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Review Article

VERSATILE APPLICATIONS OF THE IONIC LIQUID

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ABSTRACT

The first ionic liquid Ethyl ammonium nitrate was discovered in the year 1914. Ionic liquids are organic salt that is composed of organic cations and organic or inorganic anions. Ionic liquids subsist as liquid below the 100˚C or in the room temperature. A huge number of ionic liquid can be synthesized by the amalgamation of the various anions and cations through numerous kind of bonds. Ionic liquids are considered as green solvents because of non-volatility, low-viscosity, colourless, polar, wide range of solubility of biopolymers, absorption and desorption of gases. The environmental factor (E-factor), atom efficiency also supported the greenness of the ionic liquid. In chemistry, ionic liquid can replace the usual volatile organic solvents, mineral acids, solid acids etc., due to the non-corrosive green nature. For this reason, ionic liquids can be employed to numerous organic syntheses. The synthesis of ionic liquids can be done in single or multiple steps. Microwave irradiations and ultrasound assisted reactions are commonly employed to synthesise the green ionic liquids. Purification of ionic liquids can be done through crystallisation, extraction, distillation, biphasic separation and also through composed method of centrifugation or membrane separation followed by distillation. Broadly, ionic liquids are classified as basic ionic liquids, protic ionic liquids, neutral ionic liquids, chiral ionic liquids, metallic ionic liquids, poly ionic liquids, bio-ionic liquids, task specific ionic liquids etc. The major application of ionic liquids in the field of solvents and catalysts, electrochemistry, physical chemistry, analytical chemistry, pharmaceutical, biological, engineering field is well documented in this paper.

Keywords: Ionic Liquid, Green solvent, Designer solvent, Synthesis and Application, Physicochemical property, Biological activity.

1. INTRODUCTION

Most of the ionic liquids are the molten organic salt possessing melting point below 100˚C. Generally, Ionic liquids exist as liquid at room temperature. Ionic liquids are considered as third group of good molecular and/or green solvents and electrolytes after water and organic solvents. Ionic liquids are used as the sovents in many oragnic reactions. High ionic conductivity, nonvolatilty and high thermal stability are the exclusive properties of ionic liquid. However these said properties of ILs are controlled by the columbic interactions and vanderwaal's interaction between constituent cations and anions of ionic liquid [1, 2]. Since the invention of first IL Ethyl ammonium nitrate (melting point 12˚C), a number of ILs have been synthesised through standard reproducible methods using a variety of organic cations and organic or inorganic anions. Generally, the organic cations are tetra-alkylated phosphonium, N-substituted pyridinium, derivatives of N, N'-substituted imidazolium, tetra-alkylated ammonium ions. On the other hand, commonly used inorganic anions are copper (III) trichloride, nickel (II) trichloride, tin(III) chloride, iron(III) tetrachloride, choloride, bromide, iodide, nitrate, hydrogen sulphate, dihydrogen phosphate, hexafluorophosphate(V), tetra-fluoroborate and organic anions are p-methylbenezene-sulfonate, alkyl sulphate, acetate, 2,2,2-trifluoroacetate, bis{(trifluoromethyl)sulfonyl}amide [2]. On the basis of Green chemistry principles environmental factor (E-factor), atom efficiency, IL can be considered as a green solvent or catalyst in chemistry. The single step synthesis of 1-methylimidazolium halides incorporates the maximum atom efficiency. The microwave and ultra-sound assisted reactions are useful for the synthesis of desirable ILs because of the advanced selectivity of preferred product, requirement of small energy and reagents, promoting rapid reaction rates [3]. The Life Cycle Assessment (LCA) Analysis of the life cycle of ILs reveals that few ILs are toxic, non-biodegradable. Some of the IL syntheses are done using great quantities of chemicals and solvents with the generation of the huge byproducts as wastes. Researchers have to keep in mind three parameters; recyclability, biodegradability and toxicology while using and the preparation of ILs [4]. Analytical Grades ILs are desired to produce trustworthy data in chemical, physical, biological analysis. In the absence of analytical grade IL, technical grade IL can be employed and a contrast of later IL with the analytical grade may be done through (Eco) toxicological testing. The impurities in the technical grade IL can be eradicated by the use of analytical methods like Gas chromatography, Head-gas chromatography, Nessler cylinders, Karl-Fischer titration, Capillary electrophoretic method etc [2]. ILs can be classified into many types on the basis of composition of cations and anions and their distinctive physical, chemical, thermal properties [5]. Based on this, ionic liquids are classified as basic ILs, protic ILs, neutral ILs, chiral ILs, metallic ILs, poly ILs, bio- ILs, task specific ILs, switchable polarity solvent ILs, etc [2]. ILs can be applied into the various fields of chemitsry like electrochemistry, organo catalyst, solvent, physical chemistry, analytical chemistry, biological as well as phramaceutical aids for the drug development processes [2, 6-7]. Besides this ILs are used as gas storage *e.g.*, hydrogen storage using organosilicon ILs containing carbazole moiety. The theoretical total gravimetric hydrogen capacities of liquid organic hydrogen carriers (LOHC) possessing N-$(CH_2)_3$-Si bond is ~1.58% -2.05% [8]. Traditional cutting fluids have drawbacks like skin irritation, non-biodegradability, elevated toxicity, negatively influencing on the ground water. To overcome this problem, ILs are mixed with cutting fluid and this not only increases the cutting ability but also environmentally safe [9]. Supported ionic liquid phase (SILP) on porus solid materials such as aluminas and zeolites, porous polymers, metal-organic framework (MOF) materials, inorganic silicas, and carbonaceous materials are used for the purification of air by the capture of CO_2, CH_4, H_2S, and NH_3 gases [10]. Especially. imidazolium based ILs are also used as a corrosion inhibitor for metals and alloys in various electrolytic media. The advantages of imidazolium based ILs are environmentally protected, economic, highly soluble in nature [11]. The mixed ILs and porus material shows potential technology on the removal of indoor pollutants like volatile organic compounds (VOCs) such as phenol, toluene and indoor irritant gases such as SO_2, NH_3, H_2S. The mixed ILs and porus materials are environmentally safe, regenerative, recyclable, and little energy consuming. One drawback is that it can only highly adsorb single gas at a time in presence of mixed gas environment. [12]. In this connection, IL based materials are also used for the separation of gas specially CO_2 from the industrial waste stream. ILs are used as an electrolytes in metal plating, solar panels, batteries, fule cells, sensors. ILs are used as a sorbent in coating and as alubricant in lubrications. ILs are used as a matrices in the matrix-assisted laser desorption/ionization time-of-flight (MALDI-TOF) for the detection of ultra-trace of anions in the Mass Spectrometry. ILs are used a delivery agent in the drug delivery processes for the potentially safer and stable formulation. The ILs are resistant to radiation and also non-flammable. For this, ILs can be employed as a solvent in the nuclear waste processing for the greater actinide extraction from spent nuclear fuel. ILs are playing a role as plasticizers for polymer industry. ILs reduce the glass transition temperature (Tg) appreciably with superior leaching and migration resistance [13]. ILs are extensively used for enzymatic reactions of materials which are insoluble in water and common organic solvents. ILs facilitate several enzyme-catalyzed reactions with marvelous yields [14]. This review will discuss about the use of IL in the various fields of chemistry.

2. METHOD OF SYNTHESIS OF IONIC LIQUID

Generally the preliminary step of synthesis of IL involves generation of cation through the quaternization of an amine or phosphane. The quaternization reaction is done through alkylation reaction using alkylating reagent. The alkylating reagents are methyl trifilate, chlorobutane. The cations can also be synthesized by the acidification of an amine. Next anion metathesis or anion exchange reactions are employed to synthesize Lewis acid-based ionic liquids through the reaction of halide salts with Lewis acids [15].

ILs are subdivided into two categories. The first category is simple salt which is composed of simple cation and anion. The second category is binary ionic liquids. $[EtNH_3][NO_3]$ belongs to the first category while the mixtures of aluminum(III) chloride and 1,3-dialkylimidazolium chlorides belongs to the second category. The second category is more important than the first one. Besides $AlCl_3$, other Lewis acids are employed such as CuCl, $InCl_3$, BCl_3, $AlEtCl_2$ [16].

1,3-dialkylimidazolium cations based water and air stable ionic liquids are prepared through anion metathesis method. Some of the examples of anion metathesis based ILs are [Cation]$[NO_3]$, [Cation]$[AuCl_4]$, [Cation]$[CH_3CO_2]$, [Cation]$[BF_4]$, [Cation]$[PF_6]$, [Cation][CF_3

$(CF_3)_3CO_2]$ etc. The anionic source of this ILs are $AgNO_3$, $NaNO_3$, $HAuCl_4$, $Ag[CH_3CO_2]$, HBF_4, NH_4BF_4, $NaBF_4$, HPF_6, $K[CF_3(CF_3)_3CO_2]$ [2, 16]. Microwave irradiation (MW) and power ultrasound (US) are also used for the betterment of synthesis of ILs. The said two methods gives IL using solvent free technology. Using Ultrasound technique N-methyl-2-pyrrolidinium hydrogen sulphate based IL and 1-alkyl-3-methylimidazolium based cation accompanying a group of anions for example halides (Cl, Br, I), BF_4, PF_6, CF_3SO_3 and BPh_4 are synthesized. [2, 14, 17-18]. MW irradiation method is used by Verma and Namboodiri to synthesize 1-alkyl- 3-methylimidazolium halides (chloride and bromide) and dialkyl-3-methylimidazolium dihalides ILs with > 70% yield in less than 2 mins [19]. Chiral and amino acid based ionic liquids are also synthesized through MW irradiation method and these ILs are applied for various purposes. A range of imidazolium based ILs for instance discotic-discotic (triphenylene), calamitic-discotic, calamitic-calamitic (alkoxycyanobiphe) are prepared by MW irradiation method [20].

With the combined method of MW and US, 1-methylpyrrolidine, 1-methylimidazole, pyridine based cations and alkyl halides based anions ILs are prepared. These second generation ILs are air and moisture sensitive and halogen free [21].

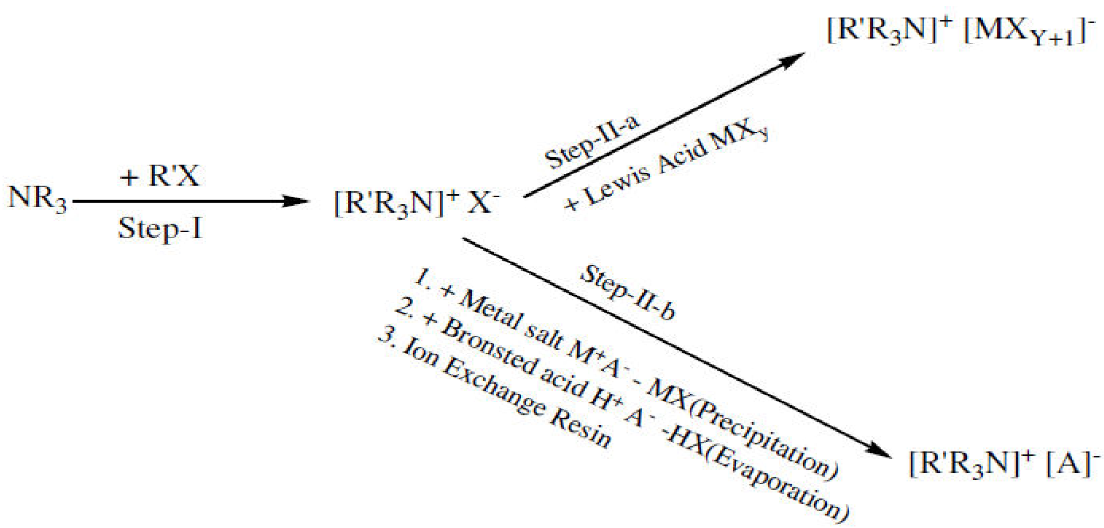


Fig. 1: General synthetic scheme for the generation of Ionic Liquid from Ammonium Salt [15].

$$[emim]^+Cl^- + AlCl_3 \rightleftharpoons [emim]^+[AlCl_4]^- \quad \text{Basic IL}$$

$$[emim]^+[AlCl_4]^- + AlCl_3 \rightleftharpoons [emim]^+[Al_2Cl_7]^- \quad \text{Neutral IL}$$

$$[emim]^+[Al_2Cl_7]^- + AlCl_3 \rightleftharpoons [emim]^+[Al_3Cl_{10}]^- \quad \text{Acidic IL}$$

Scheme 1: Series of equilibria beteween $[emim]^+Cl^-$ and $AlCl_3$. Here $[emim]^+$ is 1-Ethyl-3-methylimidazolium ion [16]

3. PURIFICATION OF IONIC LIQUIDS (ILs)

The synthesized ILs are accompanied by the by-products such as water, organic solvents, acids, salts. To keep in mind about the environmental and economic issues in this regard various methods are singly or combined employed to recover and purify the ILs. The methods which are commonly and singly employed are distillation, extraction, crystallization, membrane separation, biphasic separation. The combined methods are centrifugation and distillation, Membrane separation and distillation [2].

4. CATEGORIZATION OF ILs

ILs are categorized into several classes on the basis of combined cations and anions and physical, chemical, biological properties for example ionic conductivity, miscibility in water and organic solvents, comparative acidity and basicity. Different classes of ILs are Task Specific Ionic Liquids (TS-ILs), Chiral Ionic Liquids (C-ILs), Bio-Ionic Liquids (B-ILs), Energetic Ionic Liquids (E-ILs), Protic Ionic Liquids (Pr-ILs), Basic Ionic Liquids (B-ILs), Neutral Ionic Liquids (N-ILs), Poly-Ionic Liquids (P-ILs), Supported Ionic Liquids (S-ILs), Metallic Ionic Liquids (M-ILs), Switchable Polarity Solvent Ionic Liquids (SPS-ILs) [2].

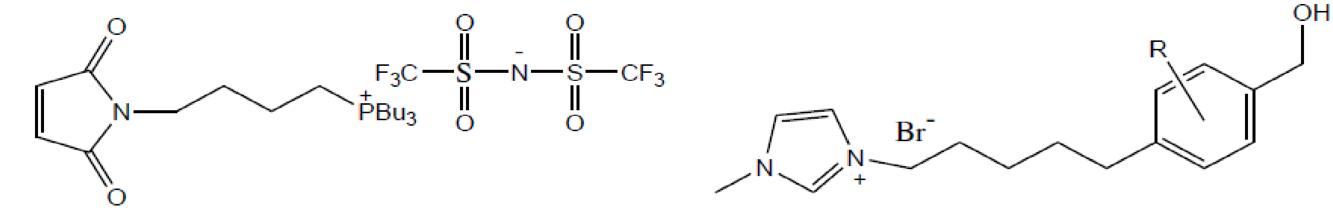
Task Specific Ionic Liquids (TS-ILs) are functionalised ILs used in organic synthesis, nanoparticle synthesis, catalytic reactions. 3-sulphopropyl tri-phenyl phosphonium *p*-toluene sulphonate is the first TS-ILs [22]. Chiral Ionic Liquids (C-ILs) are used in the field of asymmetric inductions, stereoselective polymerization, NMR chiral discrimination, liquid chiral chromatography, liquids crystal, synthesis of potential active chiral compounds etc. Example of this category is [N-(3'-oxobutyl)-N-methyl imidazolium][(+)-camphorsulfonate] C-IL [23]. Using CO_2 as activator and secondary amines as reagent to generate Switchable Polarity Solvent Ionic Liquids (SPS-ILs). A typical example of this kind is carbamate salt like $[R_2NH_2]$ $[R_2NCOO]$ [24]. Bio-Ionic Liquids (B-ILs) is prepared to overcome the problems associated with the long chains of alkyl imidazolium and alkyl benzimida-zolium based ILs. (2-hydroxyethyl)-ammonium lactate based ILs with 95% bio-degradability falls into this category [25]. Poly-Ionic Liquids (P-ILs) are extensively used in the many fields for instance electrolytes for batteries, catalysts and catalyst supports, carbon dioxide adsorbents and separation, photoresists and corrosion inhibitors, dispersants and stabilizers, pH-Triggered actuators, photo-responsive materials, polyelectrolyte membranes for fuel cells etc. By the use of N-(2-(dimethylamino) ethyl) methacrylate (DMAEMA) and a series of natural carboxylic acids (RCOOH), likely acetic acid, butyric acid, benzoic acid, oleic acid, hexanoic acid, caprylic acid, P-ILs are synthesised [26]. Due to very low vapour pressure and toxicity, higher density and thermal stability, Energetic Ionic Liquids (E-ILs) are more useful than the conventional energetic compounds like 2, 4, 6-trinitrotoluene (TNT), 2,4,6,8,10,12-hexanitro-2,4,6, 8, 10, 12-hexaazaisowurtzitane (HNIW), 1,3,5,7-tetra-nitro-1,3,5,7-tetraazocane (HMX), 4,4'-Dinitro-3,3'-diazenofuroxan (DDF), and 1,3,5-(tris-nitro) perhydro-1,3,5-triazine (RDX). N, N-dimethyl-hydrazinium hypergolic ILs is the example of this kind [2, 27].

5. PHYSICAL AND CHEMICAL PROPERTIES OF ILs

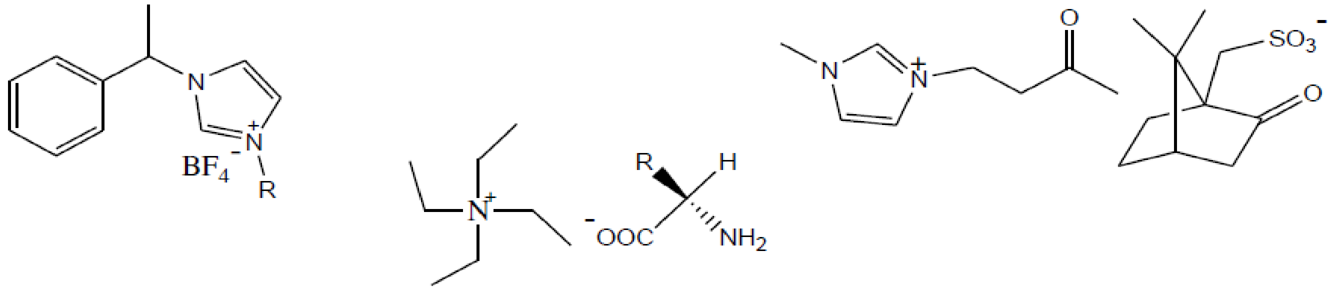
Literature survey on ILs reveals that a number physicochemical property is associated with the ILs. The properties are viscosity, surface tension, refractive index, density, polarity, thermodynamic functions, isentropic compressibility, melting point and others. The physicochemical properties of ILs can be affected by the presence of very little amount of water and organic solvent. For instance, the presence of water in the ILs can influence on the biocatalytic activity, acidity, density, viscosity, electrical conductivity, enthalpy, surface and interfacial tensions, molecular behavior of ILs. Actually the water in ILs exerts strong intermolecular interactions such as van der Waals, hydrogen bonds, electrostatic interactions. At elevated concentrations ILs are dissociated into ion pairs or individual ions. The size of cation and anion greatly influence the intermolecular and intramolecular interactions [2, 28-30].

6. VERSATILE APPLICATIONS OF ILs

The applications of ILs in the various fields of chemistry and biology increase day by day. The major reason behind these wide applications is the combination of numerous cations and anions makes the ILs thermally, physically, chemically, biologically stable. In chemistry, ILs are now extensively used as solvents, catalysts and also in the field of electrochemistry, analytical chemistry, biological chemistry, engineering chemistry, synthetic organic chemistry etc. Chemists and engineers now engage themselves to find out novel, less toxic, eco and environment friendly source for reagents, energy, fuel. IL contributes a major part in the field of green chemistry through providing green solvents and catalysts [2, 31]. Support based ILs are used for air purification and IL based adsorbents removes the indoor pollutants [10, 12]. ILs facilitate the enzymatic reactions [14]. ILs has also been applied as environmental friendly cutting fluid and sustainable corrosion inhibitors [9, 11]. Organosilicon ILs are used as Hydrogen gas storage [8]. ILs are useful and safe drug delivery agent in medicinal chemistry. ILs are playing a role as a plasticizers in polymer industry and as a solvent in nuclear chemistry for greater actinide extraction [13]. ILs are also used for the for high performance lithium metal batteries [32]. Fluorescence-enhanced IL sensors such as FIL [P66614][HDQ] is used CO_2 gas sensor and detector [33]. The application of ILs in the diverse stream of science is discussed one by one.



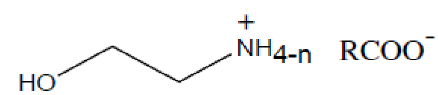
Task Specific Ionic Liquids (TS-ILs)



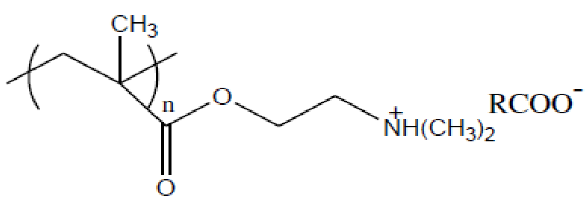
Chiral Ionic Liquids (C-IL)

$[R_2NH_2]$ $[R_2NCOO]$

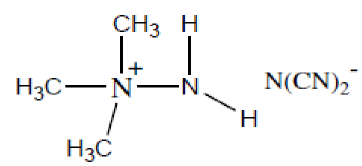
Switchable polarity solvent ionic liquids (SPS-ILs)



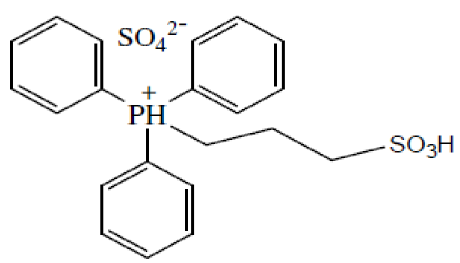
Bio-Ionic Liquids (B-ILs)



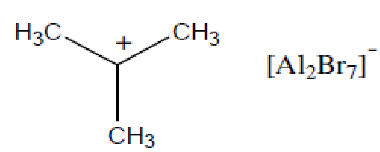
Poly-Ionic Liquids (P-ILs)



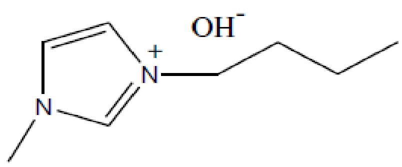
N, N-dimethyl-hydrazinium hypergolic Energetic Ionic Liquids (E-ILs)



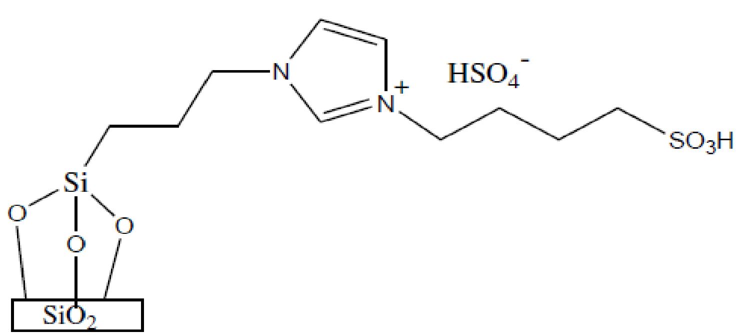
Protic Ionic Liquids (Pr-ILs)



Metallic Ionic Liquids (M-ILs)



Basic Ionic Liquids (B-ILs)



Supported Ionic Liquids (S-ILs),

Fig. 2: Different types of Ionic Liquids (ILs)

6.1. Applications Of Ils Different Fields Of Chemistry And Biology

6.1.1. Analytical chemistry

ILs have major contribution in analytical chemistry for instance in chromatography like gas chromatography (GC), GC stationary phase, high performance liquid chromatography mobile phase, matrix-assisted laser desorption ionization time of flight (MALDI-TOF), bioanalytical, capillary electrophoresis, analysis of metal ions, sensing, assorted pharmaceutical entities etc [2, 34].

6.1.2. Electrochemistry

Tuneable solubility, high viscosity and thermal stability, wide range of electrochemical potential, good conductivity of IL makes it extensively useful in the field of electrochemistry. Conductivity, viscosity, electro-chemical potential are three major controlling factors of ILs/electrodes in the application of electrochemistry. ILs are purely ionic in nature and their cations and anions are freely moving makes it excellent ionic conductor. High viscosity is attributed to different sized cations and anions their interactions. Oxidation and reduction constancies of ILs control its electrochemical potential [2].

6.1.3. Physical chemistry

Although ILs did not generate any break up topics in physical chemistry but frees ample window because of its definite physicochemical properties. In the opening, maximum research papers in Physical chemistry come from electrochemistry and this creates electrochemical window and ionic conductivity. ILs has major application as green solvent in the physical chemistry because of the extensive ability to dissolve varieties of inorganic and organic materials including polymer, co-polymers or macromolecules. Actually ILs expands the limits of conventional molecular solvents because of due to their great liquid ranges [2, 35-36].

6.1.4. Solvent

Due to the excellent physic chemical properties, ILs are now considered as the third group of solvents in comparison with conventional organic solvents. Many assorted reactions done by ILs are addition, elimination, substitution, microwave assistant, acid base reactions, transition metal catalyzed reactions, electron transfer reactions [2, 37].

6.1.5. Chemical engineering

About 2.5 decades before, the application of ILs in the Chemical Engineering field has started in the extraction procedure. It is to be mentioned that commercial plant already used ILs for their purposes. The most striking thing of IL to be used in the engineering field is due to their matchless physical and chemical properties. These properties are attributed to the different types of interactions such as van der Waals, Columbic and molecular hydrogen bonding [2, 38].

6.1.6. Biological assistance

ILs are now prevalently used an alternative reaction media and solvents instead of using organic solvents in the drug discovery technology. Actually the extra-ordinary properties of ILs can be tuned by changing the ratio of the constituent cations and anions according to the demand. ILs not only possess the physical and chemical properties but also correlates with sky-scraping biological activity. This draws the attentions of the biochemists, ecologists, pharmaceutical and medicinal scientists for the use of ILs in the biological aid such as drug delivery and drug synthesis applications. Some of the ILs show the antimicrobial activity [2, 39].

7. CONCLUDING REMARKS AND FUTURE PROSPECTS

Due to the noncorrosive, green, eco friendly nature, ILs substitute the traditional volatile organic solvents, solid acids, mineral acids and bases in the chemistry laboratory and chemical industry. In the synthetic organic chemistry, laboratory ILs can be used as solvent and catalysts for numerous organic reactions. It is to be noted that synthesis of IL involves either single step [for example Neutral Ionic Liquids (N-ILs)] or multiple steps [for example Chiral Ionic Liquids (C-ILs), Bio-Ionic Liquids (B-ILs), Task Specific Ionic Liquids (TS-ILs)]. As the change in the ratio of the constituent cations and anions of ILs tune the physical, chemical, thermal, biological properties in a flexible manner which creates the interest in the field of academics and industry. The application of ILs as a gas storage, gas separation, sustainable corrosion inhibitors, high performance Li-batteries, removal of indoor pollutants, facilitating enzymatic reactions, purification of air, sensor of CO_2, environmental friendly cutting fluids and many more is well documented.

The future prospects of ILs are used as bio-refinery to bio-chemicals to meet the demand of the clean and green energy source. In this connection it is mandatory that synthesis of/through ILs should be reproducible and purity must be maintained up to microscopic level.

Many pharmaceutical industries are using volatile organic solvents to dissolve the drugs but are able to partially soluble the drug and moreover unable to recycle it. Due to the combinations of cations (particularly organic) and anions (mixture of organic and inorganic) and wide range of polarities, ILs can easily dissolve a wide range of drugs. In this way, ILs have an excellent future prospect in the chemistry lab, chemical industry, various organic synthesis, pharmaceutical industry, drug delivery and drug discovery processes, energy generation and have a good impact on the environment.

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