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Recent Advancements in Synthesis and Applications of Amino Acid Ionic Liquids (AAILs): A Mini Review

Verma C*, **Ebenso E and Quraishi M**

[1]*Department of Chemistry, Faculty of Natural and Agricultural Sciences, School of Chemical and Physical Sciences, North-West University, Private Bag 046, Mmabatho 2735, South Africa*

[2]*Material Science Innovation and Modelling (MaSIM) Research Focus Area, Faculty of Natural and Agricultural Sciences, North-West University, Private Bag 046, Mmabatho 2735, South Africa*

[3]*Center of Research Excellence in Corrosion, Research Institute, KFUPM, Dhahran, Kingdom of Saudi Arabia*

***Corresponding author:** Chandrabhan Vermaa, Department of Chemistry, Faculty of Natural and Agricultural Sciences, School of Chemical and Physical Sciences, North-West University, Private Bag 046, Mmabatho 2735, South Africa, Tel: 27618714192; E-mail: chandraverma.rs.apc@itbhu.ac.in

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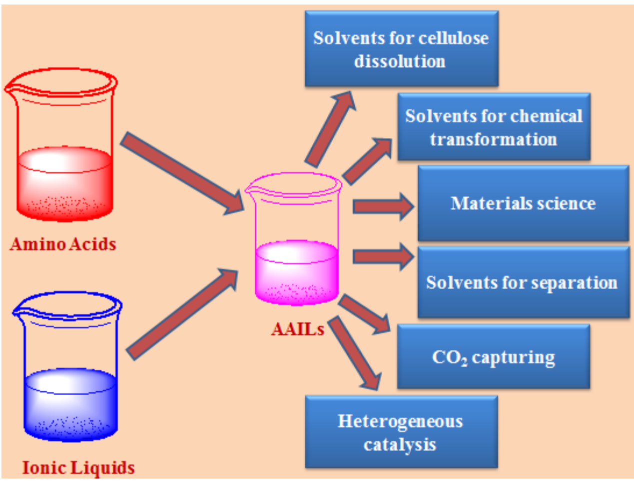
Abstract

Recently, amino acid ionic liquids (AAILs) have attracted increasing attention in the field of catalysis, solvents for separation science, cellulose dissolution and chemical transformations because of their non-toxic, biodegradable, biocompatible and low production cost. Several AAILs based on imidazolium, phosphonium, ammonium and cholinium ionic liquids have been widely employed in materials science, modern chemistry and bioscience. Some recent application of the AAILs includes heterogeneous catalysis, CO_2 capture and solvents for chemical transformation, cellulose dissolution, extraction and separation processes. Several experimental and computational techniques are being employed to gain insight about the proper understanding of their physiochemical properties. The AAILs showed high thermal stability as compared to the pure ionic liquids which is attributed to the strong Van der Waals forces and hydrogen bonding (noncovalent bonds) which make them as solvents for high temperature reactions. Generally, AAILs show decrease in viscosity, refractory index and density and increase in the conductivity on elevating the temperature. Most of the AAILs are liquids at room temperature and their glass transition temperature (Tg) decreases with increasing the size of the side chain length. Because of the increasing number of the literatures dealing with the synthesis and applications of AAILs, a review article describing the basic information and recent applications of the AAILs is highly considerable. In view of this, in presence review article we describe the collection of some major reports available on the synthesis of AAILs with little highlights on their applications.



Keywords: Amino acid ionic liquids; High temperature solvents; Green solvents; Glass transition temperature; CO_2 capture; Thermophysical properties

Introduction

Ionic liquids: Brief introduction

Ionic liquids are defined as the compounds made up of ions having melting point below 100°C [1-3]. Recently, because of the increasing ecological awareness and enormous anxieties of "green chemistry", use of ionic liquids has received wide attention in multidisciplinary studies including chemical engineering, materials science, environmental science and modern chemistry during the last few decades [4-6]. As compared to the traditional volatile compounds, the ILs are non-flammable, non-volatile and stable in water and air which make them as alternative chemicals for versatile applications [7,8]. They (ILs) are also regarded as designer chemicals because a numerous combination of anions and cations leads to the synthesis of task specific ionic liquids. A suitable combination of available anions and cations can results in to formulation of more than 1012 types of ionic liquids. After the establishment of first ionic liquid namely, ethyl ammonium nitrate (EAN) by Paul Walden several ILs have been developed by suitably varying the combination of cations and ions [7,9-11]. The ionic liquids can be broadly categorized into four major categories namely, polyionic liquids (PILs), task specific ionic liquids (TSILs), room temperature ionic liquids (RTILs) and membrane supported ionic liquids (MSILs) [12-15]. The MSILs comprises the compounds of ionic liquids sustained on metal-organic frameworks (MOFs). The ionic liquids are also called as ionic melts, fused salts, ionic fluids, liquid electrolytes, ionic glasses, liquids salts and solvents of the future and have numerous potential applications such as biological and industrial catalysts, electrochemical and biosensors, polymers and catalytic chemistry, lubricants, optical thermometers, analytic devices, solvents for chemical transformations and separation processes etc. [3,16-18]. The ILs is thermally stable therefore they can be distilled out without decomposition at reduced pressure and high temperature which makes them a suitable rout for their recyclability [19,20]. Although, ILs are safe to handle because of their low volatility (low vapour) and inflammable behavior however similar to the organic compounds their release in the surrounding environments can causes soil (land) and water pollution. Recent advancements in the field of ILs showed that they possess extensive assortment of toxicity therefore now use of the term “green chemicals” for ionic liquids is highly questioned [21,22]. More so, as compared to the traditional organic compounds synthesis, the production of ionic liquids are expensive (non-costeffective) and requires enormous amount of organic solvents for anion exchange that unfavorably affect the environment and synthetic efficacy [23]. In view of this recent development (synthesis) of ILs mainly focused on the room temperature ionic liquids (RTILs) those are generally associated with high viscosity, low volatility, high thermal and chemical stability and high conductivity using natural feed stocks such as amino acids, carbohydrates and carboxylate salts [24-28].

Amino acids (AAs): Brief introduction

Amino acids (AAs) are one of the most abundant biomaterials in the nature and are well established at the position of non-toxic, biocompatible and biodegradable chemicals [29-31]. Recently they have been extensively used as feedstocks for the production several classes of ionic liquids because of their environmental friendly behavior [24-27,32]. They are the building blocks (monomers) of the several polypeptides and proteins that contain a central carbon atom C, an acidic carboxylic (-COOH) group, a basic amino (-N) group and specific -R group (side chain). The side chain is responsible for the determination of basic characteristics of each amino acid such as size, pH and polarity [33-35]. It is important to mention that there are twenty types of amino acids that can combined together through peptide bond and form polypeptide chains or proteins responsible for several biological applications. Nowadays, AAs are widely produced and consumed in many industrial fields. The AAs can be produced or synthesized using several methods such as extraction from the natural resources, enzymatic conversion, fermentation and chemical transformations [36-40]. Except few of the AAs such as glycine and methionine, most of the AAs are being produced by fermentation and enzymatic conversion methods. The AAs are widely employed as active pharmaceutical ingredients, medical nutrition, dietary supplements, culture media for microorganism, cosmetics and food industries [40,41]. It is estimated that the global annual demands of AAs is more than 6.13 million tons and its annual market growth rate is more than 10% [42]. Because of their natural and biological origin the AAs and their derivatives are considered to be environmental benign materials to be used for various industrial and biological applications.

Amino acid ionic liquids (AAILs): Literature review

Amino acids are comprise of both acidic carboxylic (-COOH) and basic amino (-N) groups along with asymmetric (chiral) center and different alkyl (side) chains therefore their association with different ILs results in to the synthesis and designing of noble functional materials. The AAILs are characterized by their liquidus nature at or far below the ambient temperature and are being utilized as green (non-volatile) solvents for numerous chemical transformation at the place traditional volatile solvents. Recently numerous reports are published describing the synthesis and applications of the AAILs in science and technology. Literature study suggests that the number of publications dealing with the synthesis and applications of AAILs are increasing day-by-day therefore the collection of the earlier works published on the topic recent advancement on the topic “synthesis and application AAILs” is highly recommended. Therefore, in this review article we described the collection of some major publications available on the synthesis of AAILs with particular emphasis on their applications. Owing to the huge growing interest towards AAILs, recently several theoretical studies have been reported to describe the proper understanding of their physiochemical properties [43-46]. Some recent applications of the AAILs include chiral solvent for chemical transformations, chiral solvent for stabilization and dissolution of carbohydrate polymers including cellulose, nucleic acids, carbohydrates and other species of primary biological importance [47-50]. Recently, several AAILs have also been employed as electrolytes. The suitability of the AAs for the formulation of AAILs is based on the fact that AAs contain both cationic and anionic moieties through which they can interact or combine with ILs to form different types of AAILs [51-54]. More so, unlike to most of the ILs, the AAILs are essentially cost-effective and can be synthesized in large extents using biological raw stuff [55]. The area of AAILs synthesis and their applications is still remains rather new. Literature survey reveals that there is no good review available on the topic.

The synthesis and applications of the AAILs gained tremendous attention after the work reported by Fukumoto [56]. These authors reported the synthesis of different AAILs by the combination of twenty natural AALs with 1-ethyl-3-methylimidazolium [Emim]. They observed that the AAILs were miscible in common organic solvents and could dissolve native AAs. Results of the investigation showed that the properties of AAILs such as ionic conductivity, miscibility in the organic solvents and glass transition temperature mainly depend upon the nature of the side chains of AAs. Synthesis and measurements of the physiochemical properties of twenty types AAILs was outlined by Ohno and Fukumoto et al. [57]. They observed that at room temperature all the AAILs were in liquid form. The investigated AAILs showed improved decomposition temperature and reduced viscosity. The free amino group was acetylated and the AAILs retained their chirality upon their heating up to 150°C. Miscibility of the water in the ionic liquids was showed interesting result. Though, the AAILs form separate phase at room temperature but their cooling showed some miscibility of water in them. The process of miscibility and non-miscibility was reversible to cooling or heating up to some degree. The task specific coupling of imidazolium based cation with modified polystyrene with L-proline results into the formation AAILs for scavenging and catalytic abilities [54]. Gouveia et al. reported the synthesis of five AAILs by the reaction of 1-ethyl-3-methylimidazolium cation ([mim]+) and AAs based anions such as L-alaninate ([L-Ala]$^-$), glycinate ([Gly]$^-$), L-prolinate ([L-Pro]$^-$), taurinate ([Tau]$^-$) and L-serinate ([L-Ser]$^-$) [58]. Viscosity, refractive index and density of the pure ionic liquids and the AAILs were measured for sake of comparison. Results derived in the analysis showed good agreement to the results reported in literature. Significant decreases in the viscosity of pure ionic liquids were observed after their conversion in AAILs. The combination of the imidazolium based cations with anions of amino acids resulted significant improvement in the physiochemical properties of the ILs. Four other imidazolium based AAILs were synthesized and characterized by the reaction of 1-ethyl-3-methylimidazolium hydrogen sulfate with glycine [Emim][Gly], alanine [Emim][Ala], serine [Emim][Ser] and proline [Emim][Gly] [59]. Their viscosities were decreased by nearly one order of magnitude on raising temperature from 193.14 K to 353.15 K. This finding suggested that AAILs are can be used for potential applications at relatively high temperature.

A new category of task specific Tetrabutylphosphonium amino acid [$P(C_4)_4$][AA] were developed by the reaction of several AAs namely, glycine, L-β-alanine, L-alanine, L-lysine and L-serine and Tetrabutylphosphonium hydroxide [$P(C_4)_4$][OH] [60]. Synthesized AAILs were characterized using elemental analysis, spectral and thermochemical and thermophysical properties measurements. The synthesized [$P(C_4)_4$][AAs] supported on porous silica gel was used for the adsorption of CO_2. In another study, tetrabutylphosphonium cation was coupled with twenty different amino acids to gives twenty types of AAILs [61]. Out of twenty AAIL, fifteen exist in the liquid form at room temperature and it was observed that the investigated tetraalkylphosphonium based AAILs show relatively lower viscosity and higher decomposition temperature as compared to the previously reported ammonium based AAILs. Two other phosphonium cation based AAILs namely, trihexyl (tetradecyl) phosphonium methioninate [P_{6614}][Met] and prolinate [P_{6614}][Pro] were synthesized as evaluated as adsorbates for CO_2 adsorption [62]. It was observed that both [P_{6614}][Met] and [P_{6614}][Pro] react with CO_2 in a ratio of one amino group per CO_2 molecule (1:1 stoichiometry). Ouyang et al. demonstrated the synthesis of tetrabutylphosphonium based AAILs [P_{444}][AAs] by the reaction of tetrabutylphosphonium cation with L-serine [P_{444}][Ser], L-proline [P_{444}][Pro], L-valine [P_{444}][Van], L-alanine [P_{444}][Ala] and glycine [P_{444}][Gly] [63]. The synthesized [P_{444}][AAs] were characterized using NMR, FT-IR and thermal analysis methods. The investigated AAILs were used as catalyst for Knoevenagel condensation reaction of benzaldehyde with different active methylene compounds and best performance was obtained in the case of [P_{444}][Pro] which showed yield around 93%. The catalyst [P_{444}][Pro] was recyclable up to six time without any significant decrease in its activity and quantity. Various AAILs, based tetramethylammonium [TMA] and tetrabutylphosphonium [TBP] cations and AAs were synthesized and used for CO_2 capture [64]. Results showed that AAs showed better CO_2 capture performance as compared to the [TMA][AAs] whereas [TBP][AAs] showed the best CO_2 capture performance.

Recently, cholinium based AAILs are gaining significant attention because of their low toxicity and high biological degradability. Hou et al. described the synthesis of eighteen cholinium based AAILs by the reaction of cholinium cation and eighteen different natural AAs [65]. They obtained that cholinium AAILs have low toxicity and are relatively high degree of biodegradability. In another study, a series of cholinium based AAILs were synthesized by the neutralization reaction choline hydroxide ([Ch][OH]) solution with five amino acids namely glycine ([Ch][Gly]), L-alanine ([Ch][Ala]), L-β-alanine ([Ch][β-Ala]), L-proline ([Ch][Pro]) and L-serine ([Ch][Ser]) [66]. The synthesized AAILs were characterized using DSC, NMR and FT-IR methods. The thermophysical properties of AAILs were measured and it was observed that increase in caused substantial decrease in viscosity, refractive index and density and increase in the magnitude of conductivity. TGA study showed that [Ch][AAs] showed excellent thermal stabilities. Similar observation was by Santis et al. [67] where they synthesized eighteen different cholinium based AAILs, [Ch][AAs] by the neutralization of choline hydroxide with different natural AAs. Out of eighteen AAILs, fourteen AAILs were liquid at room temperature. Only AAIL derived from valine was not liquid at room temperature however its melting point was below 80°C. In another study eight type of cholinium based ionic liquids was synthesized by the direct reaction of choline chloride with eight different AAs namely, alanine [Ch][Ala], aspartic acid [Ch][Asp], serine [Ch][Ser], valine [Ch][Val], cysteine [Ch][Cys], leucine [Ch][Leu], proline [Ch][Pro] and phenylalanine [Ch][Phe] [68]. The synthesized AAILs were used to extract pectin and flavonoids from ponkan peels. Synthesis of ten cholinium based AAILs by the neutralization reaction of choline hydroxide and ten different natural amino acids namely L-Alanine ([Ch][Ala]), L-Arginine ([Ch][Arg]), L-Asparagine ([Ch][Aspr]), L-Glutamine ([Ch][Glu]), L-Histidine ([Ch][His]), L-Methionine ([Ch][Met]), L-Phenylalanine ([Ch][Phen]), L-Serine ([Ch][Ser]), L-Tryptophan ([Ch][Try]) and L-Tyrosine ([Ch][Tyr]) was reported [69]. The investigated AAILs were characterized employing NMR spectroscopy. Their biocompatibility was tested on few strains of gram-positive and gram-negative bacteria and their biodegradability was analysed by industrial sewage water. Their E_0 values range from 160-1120 mg/L suggesting that all AAILs are practically non-hazardous. Over 60% biodegradation of the AAILs was observed in 28 days. It also reported that cholinium based AAILs promote the growth of certain strains of microorganisms particularly at lower concentrations [70]. Four ammonium based AAILs were synthesised by the reaction of 4-vinylbenzyltrimethylammonium cation [VBTMA] and glycine [VBTMA][Gly], L-alanine [VBTMA][Ala], L-serine [VBTMA][Ser] and L-proline [VBTMA][Pro] as anions [71]. The synthesized AAILs were characterized using NMR and FR-IR methods. TGA analysis showed that [VBTMA][Pro] showed the highest thermal stability of 178.29°C and the thermal stabilities followed the order: [VBTMA][Gly]<L-alanine [VBTMA][Ala]<L-serine [VBTMA][Ser] < L-proline [VBTMA][Pro].

Conclusions and Outlooks

After the establishment of AAILs through the reports published by Fukumoto et al. several AAILs have been synthesized as used for verity of applications including catalysis and solvents for chemical transformation, cellulose dissolution and separation sciences [56]. The AAILs possess high thermal stability as compared to the pure ILs and therefore can be employed as high temperature solvents. Their thermal stability and glass transition temperature decreases on increasing the side chain length of the AAs. Because AAILs offer strong intermolecular van der Waal force of attraction along with very strong intermolecular hydrogen bonding, they can easily interact with cellulose and other biological macromolecules and make them solubilize. Therefore, application of AAILs for biological macromolecules such as proteins and carbohydrates polymers is highly recommended. Though AAILs have been established as potential candidates for biological, pharmaceutical, medical and chemical applications, however the reports on their applications are relatively scare. Therefore, for future studies in all area of science and technology the use of AAILs is appreciably recommended. Among the imidazolium, ammonium, phosphonium and cholinium based AAILs, the cholinium based AAILs, [Ch][AAs] are considered to be most environmental friendly, biodegradable, biocompatible and non-toxic therefore their synthesis and consumption should be enhanced. Since, increased molecular weight (side chain length) causes reduction in thermal stability and increase in the toxicity of the AAILs therefore smaller side chain with lower molecular weight is recommended for future studies.

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