hydrocarbons in soil samples. (Tenne et al., 2013)

Terpenes including d-limonene, p-cymene, and α -pinene, contain isoprene structural units and occur in all organisms and can be extracted from a variety of plants including conifers. D-limonene is a solvent obtained from citrus byproducts, regarded as one of several precious resources, receiving considerable interest in the field of analytical sciences. This compound is commonly utilized in the food and cosmetic sectors, serving as a degreaser for both industrial and household purposes, as well as a solvent for paints. Research has demonstrated that d-limonene can substitute not only n-hexane but also toluene in the Dean-Stark method and replace chlorinated organic solvents in the extraction of carotenoids (Vovers et al., 2017). p-Cymene is used primarily for odour alteration, but also as a solvent for dyes and varnishes, also used in oil extraction from rapeseeds. p-Cymene had a higher yield and improved selectivity over n-hexane, alcohols and other terpenes (Vovers et al., 2017). Improved yield was also demonstrated using p-cymene as a replacement for ethanol/acetic acid in the HCl-catalysed Biginelli reaction (Clark et al., 2013).



Chemical Structure of terpene-(A) d-limonene, (B) p-cymene, (C) α -pinene (Vovers et al., 2017)

2-Methyltetrahydrofuran (2-MeTHF) is derived from biomass feedstock, such as levulinic acid or hydrogenation of furfural (Clark et al., 2015). 2-MeTHF has been reported as a bio-based solvent replacement for tetrahydrofuran (THF). 2-MeTHF has also been identified as a promising replacement for hexane (Sicaire et al., 2015). bio-based solvents applied for purification of membrane proteins in place of chloroform-methanol mixture are 2-methyltetrahydrofuran and cyclopentylmethyl

ether (Tenne et al., 2013). In a study including nine bio-based solvents, 2-MeTHF was identified as an economically viable alternative to n-hexane in aroma compound extraction, with performance similar to the volatile organic compound solvent. The compound 2-MeTHF was found to be a suitable alternative for hazardous solvents such as tetrahydrofuran, toluene, dichloromethane, hexane, diethyl ether, and ethyl acetate

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