IJNRD.ORG ISSN : 2456-4184
INTERNATIONAL JOURNAL OF NOVEL RESEARCH AND DEVELOPMENT (IJNRD) | IJNRD.ORG
An International Open Access, Peer-reviewed, Refereed Journal

Green Solvents and Ionic Liquids-their Applications

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Abstract

Practically all Industrial manufacturers largely rely on solvents for a multitude of tasks such as separations, facilitating chemical reactions leading to desired products via catalytic, non-catalytic and bio-catalysis routes. Conventional solvents, mostly carbon based, are toxic and dangerous to environment. Careful selection of solvents based on health and environmental safety considerations therefore plays a vital role in the entire chemical processes.

Green chemistry and its concepts have been responsible for improvements in the capabilities of conventional solvents, with a new class of so-called master solvents, 'green' or 'designer' solvents, with little or milder ecological impact and other benefits like economic and productivity.

Solvents use consistently accounts for between 80 and 90% of mass utilization in a typical chemical operation. Solvent selection guides have become a vital component in the effort to enhance the greenness of the fine chemical industries. Accordingly water, Ionic liquids, supercritical fluids, Deep Eutectic solvents, bio-based, some petrochemical and non-toxic liquid polymers like PEG are part of the class of green solvents to date. 'Green' solvents are predominantly oxo-hydrocarbons (cyclic and acyclic alcohols, esters, carbonates and ethers) with some hydrocarbons and those containing other hetero atoms, with acceptable range of physical and chemical properties. Their applications include: reaction synthesis to oil extraction, sensors and biosensors, CO_2 capture, and bio-based chemicals. The pharmaceutical industry accounts for mass consumption of solvents.

Green technologies are moving from an option to a must in modern industrial applications. Ionic liquids, supercritical fluids and deep eutectic solvents are being most actively investigated as potential green solvents, due to their unique properties and possibility of tuning their cation and anion moieties to make them task-specific. In the chemical industry, they are well established for innumerable processes and applications, such as reaction media for organic transformations, in separations and extractions, as electrolytes for electrochemistry, in biotechnology, absorption of gases (CO_2) and as catalysts in organic synthesis. This article aims to provide a perspective on the green solvents, potential and successful commercialization of IL-based processes, to date.

Key words: designer solvents, ionic liquids; electrochemistry; supercapacitor, rechargeable battery, bio materials, catalysis and recycling.

1. Introduction

Solvents are ubiquitous and have a wide range of industrial and domestic applications. Conventional solvents, mostly organic or carbon base in nature, are general solvents that are routinely used for laboratory and industrial purposes because of their volatility, melting point, better dissolving power and yield. Apart from their direct exposure to laboratory and industrial workers, when released in environment and left untreated, these can cause significant potential hazards to environment and to human, living plants, and animals destroying most organs.

Industrial and manufacturing firms commonly rely on solvents for a multitude of tasks such separations, making products, cleaning and degreasing machinery and surfaces, working with materials such as coatings and paints, and facilitating chemical reactions.

Since 1990 European Solvent Industry Group [1] is advising the EU countries to follow guidelines to improve air quality by contributing to the reduction of total volatile organic compound (VOC) and emissions. The Solvents Industry Association [2] also offers advice and guidance to producers, distributors, and consumers of solvents to help to minimize the potential environmental impact.

Most of the organic solvents are toxic, dangerous and damaging to the environment. Therefore, selecting a suitable solvent is a great challenge to the research community as its impact on the environment is not forecast able. Such solvents are continued to be still used in large amount. Dwivedi s,et.al; lists different conventional solvents, their sources, and related risks to human health and environment.[3]

One of the important factor for choosing an improved solvents is the solvent properties rated by their health characteristics and environmental safety. In this context, Welton [4] summarizes few ways to make the chemical process more sustainable with the solvents having following characteristics: i) it must have dual role, ii) the product quality should be higher, iii) number of intermediate steps are reduced, iv) less amount of by-products is formed, v) and improve products. Hence, the careful selection of solvents plays a vital role in the entire industrial chemical processes. Neil Winterton [5] published, a pictorial representation of solvents used in a variety of industrial applications in Europe in 2017. About 5 million tons are used by the European solvents industry alone.

Byrne et al; [6] have reviewed several general purpose published green solvent selection guides with the aim to reduce use of the most hazardous solvents. This article serves the purpose of explaining the role of these guides, and how these can be used most effectively to enhance the greenness of chemical processes, particularly in laboratory organic synthesis and the pharmaceutical industry.

Solvents can fall into either category. The four key sectors of the chemicals industry-oil refining, bulk or commodity chemicals, fine or effect chemicals and pharmaceuticals-differ in the proportion of by-product and waste formed in product manufacture [7, 8] .This is highest for pharmaceuticals much of whose production uses multi-step liquid-phase batch operations. A multiplicity of solvents is used in the preparation, isolation and purification of intermediates at various stages of the production of a single active pharmaceutical ingredient.

2. Green Solvents

Solvents define a major part of the environmental performance of processes in chemical industry and also impact on cost, safety and health issues. The idea of “green” solvents expresses the goal to minimize the environmental impact resulting from the use of solvents in chemical production. This concept and application of green chemistry over the past two decades has led to the development of so-called neoteric or modern solvents.[5] This approach has been responsible for improvements in the capabilities of conventional solvents, with a new class of so-called master solvents, also termed ‘green’ or ‘designer’ solvents [9]. By definition an ideal fully sustainable green solvent would not have an ecological impact at any stage and would ease process conditions, making them milder and more sustainable. Such solvents are also likely to provide productivity and economic and environmental benefits [10].

The general concept of creating rankings of solvent greenness within the chemical industries has been viewed in different ways. The pharmaceutical sector in particular has been keen to establish their own institutional hierarchies of solvent greenness since the realization that the solvent is the major component of a typical reaction in the manufacture of an active pharmaceutical ingredient [11]. As a consequence process solvents are responsible for the majority of energy use, waste, and greenhouse gas emissions, which makes the minimization of solvent use and greener substitutions a priority. The solvent can have a profound influence on reaction rates and product selectivity [12] but the more general benefits of solvent use in reactions should not be overlooked either. Solvents act as a heat sink and a temperature regulator, lower mixture viscosity and improve mass transfer, and make selective extractions and separations possible [13].

Fischer and co-workers in their article [14] asked; “what is a green solvent”? The answer required two tiered assessment of environmental, health and safety (EHS) and energy demand. The system requires the user to perform calculations and provide a numerical score to compare ranking of solvents while making selection. The concept proved to be popular and subsequently repeated by other institutions.

Solvents use consistently account for, between 80 and 90% of mass utilization, in typical pharmaceutical and fine chemicals (non-polymer) batch chemical operations. Moreover, within these operations, solvents play a dominant role in the overall toxicity profile of any given process; i.e. on a mass basis, solvents account for the largest proportion of chemicals of concern used in the process. Papadakis e, et al; have developed a method for the selection of appropriate solvents for the solvent swap task in pharmaceutical processes taking into account process considerations such as batch distillation and crystallization. It is based on one of earlier reported solvent selection methods plus additional criteria such as boiling point difference, volatility difference, VLE phase diagram analysis, and azeotropic information that are particularly important for the solvent swap task[15].

Alternative solvents with low toxicity, minimal safety concerns and little impact on the environment can be selected from solvent selection guides designed for the pharmaceutical industry [6].

2.1 Solvent Selection Guides / Criteria

Three prominent solvent selection guides developed for medicinal chemistry are: Glaxo Smith Kline (GSK) [16,17], Pfizer,[18] and Sanofi [19]. The guides represent progressive improvements in solvent selection with time. The simple three-tiered and colour code approach to categorizing solvents for medicinal chemistry purposed has the advantage of easy interpretation, but at the expense of limiting the depth of information provided. When designing larger scale reactions, more information is needed about each solvent as the process is geared towards commercial scale manufacturing, where any concerns over EHS issues are magnified.

Solvent selection guides have become a vital component in the effort to enhance the greenness of the fine chemical industries, These user friendly tools communicate the issue clearly to users, creating awareness of greener alternatives and discourage the use of certain solvents in favour of others.

Pfizer were the first company to publish their colour coded, hierarchical solvent selection guide for medicinal chemists [18] The tool is a simple document listing solvents as ‘preferred’, ‘usable’, or ‘undesirable’ as presented in Table 1 below. Pfizer have prioritized user friendliness in making this solvent selection guide, if only to encourage chemists to use it. The solvents subjected to the red category are due to having these potencies: toxic, carcinogenic, mutagenic, low flash point, environmental risk of ozone layer depletion etc. [20, 21].

Table 1: PFIZER Solvent Selection Guide [18]

Preferred	Usable	Undesirable
Water	Cyclohexane	Pentane
Acetone	Toluene	Hexane(s)
Ethanol	Methylcyclohexane	Di-isopropyl ether
2-Propanol	TBME	Diethyl ether
1-Propanol	Isooctane	Dichloromethane
Heptane	Acetonitrile	Dichloroethane
Ethyl Acetate	2-MeTHF	Chloroform
Isopropyl Acetate	THF	NMP
Methanol	Xylenes	DMF
MEK	DMSO	Pyridine
1-Butanolt	Acetic Acid	DMAc
t-Butanol	Ethylene Glycol	Dioxane
		Dimethoxyethane

Equally useful as the solvent selection guides, is the criteria for solvent replacement proposed by Gu and Jerome [22] Table 2.This criteria suggests that, in practice, selecting an acceptable substitute solvent is a more complex subject. This observation is further supported by the quantitative green chemistry metrics, particularly those developed by John Andraos

[23] which highlight the difficulty of fully reconciling technical effectiveness, occupational safety and environmental impact and the impossibility of doing so perfectly. Judgments about whether one solvent or another is associated with more, or less, waste production, therefore, frequently rely on factors other than the chemical characteristics of the solvent itself.

Table 2: Criteria used to judge solvent's acceptability [22]

S. No.	Solvent Acceptability Criteria
•	Available on the required scale with a secure long-term source of supply
•	Technical performance (including solvency) no worse than the equivalent conventional solvent.
•	Stable during use and storage
•	Low or non-flammable
•	Competitively priced
•	Able to be recycled
•	Purity appropriate to use
•	Resource and energy efficient production (preferably life cycle assessed)
•	Sources from renewable intermediates and feedstocks
•	Established acceptable toxicity and ecotoxicity profiles sufficient for regulatory purposes.
•	Fully biodegradable to innocuous products.
•	Meets standards and regulations for transportation

2.2 Broad Classification and Sources

In 2015, the UN defined a new sustainability focused development plan based on seventeen sustainable development goals. This development led to the recognition of "green chemistry" and "green solvent" for a more sustainable chemistry in the future. It further motivated, more and more companies to go green, to ensure that their activities and products are part of a sustainable development process. It is in this context that so-called green more environmentally friendly solvents, or bio-solvents, and sustainable solvents have emerged as an alternative to petrochemical solvents [24].

Regular or conventional solvents can be classified in two categories namely: polar and non-polar. However it is much harder to do so in the case of for green solvents as their chemical structure and source can vary significantly. Binita et al;[25] have stated that the most important intention of green chemistry is to decrease the use of solvents or substituting them with less toxic ones. They are classified by convenient accessibility, low toxicity and option of reuse. Accordingly Ionic liquids, water, supercritical fluids, non-toxic liquid polymers like Poly ethylene Glycol (PEG) are part of the class of green solvents. Wikipedia (26) on the other hand states, green solvents are established from trials and errors in search for substitutes of existing hazardous solvents. Hence given below is the non-ending list of substances that research has found acceptable in favor of being qualified as green solvents, based on their source from which they are produced and production method.

- **Water**

Water is considered to be the nature's 'green solvent' for its bio-catalytic processes It is the cheapest and most abundant solvent for a large range of reactions and processes in industrial chemistry. Huge interest in using water as a solvent may be attributed to its easy accessibility and low cost, along with its green properties: non-toxicity, renewability, safety and ease of handling, ease of treatment and degradation, etc. As per Simon and Li. [27] water exemplifies well the physicochemical basis for the explanation of why a solvent that appears so 'obviously' green still struggles to achieve more widespread application as a process solvent There are, at least two key problems with water. First, it has a very high heat of vaporisation combined with a very low molecular mass -18 Daltons. Second, to meet regulations concerning the discharge of waste water into rivers and other natural waters, reduction in the concentration of potentially polluting organic solutes often requires very extensive treatment prior to discharge, making the use of water as a reaction medium much less attractive [28].

- **Ionic liquids-ILs**

ILs comprise a large category of organic salts that, due to differences in their cation and anion sizes, are normally liquids (generally fluid at room temperature) at a temperature less than 100^0C. Indeed, they are named green solvents, as their low volatility allows them to limit VOC emissions compared to conventional solvents. The most popular variously substituted cations include imidazolium, pyridinium, ammonium and phosphonium. Anions include halides, tetra fluoroborate, hexafluorophosphate, and nitrate. Bubalo et al; [29] state that ionic liquids are non-flammable, chemically, electrochemically and thermally stable, with negligible volatility. Optimistically, ionic liquids from renewable and biodegradable materials have recently emerged. But their Eco toxicity and poor degradability had been recognized in the past because the resources typically used for their production are petroleum based, as is the case for imidazole and halogenated alkanes. Plus, their availability is up for debate because their production cost is high [5].

- **Super Critical Fluids**

A supercritical fluid is the phase of a material at critical temperature and critical pressure that combine useful properties of gas and liquid phases. These fluids are classified under green technology and are hence environmentally benign [30].

A supercritical fluid provides a gas-like characteristic where the motion of the molecules are quite similar to gas molecules. On the other hand, a supercritical fluid behaves like a liquid because its density property is near liquid and, thus, it shows a similarity to a liquid in terms of dissolving effect. Other important characteristics are diffusivity, viscosity and recyclability. Most commonly used supercritical fluid is carbon dioxide (CO_2) which is popularly used in Decaffeination. Supercritical water (SCW) behaves as a dense gas with a dissolving power equivalent to that of organic solvents of low polarity. supercritical water boiler is expected to be used in India only after 2030 [31].

These unique materials have lead applications in field of extraction and analytical in chromatography. Prominent applications are in the pharmaceutical industry for micro ionisation and in dry cleaning, drying, impregnation transestrification and as refrigerant. Their main limitation is, they requires a lot of energy in reaching its supercritical state and then maintaining it for the duration of operation.

- **Deep Eutectic solvents-DESs**

Deep Eutectic Solvents (DESs) have been slowly emerging, as a green alternative to ILs, since 2004 [32] and can be prepared by mixing solid compounds which form a eutectic mixture with a melting point lower than either of the individual components melting points. [33]. DESs formation mainly occurs due to the generation of intermolecular hydrogen bonds between hydrogen bond acceptor (HBA) and hydrogen bond donor (HBD). DESs share many physicochemical properties with ILs (high viscosity, low volatility, non-inflammability, chemical and thermal stability). Ease of DESs storage and synthesis as well as the low cost of their starting materials are the other major benefits over ILs.

Natural sources based, a new class namely ‘Natural Deep Eutectic Solvents’ (NADESs) are mixtures of compounds prepared via HBA & HBD have a much lower melting point than that of any of their individual components . Besides all the advantages of DESs, NADESs are considered as environmentally friendly and ‘readily biodegradable’ due to the natural origin of their components. The compounds found to form this liquid phase are primary metabolites like organic acids (lactic, malic, citric acids, etc.), sugars (glucose, fructose, sucrose, etc.); amino acids, choline chloride, etc. [34, 35]. These compounds differ from ILs in terms of bonding, Ionic vs hydrogen bond and are non-toxic. A new family of deep eutectic designer solvents were synthesized and formed by crown ether (CE) complexes as HBA and PEG as HBD. The designer solvents unlock the potential for numerous applications in chemistry and material science especially the ultra-deep extraction of non-basic *N*-compounds from fuel oils. [36].

- **Switchable Solvents**

Switchable solvents are solvents that reversibly change physical properties to a great extent. This unique property is a consequence of a reversible reaction in response to an external stimulus such as change in temperature, pressure, pH and/or the addition or removal of a gas. As a consequence of the switching ability of the chemical reaction, the solvent can be effortlessly changed back to its actual original form. Reversible ionic liquids (RILs) are a type of switchable solvent which can change from a liquid to a solid or a gas, depending on the applied stimulus. This reversible phase transition is due to the formation or disruption of ionic interactions between the solvent molecules. Switchable polarity solvents (SPSs) a type of ionic liquids , can switch between polar and non-polar states in response to a stimulus, such as a change in pH or temperature. RILs have been used in applications like the separation of chemicals, catalysis, and energy storage.[37,38].

This switch in polarity can be used to selectively extract or dissolve different types of molecules, making them useful in a range of industrial processes, including extraction, separation, and purification [39]. "Switching" the physical properties and/or polarity of solvents opens the pathways for subsequent basic research and future chemical process development.

- **Petrochem solvents with green features**

Denis Prat, and his team reviewed published solvent selection guides and brought these together into a single guide with a six point scale namely: recommended, recommended or problematic, problematic, problematic or hazardous, hazardous and highly hazardous solvents. Out of the 51 solvents considered, beside water the following eight solvents were found to be in the recommended /acceptance category; ethanol, isopropyl alcohol, n butanol, ethyl acetate iso propyl acetate, n-butylacetate, Anisole and Sulpholane. All these solvents are basically produced in bulk from petroleum feed stocks, although some of them may be obtained from the renewable resources as well [40].

Other solvents that make into petroleum solvents include low molecular weight polymeric solvents such as Poly(ethylene glycol) PEGs with molecular weights below 600 Dalton which are viscous liquids at room temperature. It is a reusable solvent medium for organic synthesis [41]. Poly(propylene)glycol (PPG) has been used as medium in Indium metal mediated synthesis of homoallylic amines [42].N. Leininger and team reports use of aqueous solutions of low molecular weight polypropylene and polyethylene glycol as novel, safe, environmentally friendly solvents. Many organic compounds have been found to be soluble in these polyglycol solutions; thus, they could be used as replacement solvents in various chemical processes and as the medium for conducting chemical reactions.. In particular, three classes of organic reactions, SN1, SN2, and Diels-Alder, have been conducted in the polyglycol solutions [43].

Potential use of volatile methylsiloxanes (VMSs) as solvents for chemicals synthesis has been explored. Environmental impact studies of these solvents have shown these to be significantly lower than those of the non-polar organic solvents that they have the potential to replace. Methylsiloxanes as solvents for synthetic chemistry applications, both in organic and inorganic transformations has been described by Mohd Azri Ab Rani et.ai [45].

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Propylene Carbonate (PC) , Molecular formula $C_4H_6O_3$) is a polar aprotic solvent used as a "green" sustainable alternative solvent for chemical transformations. It is also compatible with other solvents providing an efficient ingredient in co-solvent formulations and is an effective substitute for more hazardous solvents such a MEK, methylene chloride, NMP, and perchloroethylene etc, due to its low vapor pressure and negligible photochemical reactivity. Its major applications are in the manufacture of paints, adhesives, coatings, surface cleaners, degreasers, strippers, and inks formulations as well as in lithium-ion batteries, as electrolytic solvent, and in the removal of CO_2 and H_2S from natural gas [46].

- **Bio-based Solvents**

Making use of renewable resources in producing solvents is a promising and important strategy to move towards sustainable chemical processing and to replace organic solvents derived from fossil raw materials. To this end, bio-based feed stocks such as carbohydrates, carbohydrate polymers, proteins, alkaloids, plant oils and animal fats have been used to produce bio-based solvents [36, 46].

The main processing methods include biochemical and thermochemical conversion [47]. Using one or a combination of these processing techniques, several classes of bio-based solvents (including alcohols, esters ethers (alkanes aromatics and neoterics) can be manufactured. A novel array of ionic liquids and deep eutectic solvents based on inexpensive bio-derived components has been developed, and because these solvents have low volatility, it presents a solution to the volatile organic solvent concern. Additionally, they are usually recyclable after use, with the benefit of tune ability to bring about smart solvents for tailored applications.

- **Solvents from waste materials**

Three types of waste materials namely; substances such as starch and vegetable oils and wastes derived from lignocellulosic biomass, used cooking oils from food industry and fusel oil –a byproduct of ethanol manufacturing process from sugars, are the key sources for the recovery of various solvents and other valuable products. In addition they do support environmental conservation as well.

Integrated bio refineries combine many processes in sequence to valorize lignocellulosic biomass into value-added products. Integrating appropriate processes enhances the yield, reduces the reaction time, cost-effective, and sustainable. This approach also supports new products and paves its path towards sustainability in the framework of the circular economy.[48] These refineries exploit food base substances to make ethanol, lignocellulosic waste into solvents like 2-methyletetrahydrofuran and esters of levulinic acids. These solvents have the potential to replace solvents like tetrahydrofuran, toluene, dichloromethane and diethyl ether in many applications.

Fusel oil (a non-ideal mixture) is used to isolate C6 alcohols and other fragrances and flavor ingredients. For obtaining isoamyl alcohol from fusel oil, use of dividing wall column indicate significant energy and economic savings compared to conventional configuration[49]. Other green solvents such as isoamyl acetate or isoamyl carbonate could also be obtained.

Mazubert A et al; have reviewed the intensification of fatty acid methyl esters (FAME) production from waste cooking oil (WCO) using innovative process equipment. In particular, it addresses the intensification of WCO feedstock transformation by transesterification, esterification and hydrolysis reactions along with it catalyst choice and product separation. The results indicates that continuous flow equipment that integrate both reaction and separation steps appear to be the best means for intensifying FAME production [50]. Byproduct of the synthesis Glycerol, can in turn be used to produce various solvents such as 2,2-dimethyl-1,3-dioxolane-4-methanol, usable as a solvent in the formulation of inks and cleaners

The solvents which have been claimed to be ‘green’ are predominantly oxo-hydrocarbons (cyclic and acyclic alcohols, esters, carbonates and ethers) with some hydrocarbons and those containing hetero atoms. Class wise solvents and their representatives along with their source are shown in Table 3.

Table 3: Class wise representative solvents

Class	Representatives Solvents	References
Alcohols	ethanol; butanol; tert-amyl alcohol (or 2-methylbutan 2-ol); glycerol;	51 (Banker et al; 2013)
	blends of acetone, butanol and ethanol (ABE)	
Esters and carbonates	methyl acetate, ethyl acetate,dimethylglutarate	52.(Mouret et al; 2014);
	glycerol triacetate; ethyl lactate; γ-valerolactone	
	5-(dimethylamino- 2-methyl-5-oxopentanoate, *Dimethyl carbonate*;	53 (Lebarbe et al; 2014)
	and glycerol carbonate	54 (Christy et al;2018).
Ethers	3-dioxolane; cyclopentyl methyl ether; isosorbide dimethyl ether;	
	2,5-dimethylfuran;2-methyltetrahydrofuran; ethyleneglycol monomethyl ether;	
	ethylene glycol dimethyl ether; triethyleneglycolmonoethyl ether [2-(2-ethoxyethoxy)ethanol]	55 (Shakeel et al;2014)
	1,2,3-trimethoxypropane; and dihydrolevoglucosenone	
Others	limonene; farnesane, benzotrif luoride;1,1,1,3,3-pentafluorobutane;	
	piperylene sulfone;	
	dimethylsulfoxide;hexamethyldisiloxane;	
	N-(2-methoxy-2-ethoxyethyl) dibutylamine	56 (Samori et al;2014).

For viewing the chemical structures of all the above solvent reader may refer to publication by Niel Winterton listed at reference number[5].

2.3 Physical properties

The selection of a solvent for a given reaction conditions is based on its important physical properties. For example their dissolution characteristics make it possible to assess the use of a particular solvent for a chemical reaction, such as an extraction and properties such as evaporation as it reveals the potential emission of volatile organic compounds. Wikipedia [26] lists some physical properties of green solvents falling in the categories like water, solvents derived from carbohydrates, waste materials, by extraction and that of propylene carbonate- a petrochemical solvents.

For the remaining categories, some specific publications have described their properties in detail. For example, Nowosielski et al. [57] carried out fundamental research by investigating important physical properties (density, speed of sound, refractive index, and viscosity) of both pure and aqueous solutions of a number of deep eutectic solvents. Benworth b et al; [58];examined Complex hydrogen bonding behavior of DESs, which is postulated as the root cause of their melting point depressions and physicochemical properties; to understand these hydrogen bonded networks.

The rational selection of the appropriate ionic liquid solvent for a particular reaction requires general understanding of the properties of ionic liquids, and the details of some properties of the specific ionic liquid solvents being considered. John S. [59] looked into solvent properties of ionic liquids that are relevant to catalysis. A roadmap for the literature values of density, viscosity, melting and glass transition temperatures, thermal stability, empirical solvent parameters, absorption, toxicity, surface tension, heat capacity, and thermal conductivity is provided.

Fatty Acid Methyl Esters (FAME). Their physical characteristics are closer to those of fossil diesel fuels than pure vegetable oils, but properties depend on the type of vegetable oil. A mixture of different fatty acid methyl esters is commonly referred to as biodiesel, which is a renewable alternative fuel' ETIP-bio energy factsheet [60]report comparison of fuel properties. FAME as compared to diesel is higher in density, lower in heating values, higher in viscosity, comparable in cetane number but lower in fuel equivalence'.It is also non toxic and biodegradable.

Some of the key properties of switchable solvents: are: High selectivity that helps in purification processes, low volatility, high stability and low reactivity, makes it easy to handle & storage, variable polarity allows for fine tuning of solvent properties, renewable attractive for green chemistry applications, non-toxic safer to use and handle and finally high solubility, useful for dissolving and extracting complex mixtures [61].

In terms of quantitative values, the switchable properties are caused by two parameters namely: strength of their conjugate acid's -pKa and octanol-water partition coefficient ratio $K_{ow.}$ They must have a pKa above 9.5 to be protonated by carbonated water and also a log (K_{ow}) between 1.2 and 2.5 to be switchable because they will not be too hydrophilic or hydrophobic. It also depends on the volumetric ratios which influence their switchable properties. For example, N,N,N′-Tributylpentanamidine is switchable, and for a volumetric ratio of compound to water of 2:1, it has a log(K_{ow})= 5.99, which is higher than 2.5. It can be explained by the fact that the volume of the compound is twice higher than the water volume [62,63].

In recent years much attention has been paid to physicochemical properties of green solvents and their applications in green chemistry, which as per Buxing [64] include:

- phase behavior, intermolecular molecular interaction, and the microstructures in complex SCFs, ILs, supercritical (SC) CO_2 /IL systems;
- effects of phase behavior and intermolecular interactions on the properties of chemical reactions in SC CO_2 , ILs, SC CO_2 /water, water/ ILs, and SC CO_2 /ILs;
- colloid and interface science of green solvent systems, including chemical thermodynamics, microstructures, and functions;
- synthesis of highly efficient catalytic materials using green solvents;
- development of greener routes for the transformation of CO_2 , biomass and aromatics into value-added chemicals in green solvents, and optimization of the reaction processes using the designable and tunable features of green solvents.

Based on above applications two recent discoveries include:

- Carbon fibers from plant-derived material where biochar based carbon fibers prove themselves as potential fillers in composite.- Politecnico di Torino, Italy
- DNA-based solar cells: Towards environmentally responsible approaches for solar light harvesting.- University of Connecticut USA

2.4 Applications

Industrial applications of green solvents include: wide range from reaction synthesis to oil extraction, sensors and biosensors, CO_2 capture, lignocellulosic biomass utilization, and bio-based chemicals [65]. Pharmaceutical industry accounts for mass consumption solvents in the range of 80–90%. Bio-solvents such as butanol, polyethylene glycol, bio-renewable solvents, and supercritical solvents are widely used in the extraction and solubility of drugs [66]. In the past two decades, supercritical fluids, ILs, and deep eutectic solvents have been the most actively investigated as possible green solvents, especially in the domains of food, flavor, and fragrance, as well as medicinal plant processing [67]. Organic carbonates are a family of organic molecules and chemical intermediates that are known for their high biodegradability, low toxicity, and flexibility which lead to their wide application in bio-catalysis [68]. Another interesting area is rare earth elements recovery and ionic solvents, such as ILs and deep-eutectic solvents, have gotten a lot of interest since they offer an alternative to traditional metal recovery methods [69].

Table 4 below, compiled from various references depicts different solvents types, their sources, industrial, and general uses [3].

Table 4: Green solvents: Sources, industrial and general uses [3]

	Solvent	Source	Industrial Uses	General Uses
1.	Water	Naturally occurring	Petroleum and food industry	(Universal solvent) Extraction of solvents, industrial purposes, etc.
2.	Glycerol	Animal fat	Food and cosmetic industry	Isolation of reaction products, personal care products, mouthwashes, etc.
3.	Ethyl lactate	Processing of corn	Food and beverage industry	Paint industry, removing greases,adhesives, magnetic tape coatings, etc.
4.	Supercritical carbondioxide	Liquid state of CO_2	Pharmaceutical industry	Polymer-related applications, Extraction of essential oils
5.	Ionic liquids	Organic salts	Electronic industry	Biocatalysis
6.	n-butane	Natural gases	Refinery and petrochemical industry	Aerosol propellant, fuel additives, etc.
7.	Bioethanol	Fermentation of sugar and vegetable residues	Power and chemical industry	Motor fuel, additive in gasoline, etc.
8.	Cyclopentyl methyl ether	Addition of methanol to thecyclopentene	Bio refineries	Biotransformations, bioextractions,chromatography, solid-phase peptidesynthesis.
9.	Dimethyl carbonate	Reaction of methanol withcarbon monoxide and oxygen	Paint and cleaning industry	Raw material for organic synthesis
10.	Ammonium	Volatilization of urea,manures, slurries	Fertilizer and pesticideindustry	Textiles, dyes, etc.

3. Supercritical Fluids, Ionic liquids and Deep Eutectic Solvents

3.1 Basic concepts

Green technologies are moving from an option to a must in modern industrial applications. As stated earlier, solvents are the core of the pharmaceutical, chemicals including petroleum, food, cosmetic and biotechnological process technologies. In the past two decades, supercritical fluids, ionic liquids, and deep eutectic solvents became the most actively investigated as potential green solvents. The discussion below assesses recent information about these novel green solvents[70].

Supercritical fluids, ionic liquids, and deep eutectic solvents, each has its own specific physicochemical properties (Table 5) which consequently confine their applications. The first generation of green solvents were supercritical fluids. Their properties are somewhere in between gas and liquid. Since the end of 1970s, supercritical fluids are widely used for the extraction of chemicals from various sources including, for example, the decaffeination of coffee using SC CO_2, and essential oils from spices [71,72] Table 5**:** Compares density and viscosity of supercritical CO_2, ionic liquids, and deep eutectic solvents with organic solvents.

Table 5: Comparison of properties of $ScCO_2$, Ils and DES [72].

Gases		Density (g/cm^3) (0.1–) $x10^{-3}$	Viscosity (g/cm s) (1–3) x 10^{-44}
Organic Solvents		0.6-1.6	(0.2–3) x10^{-2}
Supercritical CO_2	Tc, Pc	0.47	3 x 10^{-4}
	Tc, 6Pc	1.0	1 x 10^{-3}
Ionic liquids	C4mimBF4	1.14 0	0.115
	C4mim(CF3CO_2)2N	1.43	6.9 x10^{-2}
Deep eutectic solvents	ChCl-ethylene glycol (1:2)	1.12	0.36
	ChCl-urea (1:2)	1.24	6.32

Tc: critical temperature. Pc: critical pressure. C4mim: butylmethylimidazolium

Supercritical carbon dioxide ($ScCO_2$) is a good example of supercritical liquids. It is known as a green solvent because it acts as a good solvent for many non-polar, few polar and low molecular weight compounds. These liquids are the perfect replacement for organic solvents for industrial and lab processes due to their great solubility in many polymers. Science Doze lists as many as ten advantages of $ScCO_2$, as solvent, compared to traditional organic solvents with five negative effects [73]. An important application include, Polymerization of fluorine and silicon-containing monomers performed in $ScCO_2$ as they are less soluble in organic solvents. For example synthesis of Fluorinated poly acrylate is achieved through this approach.

3.2 Synthetic ionic liquids

Synthetic ionic liquids are made up of ions that are liquid at 100°C, in contrast to high-temperature molten salts [74]. It is rather a new class of sustainable solvents. Applications and potential of ionic liquids are comprehensively reviewed by Plechkova and Seddon [75]. Ionic liquids are liquid salt mixtures, in which individual ionic components bind with each other through ionic bonds [76]. They have characteristic physicochemical properties, which distinguish them from conventional organic solvents and high conductivity, with a wide range of electrochemical, and polarity properties. Because of these features they replaced conventional organic solvents in many chemical processes.

The molecular structure of ionic liquids is made up of different cations and anions. The cation is usually played by a large to bulky and asymmetrical structure organic compounds (with a positive charge), but the anions are much smaller in volume than the cations (with a negative charge) and their structure is inorganic. Due to the difference in size between anions and cations, the bond between the two components of ionic liquids is weak. Some ionic liquids are liquid at room temperature, called RTILs (Room temperature ionic liquids) [74, 77]. Figure 1. below show typical cations and anions that make up the ionic liquids.

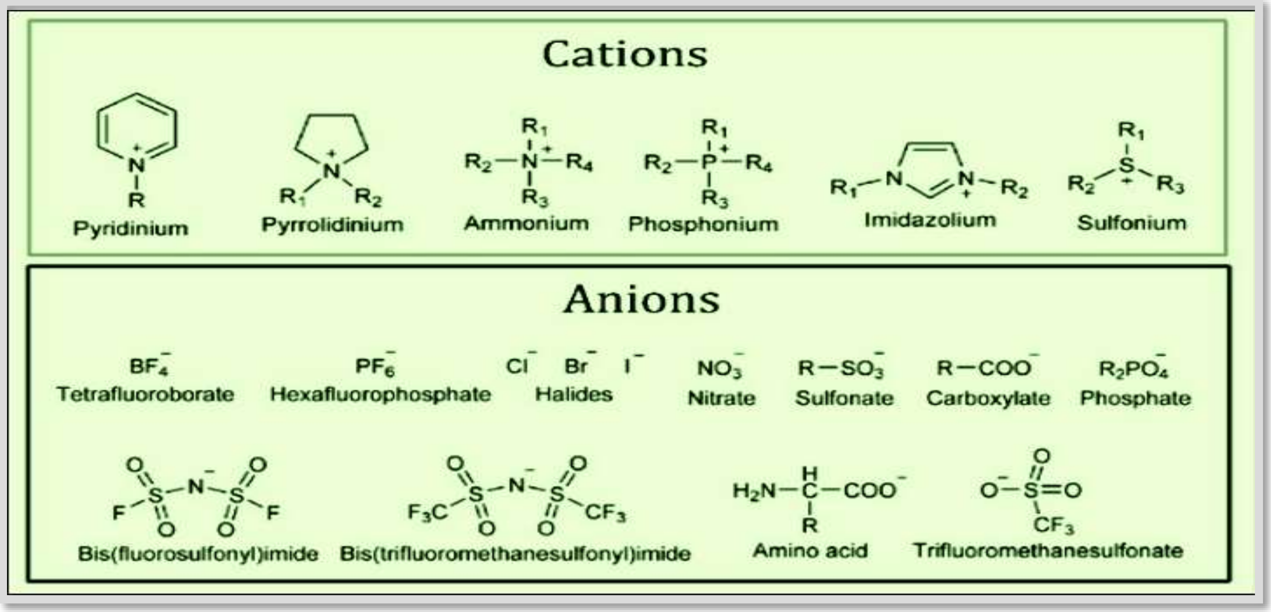

Figure 1--Typical cations and anions in ionic liquids. [78]

Typical basis of the cations are ammonium, imidazolium, sulfonium, piperidinium, and pyridinium ions. Halides, tetrafluoroborate, nitrate, sulfonate, carboxylate, phosphate, amino acid, among others, serve as a base of the anions for the preparation of ILs.[79]. There are infinite combination of anions and cations which leads to IL of specific uses or enhanced physical-chemical properties. While anions are responsible for qualities like air and water stability and cations on the other hand are responsible for melting temperature and organic solubility. Ehsan and Sajjad [74] based on such combinations/properties have classified ionic liquids into three generations namely:

First generation: Ils with unique tunable physical properties .

Second Generation: Ils with targeted chemical properties combined with chosen physical properties. These compounds have one or more specific functional groups on the cation that can interact and play a specific chemical role. They are used as lubricants and complex ligands

Third generation: Compounds with targeted biological properties combined with chosen chemical and physical properties. Such compounds have a structure that has classical ionic units and are biologically active and their toxicity has been investigated. Compounds which have very low toxicity ions can be used as drugs.

3.2.1 Room temperature ionic liquids-RTIL

RTILs are salts that are liquid at room temperature. They are intrinsically interesting because they simultaneously have properties that are similar to organic liquids and liquid salts [80]. RTILs are usually composed of an organic cation, usually bulky and low symmetry in nature, and an inorganic anion. These ILs are dominated by salts [81] derived from 1-methylimidazole, i.e., 1-alkyl-3-methylimidazolium. Examples include 1-ethyl-3-methyl- (EMIM), 1-butyl-3-methyl-(BMIM), 1-octyl-3 methyl (OMIM), 1-decyl-3-methyl-(DMIM), 1-dodecyl-3-methyl- docecylMIM). The organic cation has alkyl chains of various lengths. The disorder in the liquid produced by the presence of the alkyl groups lowers the temperature for crystallization below room temperature and can also result in super cooling and glass formation rather than crystallization. The anions present in the IL are both inorganic and organic anions: fluorous anions such as PF6 ´, BF4 ´, CF3SO3 ´, (CF3SO3)2N´ and non-fluorous anions such as Al2Cl7 ´, Al3Cl10 ´, Au2Cl7 ´, Fe2Cl7 ´. The most widely used ILs are the ones with PF6 ´ and BF4 ´ anions [82]. The properties of ILs are determined by mutual fit of cation and anion, size, geometry and charge distribution, hydrogen bonding, polarity and dispersive interactions. Example of application include; Self-metathesis of oleochemical feedstocks with Grubbs and Hoveyda-Grubbs catalysts using 1,1-dialkyl and 1,2,3-trialkyl imidazolium type ionic liquids [bmim][X] where X = BF_4^-, PF_6^- and NTf_2^- and [bdmim][X] where X = BF_4^- and PF_6^-.[83].

3.2.2 Specialized ILS

Specialized ILs like Protic ionic liquids (PILs), a novel category of inexpensive and sustainable solvents, have been employed to solve industrial and medicine solubility problems. Polyphosphoric acid as a solvent-free protic liquid electrolyte, which excludes the demerits of solvent and exhibits unprecedented superiorities, enables $MoO_3/LiVPO_4F$ rocking-chair battery to operate well in a wide temperature range from 0 °C to 250 °C and deliver a high power density of 4975 W kg^{-1} at a high temperature of 100 °C. The solvent-free electrolyte could provide a viable route for the stable and safe batteries working under harsh conditions [84].

Other innovative applications of ILs are in some organic electronic devices such as organic light emitting devices, photovoltaics, and organic field effect transistors.[85]

3.2.3. Poly (ionic liquids)-PILs

PILs, also called polymerized ionic liquids, refer to a subclass of poly electrolytes that feature an ionic liquid species in each monomer repeating unit, connected through a polymeric backbone to form a macromolecular architecture. Some of the unique properties of ILs are incorporated into the polymer chains, giving rise to a new class of polymeric materials. PILs are generally synthesized by the direct radical polymerization of IL monomers. They are commonly reported to have rather broad glass transition temperature (T_g) ranges, despite their high charge density. Due to the superior process ability afforded by their polymer nature, they are able to form transparent films of different thickness by spin coating or solution casting. Examples of recent applications of PILs includes thermo responsive materials, carbon materials, catalysis, porous polymers, separation and absorption materials, and energy harvesting/generation as well as several biological applications[86-88].PILs have been recently recognized as innovative poly electrolytes attracting rapidly increasing interest in a multitude of fields of polymer and materials science.

3.3 Synthesis of ionic liquids

Ionic liquids can be mainly grouped into two main categories: simple salts -made up of single cation and single anion and ionic liquids- salts in which equilibrium is involved [89]. Two major steps involved in their synthesis are;[90].

- Formation of the desired cation**:** can be achieved by the protonation of an amine or through quaternization reaction (SN2-reaction) with an alkylating agent and heating the mixture. Typical alkylating agents used are alkane halides.

- Exchange of anion***:*** In cases where it is impossible to form the desired anion directly within the first step, two different pathways to vary the anion are possible. The anion-exchange can be realized via Lewis-acid-base reaction or via anion metathesis [91,92]. Both types of reactions are carried out from the halide salts of ionic liquids. Typical Lewis acids used in this context are $AlCl_3$, BCl_3, $CuCl_2$, $FeCl_2$, or $SnCl_2$. Figure-3-below present the synthesis path for preparation of an imidazolium based ionic liquid. [90].

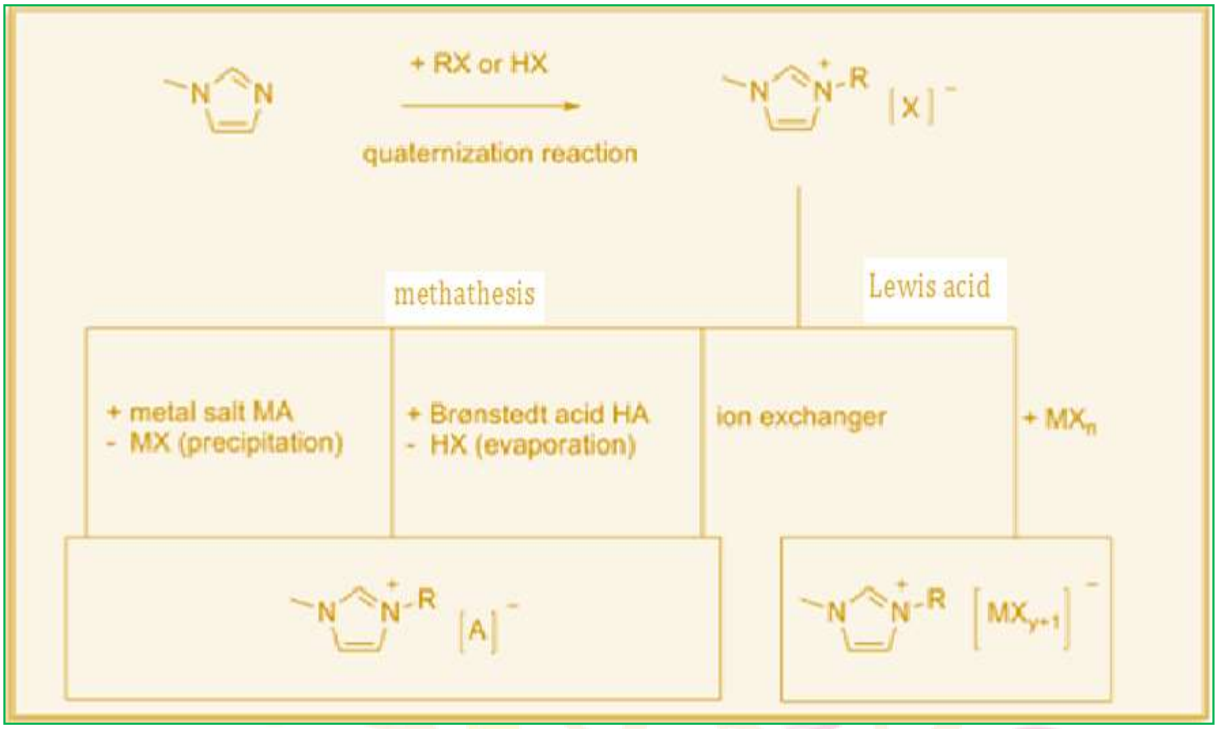

Figure-2: Synthesis path for preparation of an imidazolium based ionic liquid [90]

Several new and upgrade on conventional methodologies have been developed for synthesis of Ionic liquids such as irradiation with microwave, sonication, ring opening, acid-base neutralization, power ultrasound and many more [77,93] .

3.4 Ionic liquids-purity

The purity requirement of an ILs depends on its application. The presence of impurities can change their physical and chemical properties. Therefore, the purification of ionic liquids is essential. The most important ionic liquid pollutants are halides or organic and water based substrates, which usually come from unreacted materials. Ionic liquids have a strong tendency to absorb moisture, so hydrophobic ionic liquids are also moisture-absorbing. In general, ionic liquids are dried by heating under vacuum, but it is difficult to completely remove water (due to the strong hydrogen bonding). The presence of water reduces the density and viscosity and modulates the chemical properties [94, 95]. Therefore, in order to minimize impurities, and maximize the purity of an IL, it's better to remove impurities from the starting materials, select a method of synthesis that brings in few side reactions and accommodates the easy separation of impurities from samples [96.].

3.5 Birth of deep eutectic solvents (DES).

The high expectations of ILs as green media were however faced with some challenges [97, 98]. Their toxicity, costly synthesis and poor degradability impeded their application in industry [99-101]. To overcome these problems of ILs, a new type of solvents were developed, DES, also termed as deep eutectic ionic liquids (DEILs), low-melting mixtures (LMMs) or low transition temperature mixtures (LTTMs) [98]. The first set of DES was obtained by mixing a quaternary ammonium salt with hydrogen bond donors such as organic acids, urea or glycerol that form a complex with the halide anion of a quaternary ammonium salt (e.g. choline chloride) and various carboxylic acids [101]. The physicochemical properties of DES are similar to those of ILs, except that most DES are made of non-toxic, easily accessible, cheap sustainable compounds and include also non-ionogenic compounds [102,103] as well.

3.6 Physical properties of ionic liquids

To optimize the use of ILs and design the desirable ILs, knowledge of the physical and chemical properties of ILs is of prime importance. From the industrial viewpoint, a fundamental understanding of f ILs properties is a must before its industrial application. For academic research, these properties are also indispensable to validate the theoretical models or select proper

ILs. For example, densities, viscosities, and surface tensions determine important parameters including rates of liquid-liquid phase separation, mass transfer, power requirements of mixing, and pumping [104].

Mohd Farooqh Javvad Ali et al; and others [105-110] have reviewed various properties of ionic liquids such as density, viscosity, hydrophobicity, solvation of ionic liquid system. These properties are susceptible to be influenced by the chosen anion or alkyl chains of the cations. Brief details of some critical properties are:-

- **Density**

The density of the ionic liquids is generally higher than the density of water. The magnitude of density, ρ, depends upon the constituent cation and anion. For instance the ρ, value of ionic liquid varies with the length of the N-alkyl chain on the imidazolium cation. As a thumb rule, the density of comparable ionic liquids decreases with the increase in the bulkiness of the organic cation. It lies in the range 0.80-2.10 g/cm^2.

- **Viscosity**

Viscosity of ionic liquids is of immense importance and plays a major role in many conditions. Ionic liquids are generally viscous with a broad range of viscosity from 7-1800 mPa-s when compared with aqueous amine solvents. The viscosity of [EMIM][Tf2N], for instance is 28 cP, while that of [BMPYRR][NMs2] 1680 cP at 298 K. [BMPYRR] stands for 1-butyl-1-methylpyrrolidinium, whereas [NMs2] for bis-(methanesulfonyl)amide species [111].

- **Hydrophobicity**

The formation of hydrophilic or hydrophobic ionic liquids depends upon proper selection of cations and anions. The increase in the length of alkyl chain in the cations is responsible for the development of hydrophobicity. The anions such as [PF6] – and (CF3SO2)2N– are responsible for making ionic liquids immiscible in water, whereas, the nitrates, acetates and trifluoroacetate are responsible for making ionic liquids miscible with water.

- **Melting Point**

This is the most significant characteristic property of ionic liquids that can be correlated with the structure and composition of ionic liquids. Selection of both the cation and anion determine the melting point of an ionic liquid. The melting point of the ionic liquids is very less. It ranges between –100 to 113^{o}C. Most of the ionic liquids are in molten form at the room temperature. By increasing the substituent chain length, the melting point can be increased. Melting point of the ionic liquids decreases with the increase in the amount of water molecules available in the ionic liquids.

- **Vapor Pressure**

The vapor pressure of the ionic liquids at room temperature is negligible.

- **Polarity**

Polarity is one of the most important properties of ionic liquids and an essential requirement when choosing an IL for a specific industrial application. It is a description of the potential behaviors of the solvent in a relationship with the solute but not an absolute property of the pure liquid. Hence, there is no single measure of polarity. Kamlet–Taft solvent polarity scales a, b, and p have been adapted for use with ionic liquids [112].

- **Thermal stability**

Most of the ionic liquids are stable at and above 400^{o}C. The thermal decomposition depends on the nature of anions rather than on that of cations. Further thermal decomposition also decreases with increase in hydrophilicity of anions.

- **Solvation Properties**

Solvation is the process of the interaction between a solute and a solvent. The solvent's polarity is critical in determining how well it solvates the solute. The solute particles interact with the solvent. By changing the constituent ions, the solvating properties of the ionic liquids can be changed. With the help of ethers, hydrocarbons and various organic solvents, the ionic liquids can be made miscible or immiscible. This converts them into a very beneficial solvent when combined with the characteristic of negligible vapor pressure.

4. Ionic Liquids-Applications

Since their conception, ionic liquids have been investigated for an extensive range of applications, in solvent chemistry, catalysis, and electrochemistry. This is due to their designation as ‘designer solvents’ also branded as “solvents of the future” in 2003”, whereby the physiochemical properties of an IL can be tuned for specific applications [113]. Some such properties of ILs include high polarity, negligible volatility, high thermal stability, high ionic conductivity, low melting point, and structural design ability [114]. The structural design ability can be exploited in tuning the properties of ILs and making them task specific for challenging applications where molecular liquids cannot be used [56]. In view of such an observation, while ILs have uses as alternative solvents, many other applications are also of importance [115].

ILs have been used in a number of industrial processes, particularly to improve catalyst recovery and product separation through the formation of biphasic systems. Their solvation properties are greatly influenced by the ions’ ability to act as hydrogen bond acceptors/donors, and the degree of delocalisation of the anionic charge [116]. ILs could be used, particularly as replacements, for traditional solvents in industrial processes which are often toxic, flammable and highly volatile [117].

Due to unique properties of the ILs, there is a vast interest in applying them as a reaction medium in a wide variety of chemical transformations that until recently could only be carried out in organic solvents [118]. Many publications describe numerous applications /uses of ILs. Some important of them are: reaction media for organic transformations [119], in separations and extractions [120], as electrolytes for electrochemistry [121], in nanotechnology [122], in biotechnology [123], and in engineering processes [124], absorption of gases (CO_2) [125], as catalysts in organic synthesis [126], aldol condensation [127], organometallic and radical polymerization [128]. Some ILs applications, subject wise are depicted below in Figure 3.

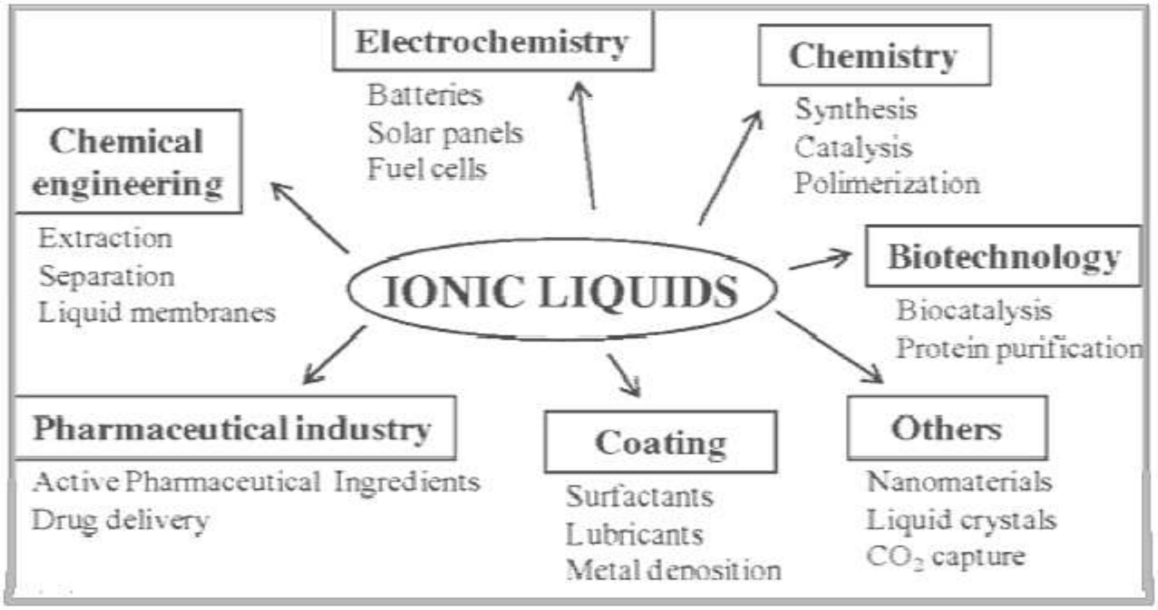

Figure 3: Illustration of some well-known subject wise applications of ionic liquids.

4.1 Pilot scale or potential and commercial applications

Since the commercialization of the first known IL-based process in 1996, there has been an explosion in their use for a diverse range of applications. As seen from the figure 3, ILs have vast applications in different chemical, biotechnological and other disciplines. It is nearly impossible to describe the same in any one write up. Accordingly, this article will primarily confine to types of applications/processes falling in the scope of electrochemistry, pharmaceutical and petroleum industry. Further description in this part of the chapter therefore relates to ILs based important subjects of research that hold the potential of commercialization in the next few years followed by some known processes that are operating at commercial scale.

4.1.1 Potential or pilot scale applications

1. **Biopolymer processing**

Biopolymers such as chitin/chitosan, starch, alginate, collagen, gelatin, keratin, and fibroin [129] have been processed and modified using ILs via dissolution, extraction, purification. For example, ILs enables the preparation of biopolymer materials in different forms (e.g. sponges, films, micro particles, nanoparticles, and aerogels) and better biopolymer chemical reactions, leading to biopolymer-based drug/gene-delivery carriers [130].

ILs also enable the synthesis of chemically modified starches with high efficiency and degrees of substitution and the development of various starch-based materials such as thermoplastic starch, composite films, solid polymer electrolytes, nanoparticles and drug carriers [131].

2. Materials for biomedical applications

ILs and their tailor able properties as well as their rich synergetic effect in their combinations with natural and synthetic polymers have been used for biomaterials preparation, improving dissolution and processability. As an example, the exploration of the magnetic properties of ILs and magnetic ILs/polymer materials, for tissue regeneration hold interesting potentials. [132].

3. **Protein and DNA Chemistry**

IL's ability in stabilizing or destabilizing the 3D-structure of a protein or the double-helical structure of DNA has been assessed superior to the water and volatile organic solvents. Ions interaction toward the protein backbone is driven by specific ion-protein interactions and not necessarily due to the perturbation of the bound water to the protein. These observations were confirmed as valid in several studies utilizing ionic liquids. Comparison of the stabilizing power of SCN−, SO2− 4 , HSO−4 Cl−, Br−, I−, and CH3COO− for ionic liquids with 1-butyl-3-methylimidaolium ([bmim]+) cation and for inorganic salt with sodium ion on α-chymotrypsin (CT) showed that the SCN−, SO2− 4 , HSO−4 Cl−, Br−, I−, and CH3COO− of sodium salt have. a destabilizing effect whereas the Cl−, Br−, and CH3COO− with [bmim]+ have a stabilizing effect on the CT.

In the case of DNA, ionic liquids emerged as a natural long-term stabilizer and nuclease inhibitor that causes slow degradation to the stored DNA [133].

4. **Recovery of valuable metals and waste recycling**

Due to their multitude and unique tune able characteristic, ILs and DESs, have enabled energy-efficient design, recovery, recycling, and separation of valuable metals from urban mining and electronic wastes. Functionalized ILs, solid extraction, SILPs, and agricultural waste-based IL show great promise for sustainable high-value metal recovery [134].

ILs can aid the recycling of synthetic goods, plastics, and metals. They offer the specificity required to separate similar compounds from each other, such as separating polymers in plastic waste streams. This has been achieved using lower temperature extraction processes than current approach and could help avoid incinerating plastics or dumping them in landfill [135].

5. Ionic liquids in two-dimensional materials

ILs are being intensively researched to be used as readily available "designer" solvents, characterized by a number of tunable properties. High ionicity among them is outstanding in the preparation and property modulation of two-dimensional (2D) materials. Electric-field controlled ILs-gating of 2D material systems have shown novel electronic, magnetic, optical and superconducting properties, attracting a broad range of scientific research activities particularly in the recent developments of ILs modified 2D material systems from the electrochemical, solar cells and photocatalysis aspects [136].

6. Organometallic chemistry

Carborane anions have a number of unique properties: they are extremely inert, non -coordinating and stable and can be derivatized and tailored for specific applications, to improve solubility or polarity. Most modern homogeneous catalysts are based on cationic transition metal complexes, and thus follow the importance of the suitable anion that would not block the reactive metal center. Carboranes anions are considered perfect to address this issue. Currently researchers are working to develop catalytically active ruthenium-based cationic systems coupled with carborane anions. Another recent research project involves synthesis and reactivity studies of the potential catalysts based on rhodium pincer complexes with a view to develop a better catalyst for sterecontrolled alkyne dimerization [137].

7. Dissolution and recovery of lithium

The direct dissolution of Li_2CO_3 or LiTFSI (obtained from Li_2CO_3) has been demonstrated in [MPPi][TFSI]. The solutions prepared from both chemical species were identical in terms of their electrochemical responses indicating Li+ is the relevant species dissolved in IL. The production of nucleation sites using cyclic voltammetry was demonstrated and strongly impacted the bulk deposition of lithium on a gold electrode. The pulsed potential method produced uniform and compact deposits that adhered to the electrode surface, thereby suggesting that this could be the potential route for the recovery of lithium from spent new generation LIBs in the form of LiH [138].

8. Lubricants and tribology

Development and synthesis of ILs-based lubricants for tribological applications is an important current research subject. ILs, as neat lubricants or additives in lube oils, with a focus on ILs chemistry, synthesis, miscibility, and other relevant rheological, thermal, and tribological properties at macro to nanoscales are being investigated, as also lubrication mechanisms based on the tribofilm that forms on the rubbing interfaces owing to tribochemical reactions among the ILs, base oil, and solid bodies, which gives key insights into anti-wear properties. Initial results indicate ILs exhibit superior anti-wear properties compared to fully formulated oils [139].

9. CO_2 capture

Recent studies using ILs and their modified versions on CO_2 capture are a great boom to environment protection. Two examples of research paths showing improved CO_2 separation and economics are:

- **Low pressure CO_2 capture**

New porous ionic liquids, based on the ZIF-8 metal-organic framework (MOF) and phosphonium acetate or levulinate salts, show an increased capacity to absorb CO_2 at low pressures. This ILs combination absorb reversibly 103% more CO_2 per mass than pure ZIF-8 at 1 bar and 303^0 K. The study showed how the rational combination of MOFs with ionic liquids could pave the way towards a new generation of high-performance liquid materials for effective low-pressure carbon capture [140].

- **Application of DESs**

Mihaila et al; [141] report on the application of deep eutectic solvents. The CO_2 absorption-desorption capacity of two choline chloride-based DES, ethaline (ChCl:EG, 1:2 molar ratio) and reline (ChCl:U,1:2 molar ratio), was studied. The tests showed that ethaline had 3x better CO_2 absorption and desorption capacity than reline, attributed to weaker intermolecular interactions. Increasing temperature and pressure increases the absorption capacity of the two eutectics due to physical changes (lower viscosity and density), allowing easier penetration of the CO_2 molecule into the 'eutectic cage'. Conversely, reducing temperature and pressure induces desorption. This project is at the demonstration stage.

10. Oxidative desulfurization (ODS) of gas oil

Hoda A. Mohammed et al; [142] have successfully synthesized and characterized a novel Gemini IL (N1,N1,N3,N3-tetramethyl-N1,N3-diphenylpropane-1,3-diaminium dichloride) using 1 H-NMR and FT-IR spectroscopy. It was then used as a catalyst in ODS of real gas oil with a sulfur content of 2400 ppm. Te N1,N1,N3,N3-tetramethyl-N1,N3- diphenylpropane-1,3-diaminium dichloride IL exhibited a high desulfurization efficiency of 84.7% under optimal conditions (H_2O_2:Oil=0.4:1, IL:Oil =0.02:1, reaction temperature=70 °C, and reaction time=3 h). Based on the thermodynamic analysis of the ODS process, the values of $\Delta H°$ indicated that the reaction is endothermic with increase the temperature. Te IL can be directly reused, and they exhibited good recyclability for six times. Swapnil A. Dharaskar et al; [143] earlier attempted similar approach for the Extractive Deep Desulfurization of Liquid Fuels Using Lewis-Based Ionic Liquids but with less promising results.

4.1.2 Commercial Applications

1. Ionic liquids in bio-medicines

ILs and IL-modified materials have been adopted as highly promising tools to improve the downstream processing and formulation approaches of biopharmaceuticals, due to their remarkable properties, selective, and easy to scale-up techniques for the recovery, purification, and formulation. Several IL-based approaches, most of them relying on IL-ABS (IL-based Aqueous Biphasic Systems) and SILs (supported Ionic Liquids), have been proposed to enhance the recovery yields and purification efficiency of a myriad of biomolecules, including therapeutic enzymes, antibodies, nucleic acids, viruses, and interferons[144].

In the downstream processing, IL-based ABS/ATPMS (Aqueous Micellar Two-Phase Systems and TTP (Three-Phase Partitioning) systems and SILs have shown the ability to improve the purity and yield of a myriad of biopharmaceuticals. Research showing the possibility of performing biomolecules' purification in continuous mode with ABS and CPC, and the identification of ILs as multimodal ligands in preparative liquid chromatography, seems particularly relevant. It is also important to emphasize the capacity of ILs to be recovered and reused, contributing to the development of more sustainable and low-cost processes. In the formulation field, ILs have been shown to have high potential to improve the thermal and chemical stability of several therapeutic compounds, and to inhibit their aggregation, denaturation, enzymatic degradation, and fragmentation.

ILs and SILs-based processes represent good alternatives when aiming to reduce bio-manufacturing-associated environmental, economic and health burdens. The research conducted so far shows the remarkable potential of IL-based approaches in the biopharmaceutical field.

The use of ionic liquids within biomedical nanocarriers [145] was initiated by their use for synthesis and templating of silica nanoparticles for antibacterial, gene transfection, and drug release applications. ILs have also recently been used in the synthesis of nanocarriers to improve many of their characteristics, including biocompatibility, stability, and drug loading. IL-based nanocomposites have been used to increase drug solubility and efficacy of the anticancer drug rutin. Rutin has a strong promise as a treatment for renal cell carcinoma but is limited by its poor water solubility and pharmacokinetics. The researchers were able to encapsulate choline amino acid-based ILs (phenylalanine and glycine) in poly (lactic-*co*-glycolic acid) nanoparticles and achieve similar toxicity of free rutin with a tenfold increase in drug concentration in water[146]

Rutin has also been named as rutoside, vitamin P, quercetin-3-O-rutinoside, and sophorin, with the chemical formula, C27H30O16 and a molecular weight of 610.53.Rutin is demonstrated to inhibit the proliferation of breast, colon, lung, and prostate cancers and other tumors. By way of commercial manufacture, two main facilities are at Athena facilities at RPOUVY north of Paris in France and at Ambernath east, of Mumbai in India [147].

2. Electrochemical Devices

- **Electrochemical sensor electrodes-AZO sensors London**

High electrochemical sensitivity has been achieved by using graphene, a 2-dimensional (2D) nanocarbon structure, as the main electrode material or as an addition to increase electron transport between a target analyte and the electrode surface. Screen-printed graphene electrodes (SPGEs), in particular, have been shown to have a significantly greater electrochemical response than screen-printed carbon electrodes (SPCEs).

SPGEs and SPCEs surface-modified with imidazolium-type and pyridinium-type ILs, in particular, have demonstrated dramatically better electrochemical responses to a variety of analytes.

Disposable screen-printed IL/graphene electrodes (SPIL-GEs) were created in a new study published in the journal *Electrochemistry Communications* utilizing pastes made by mixing ILs with electrolytically exfoliated graphene and carbon paste (CP) in a ball mill.

The electrochemical response of the optimum SPIL-GE was compared to SPCEs and SPGEs for three common electroactive analytes: (Fe(CN)$_6$) redox couple, DA, and HQ. For the three analytes, the results showed that SPIL-GE had higher oxidation currents and lower anodic potentials than SPGE and SPCE. As a result, the SPIL-GE may be a viable option for advanced electrochemical sensing applications [148].

- **Ionic liquid based sodium oxygen batteries**

The electrochemical properties and performance of four different air cathodes in ionic liquid based sodium oxygen (Na-O$_2$) cells were examined, focusing on the roles of the cathode morphology, structure and composition. Cathodes with a microporous layer (MPL) and hydrophobic character gave higher discharge capacity in both half cells and full Na-O$_2$ cells employing a 16.6 mol % [NaTFSI]/[C$_4$mpyr][TFSI] electrolyte. The highest discharge capacity was found for a cathode with a pore size *ca*. 6 nm; this was over 100 times greater than that delivered by a cathode with a pore size less than 2 nm. The air cathode with the highest specific surface area and the presence of a microporous layer (BC39) exhibited the highest specific capacity (0.53 mAh cm^{-2}) [149].

- **Rechargeable lithium ions batteries**

Rechargeable batteries are the scientific response to rapid development of smart and wearable gadgets, electric vehicles and many other ultraportable devices. These batteries have features like enhanced safety, high-performance, high gravimetric and volumetric energy density, built with the support of new battery chemistry. The new chemistry consistently explores better electrolytes that are environmental-friendly, nonflammable, reusable and most importantly ultra-customizable for high-performance applications. In future there is a scope to use ILs as sole electrolyte (ILs-based gels or polymer electrolytes) containing lithium salts in combination with the advanced electrode materials to provide high thermal stability. LIBs were introduced to the market in 1991 by Sony (Tokyo, Japan) [150].

- **Supercapacitors**

Electrochemical capacitors, also known as supercapacitors (SCs) are promising energy storage devices that store electrical energy at the interface between the electrode and electrolyte. Micro-supercapacitors (MSCs) offering high-energy and power performance are also a class of supercapacitors with a miniaturized configuration. SCs and MSCs are emerging as high-performance electrochemical energy storage and clean renewable energy generation devices that supply power for various electronic devices, including hybrid vehicles, portable electronics, military devices, space equipment, next-generation electric cars, microdevices, and internet of things. IL-based gel electrolytes exhibiting a wide potential window, less volatility, high conductivity, and good mechanical strength are considered an advantageous alternative to typical liquid electrolytes [151]. In 2020, Asbani et al. developed and tested 3D MSCs using MnO$_2$ electrodes and a sol–gel derived IL-based gel containing PVDF in DMF and EMIMTFSI The IL-based gel-based MnO$_2$ MSC delivered excellent cycle life over 30,000 cycles [152].

- **Electro deposition**

Green ILs solvents due to their inherent conductivity, act as essential electrolytes for the electro-deposition of metals to minimize the conventional disadvantages of other electro-deposition techniques [153]. Suryanto et al; [154] demonstrated the electro deposition of silver onto metals and metal oxide substrate using protic ILs(ethylammonium nitrate, triethylammonium methylsulfonate and bis(2-methoxyethyl)ammonium acetate). The results revealed that the electro deposition is achieved by a three-dimensional growth process controlled by progressive nucleation and diffusion. Silver nano particle as electro catalysts on electrode exhibits outstanding catalytic activity in the oxygen reduction process. The study concludes that protic ILs might be an alternate electrolyte for metal electrodes and nano structured electro catalysts.

Motobayashi group (Osaka Japan) has developed a new electro deposition method of cobalt (Co) in the presence of IL, which produces interfacial multilayer of ions. This method generates high over potential (HOP), which helps to reorganize the interfacial multilayer structures.

The IL technology is used in a variety of metal processing applications, including electroplating and electropolishing processes. Scionix's founder (Abbott) has stated that there are two processes running on a commercial scale (>1 tonne), and seven further pilot scale processes (50-250 kg) for a range of different metals [155,156]. Presently research work on developing methodologies for more complex casting methods such as single crystal super alloy castings which is important for the automotive and aerospace industries, is in progress.

3. Applications in Petroleum Industry

- **Alkylation using ionic liquids**

The predominant chemistry for alkylation process can be represented as:

iC4 + C4= ➔ C8 alkylate
iC4 + C3= ➔ C7 alkylate
iC4 + C5= ➔ C9 alkylate

A strong acid is required to catalyze the reaction and refineries currently use technologies with either hydrofluoric acid (HF) or concentrated sulfuric acid (H_2SO_4) catalyzing the reaction.
Research and development in ionic liquids led to the discovery that certain ionic liquids are very effective at alkylation of olefins. The ionic liquid catalyst used in the ISOALKY Process is a state-of-the-art chloroaluminatebased ionic liquid catalyst. The ionic liquid alkylation catalyst can be characterized by the general formula Q+A−, wherein Q+ is an ammonium or phosphonium cation and A− is a negatively charged ion such as AlCl4 − or Al2Cl7.The specific ionic liquid selected for use as the ISOALKY Catalyst was chosen for its activity and selectivity for alkylation process and long-term stability. A trace amount of anhydrous hydrogen chloride (HCl) co-catalyst is needed to maintain the alkylation reactivity for extended periods of operation, and this is generated *in-situ* by organic chloride promoter addition [157].

ISOALKY is a groundbreaking new technology for refiners, and a lower-risk and economical solution compared to conventional liquid acid technologies that produce alkylate. Ionic liquids have strong acid properties that enable them to produce alkylate without the volatility of conventional acids, allowing for simpler handling procedures [158].

- **Mercury removal from natural gas**

Abai, M.et al have investigated application of chlorocuprate(II)-based ionic liquids for the direct oxidation of elemental mercury,[159] and the use of these ionic liquids in a solid-supported ionic liquid phase (SILP)[160] for reactive capture of mercury from gas streams, which has led to the scale-up and deployment on industrial plants in Malaysia, operating for the past 5 ears[161,162].

5. Conclusions and Future Trends

Wide range applications of green solvents include: from synthesis to oil extraction, sensors and biosensors, CO_2 capture, biomass utilization and bio-based chemicals. Pharmaceutical industry accounts for mass consumption of solvents in the range of 80–90%. Bio-solvents such as butanol, polyethylene glycol, bio-renewable solvents, and supercritical solvents are widely used in the extraction and solubility of drugs.

Due to their specific properties, ILs have been explored for a big range of applications, of which biomedical, electrochemical, chemical separations and pharmaceutical applications, are of particular interest. Their use in industry is not confined to only bulk solvent applications and catalysis, with many niche processes now arising in areas of performance additives and analytical materials. Strong solvation properties and their tunable nature offers significant advantages in comparison to conventional solvents, however, the drop-in replacement of ILs in a process does not often occur without further consideration of process compatibility. Currently in the area of biomedical materials, the most commonly used ILs remain protein-derived amino-acids cations and anions and choline cations, but also other types are emerging in fields such as cancer treatment, biocides, or bio-sensing. The most common use of ILs is as green solvents for a wide range of polymers and other materials. As an example, the exploration of magnetic properties of ILs and magnetic ILs/polymer materials for tissue regeneration hold interesting potential.

IL-based gel electrolytes exhibiting a wide potential window, less volatility, high conductivity, and good mechanical strength are considered an advantageous alternative to typical liquid electrolytes. In 2020, Asbani et al. developed and tested 3D MSCs (super capacitors with a miniaturized configuration) using MnO_2 electrodes and a sol–gel derived IL-based gel containing PVDF in DMF and EMIMTFSI. The IL gel-based MnO_2 MSC delivered excellent cycle life of over 30,000 cycles.

Rechargeable lithium batteries are the scientific response to rapid development of smart and wearable gadgets, electric vehicles and many other ultraportable devices. These batteries have features like enhanced safety, high-performance, high gravimetric and volumetric energy density, built with the support of new battery chemistry that consistently explore better electrolytes that are environmental-friendly, nonflammable, reusable and most importantly ultra-customizable for high-performance applications.

The ionic liquid catalyst used in the ISOALKY process to produce high octane alkylate-a component of gasoline pool, is a state-of-the-art chloro-aluminate based ionic liquid catalyst. The ionic liquid alkylation catalyst can be characterized by the general formula Q+A−, wherein Q+ is an ammonium or phosphonium cation and A− is a negatively charged ion such as $AlCl_4$ − or Al_2Cl_7.The specific ionic liquid selected for use as the ISOALKY Catalyst was chosen for its activity and selectivity for alkylation process and long-term stability.

ILs are typically more expensive than conventional solvents, however, the initial increase in capital cost can be offset by improvements in solvent recyclability, catalyst recovery, reaction rates, selectivity, and product separation, as has been shown by a number of processes discussed earlier. It is the major drawback of applying ionic liquid in large quantity which so far has restricted their use in the oil and gas industry. However, if cost effective mechanisms are found to synthesize ILs on a large scale, they could be widely used to replace many potentially harmful and volatile organic solvents and chemicals currently used in the oil industry with economic benefits [163]. Additionally, attention should be given to alternative classes of solvents such as switchable solvents and porous liquids which have the possibility to provide benefits to the same types of processes in the future.

The solvent and catalyst application market will remain the major end-use market for ionic liquids by value during the forecast period, followed by separation processes, electrochemical devices and biotechnology. However, the electrochemical device market is expected to grow at the largest CAGR during the forecast period [164].

Author Contributions:: original draft preparation, conceptualization, writing—review and editing of the manuscript.

Funding: All funding managed by the author himself

Acknowledgments: Author deeply acknowledges the support of my daughter Jasdeep in the preparation and formatting of this manuscript.

Conflicts of Interest: Authors declare no conflict of interest.

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