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Ionic liquid as antioxidant in recent advancement of biodiesel or drug delivery

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Abstract

Advancements in the realm of ionic liquids (ILs) have expanded their applications beyond traditional uses, extending into various pharmaceutical and biomedical fields. Scientists are increasingly drawn to the versatility of ILs, often referred to as Solutions for Your Success, owing to their myriad applications in the pharmaceutical sector, addressing diverse diseases and facilitating innovative drug delivery methods. A pivotal physicochemical characteristic influencing a drug's efficacy at the target site is its solubility. The significant hurdle of poor solubility often leads to the failure of promising drug candidates at various stages of drug discovery. ILs emerge as promising drug delivery vehicles, particularly for drugs with low solubility.

In the context of drug delivery, ILs can be tailored to form salts that meet the theoretical requirements for efficient drug delivery. This customization involves combining different anions and cations to create ILs suitable for specific drugs and applications. The modularity of the physicochemical properties of ILs allows for customization based on the unique demands of different applications.

This study aims to illuminate the utilization of ILs as antioxidants, exploring their recent advancements in drug delivery and biodiesel applications. By investigating their role in drug delivery, the research contributes to a deeper understanding of how ILs can address challenges associated with poor drug solubility and potentially revolutionize pharmaceutical advancements.

Keywords: Ionic liquids, biodiesel, drug delivery, solubility, antioxidants

Introduction

Ionic liquids (ILs) have emerged as versatile and innovative chemicals that offer significant advantages in various fields, including biodiesel production and drug delivery. Their unique properties, such as low vapor pressure, high thermal stability, and the ability to dissolve a wide range of compounds, make them highly suitable for enhancing the performance and efficiency of processes in these areas. This introduction will briefly explore the recent advancements in the use of ionic liquids as antioxidants in biodiesel production and drug delivery, highlighting their benefits and the challenges that lie ahead.

In Biodiesel Production

Biodiesel, a renewable and environmentally friendly fuel alternative to fossil fuels, has gained substantial interest in recent years. However, one of the main challenges in biodiesel production and storage is the oxidative degradation of unsaturated fatty acids, which can lead to the formation of sediments and a decrease in fuel quality. Recent advancements have shown that ionic liquids can be employed as effective antioxidants in biodiesel. These substances can stabilize the biodiesel by scavenging free radicals and inhibiting oxidative processes, thus improving its storage stability and combustion quality.

The use of ILs as antioxidants in biodiesel not only enhances its oxidative stability but also contributes to the sustainability of the production process. Some ionic liquids are biodegradable and can be synthesized from renewable sources, aligning with the eco-friendly nature of biodiesel itself. Moreover, their effectiveness at low concentrations makes them economically viable as well.

In Drug Delivery

In the realm of drug delivery, ionic liquids have been explored for their potential to improve the solubility, stability, and bioavailability of drugs. The tunable solubility of ILs allows for the dissolution of poorly soluble pharmaceutical compounds, enhancing their effectiveness. Furthermore, ILs can be engineered to possess antioxidant properties, which is particularly beneficial in the delivery of drugs susceptible to oxidation, a common cause of degradation and loss of efficacy.

Recent advancements in this area include the development of IL-based drug delivery systems that can target specific sites within the body, reducing side effects and improving therapeutic outcomes. The antioxidant properties of certain ILs also protect sensitive drugs from oxidative damage during storage and transport, extending their shelf life.

Challenges and Future Perspectives

Despite their promising applications, the use of ionic liquids in biodiesel production and drug delivery faces several challenges. The toxicity and biodegradability of some ILs remain concerns that need to be addressed through the design of greener, more environmentally friendly variants. Additionally, the cost of synthesizing and purifying specific ionic liquids can be high, potentially limiting their widespread adoption.

The future research in this field is likely to focus on developing novel ionic liquids with tailored properties, such as enhanced antioxidant capabilities, lower toxicity, and higher biodegradability. Advances in synthesis methods may also reduce costs and improve the feasibility of using ILs in these applications.

In conclusion, the recent advancements in the use of ionic liquids as antioxidants in biodiesel production and drug delivery highlight their potential to significantly impact these fields. By overcoming the current challenges, ILs could play a crucial role in developing more sustainable and efficient biodiesel fuels and more effective drug delivery systems.

Related study

A literature review involves an exploration of existing materials pertaining to the research topic. This process aids researchers in gaining a comprehensive understanding of the specific field they are investigating. Additionally, it serves as a valuable tool for exchanging information, aiming to prevent redundancy among researchers and allowing them to ascertain what is already established in similar studies. Familiarity with previous research literature is crucial for researchers as it enables them to align their findings with those of prior studies, contributing to a more informed and validated research endeavor.

Ionic Liquid as an Antioxidant in Recent Advancements of Drug Delivery

Moshikur *et al.* (2021) [14] conducted early research aimed at developing ionic liquids as environmentally friendly, nonvolatile, stable, and noncombustible solvents. Ionic liquids (ILs), "a distinct class of materials composed of ions, possess unique characteristics, including low vapor pressure, strong solvating power, and high thermal stability. During the initial phases of drug discovery, these attributes were found to be highly advantageous. ILs were employed as novel solvents for chemical synthesis, contributing to the development of eco-friendly alternatives. Beyond their use as solvents in various activities, such as organic synthesis, ILs demonstrated versatility by enhancing the solubility of compounds in aqueous media. This property facilitated the purification and isolation of products, particularly during the initial stages of drug discovery.

Shi *et al.* (2013) [21] defined ILs as salts exhibiting infinitely variable qualities, including volatility, toxicity, instability, and flammability, with lower melting points around 100 °C. The application of ILs in drug development has evolved beyond drug delivery systems, incorporating the formation of micelle structures, nanoparticles, and solid dispersions. These IL-based drug delivery systems aim to enhance drug solubility and bioavailability.

In the earlier perception of ILs, they were primarily seen as salts of imidazolium, pyrrolidinium, quaternary ammonium, phosphonium cations, or pyridinium, mixed with various anions to create salts meeting IL requirements (Agatemor *et al.*, 2018) [2]. Recent research has expanded the scope of ILs to include other cations such as guanidinium, cholinium, and metal-based cations, thereby increasing the chemical space for creating ILs with unique properties.

Adawiyah *et al.* (2016) [1] note that application-driven studies of ILs, especially in drug delivery, are thriving and expanding. These advancements offer new horizons in therapeutics, showcasing the potential for ILs with distinct properties to be utilized in theoretical studies and practical applications. The heterogeneity of the structure of an asymmetric sterical cation, influencing interactions with anions and the arrangement within a crystal lattice, is a key factor supporting the characteristic low melting point of IL compounds.

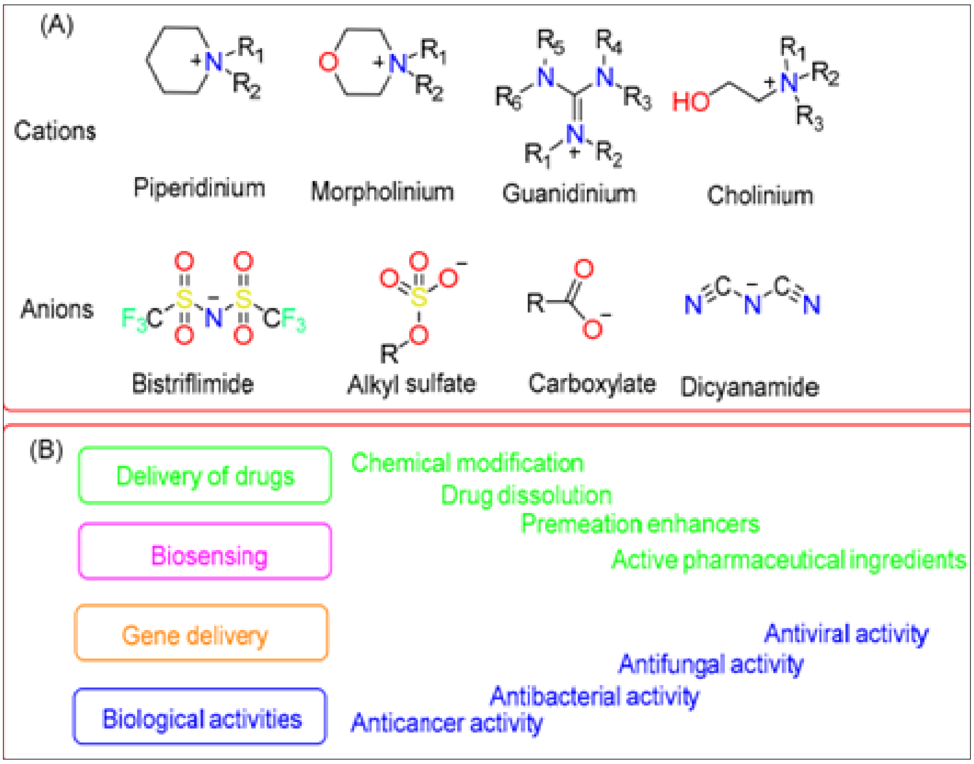

Fig 1: A list of different cationic and anionic molecules used in pharmaceuticals (A) and their various applications in different biomedical fields

Hansen *et al.* (2021) [8] advocated for substituting organic unsafe solvents with more environmentally friendly alternatives that exhibit reduced volatility and flammability. They pointed out that ILs can be particularly advantageous in this context due to their minimal vapor pressure. Furthermore, Grodowska *et al.* (2010) [6] highlighted the significance of carefully selecting cations and anions in ILs. They observed that such selections could lead to unique interactions with specific solute groups, a factor crucial for enhancing the solubility of compounds with inherent complexity, including active pharmaceutical ingredients and their precursors.

Table 1: Application of ILs as a useful solvent for Ionic liquid as antioxidant in recent advancement of biodiesel

Cations/Anions	Applications	References
Cations such as (C_4MIM) in combination with anions (NTf_2), (BF_4), or (PF_6)	Synthesis of APIs	Hayes *et al.* (2015)
1-butyl-3-methylimidazolium hexafluorophosphate ((C_4MIM) (PF_6))	Synthesize hybrids of pyrimidine nucleoside; thiazolini-4-one	Zhang *et al.* (2009)
(Emim)Cl	Synthesis of 5-hydroxymethylfurfural (HMF) from monosaccharides	Zunita *et al.* (2022)
(C_4MIM) (X) (where X = BF_4 or PF_6)	Preparation of L-4-boronophenylalanine (L-BPA)	Wolan *et al.* (2003)
Pinacol borane protected p-iodophenylalanine with (C_4MIM) (BF_4)	Synthesis of L-BPA	Kurata *et al.* (2010)
Imidazolium-based ILs in combination with Friedel-Crafts reaction and nucleophilic displacement process	Synthesis of Pravadoline	Earle *et al.* (2000)
Ru-BINAP catalyst and (C_4MIM) (BF_4)	Synthesis of (S)-naproxen	Monteiro *et al.* (1997)
(C_4MIM) (PF_6) and (C_4MIM) (BF_4)	Synthesis of (R, S)-ibuprofen	Monteiro *et al.* (1997)

Ionic liquid as antioxidant in recent advancement of biodiesel

Dong *et al*. (2019) [4, 32] evaluated biodiesel as a potential replacement for fossil diesel due to its renewable and environmentally friendly attributes. The overuse of fossil resources has led to numerous energy shortages and environmental pollution issues (Dong *et al*., 2019) [4, 32]. The aim is to provide sustainable and green substitutes for fossil fuels (Mao *et al*., 2022) [3, 12, 28, 46]. Biodiesel, derived from methyl esters of long-chain fatty acids, is a popular substitute for fossil diesel, boasting advantages such as renewability and environmental friendliness.

The production of drugs/molecules

Transesterification of triglycerides in oil and esterification of long-chain fatty acids are recognized as the primary processes for biodiesel production from renewable oil sources like vegetable and animal fats (Pan *et al*., 2020) [18]. The use of a catalyst is essential in these processes, impacting the cost, efficiency of biodiesel production, and reaction conditions (Mukhtar *et al*., 2022) [15, 48].

ILs, described as organic salts with melting temperatures below 100°C composed of anions and cations, have garnered attention as catalysts in the conversion of renewable oil to biodiesel. ILs offer exceptional qualities, including robust solubility, a wide liquid temperature range, low toxicity, thermal and chemical stability, and a lack of vapor pressure (Cheng *et al*., 2022 [3, 28]; Panchal *et al*., 2022) [19].

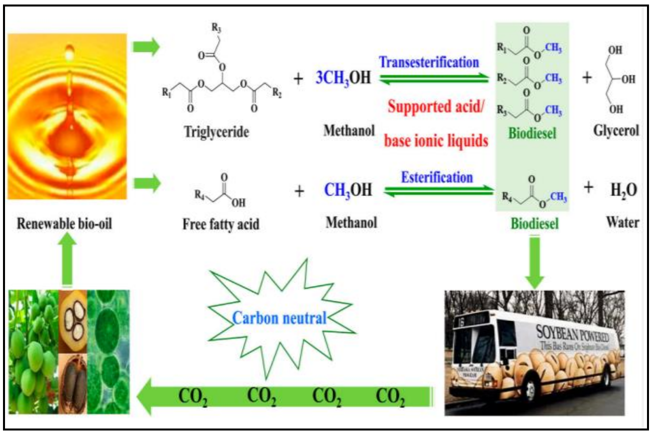

Fig 2: Biodiesel production cycle from renewable bio-oils via transesterification and esterification catalyzed by the supported acid/base ionic liquids.

Meanwhile, by altering the structure of the anions and cations in ILs, the physicochemical properties and functions of ILs can be created and adjusted. Because of these exceptional benefits, ILs have been used in the production of biodiesel. For example, Han *et al.* (2022) reported that SO_3H-functionalized ILs showed exceptional catalytic activity in the esterification of fatty acids to biodiesel, while Panchal *et al.* (2022) [19] reported that basic ILs demonstrated notable catalytic performance in the transesterification of triglycerides to biodiesel. However, because ILs are often soluble in polar solvents, recycling them throughout the biodiesel production process is challenging. Another serious flaw in ILs is their high viscosity, which makes them difficult to use. To increase the use of ILs in the industrial production of biodiesel, these shortcomings must be fixed.

The immobilization of ILs on solid supports to create IL-functionalized materials is a workable solution to the aforementioned problems. Ionic liquid-functionalized materials combine the benefits of heterogeneous and homogeneous catalysts, inheriting the former's heterogeneous properties from the solid support and the latter's homogeneous characteristics from highly soluble ILs on the support surface. Furthermore, materials functionalized with IL can be used in fixed-bed reactors to produce biodiesel continuously. Consequently, a variety of IL-functionalized materials have been created for the immobilization of ILs on supports in the catalytic synthesis of biodiesel production. These supports include polymers, silica, magnetic nanomaterials, nitrogen-doped carbon, and metal–organic frameworks (MOFs) (Pan *et al*., 2016). A recent review concentrating on the heterogenization of ILs via the immobilization of ILs on solid supports for biodiesel production is still needed, despite the fact that numerous excellent studies on ILs as catalysts or solvents for the synthesis of biodiesel have been published (Ong *et al*., 2021) [16].

Wang *et al.* (2019) synthesized an acidic poly (ionic liquid) via self-polymerization of an acidic IL monomer with a double bond group, achieving a 91.6% yield in the esterification of palmitic acid to biodiesel at 65°C for 8 hours. However, these polymers typically exhibit low specific surface area and hydrophobicity. To enhance these properties, Liang *et al.* (2016) [40] investigated copolymerization of the acidic IL monomer with divinyl benzene (DVB). Increasing DVB content improved hydrophobicity and specific surface area but reduced acid density, allowing for adjustment of physicochemical properties. Various ionic liquid polymers were developed through copolymerization for biodiesel production. Feng *et al*. (2017) [33] synthesized poly (ionic liquid) by copolymerization of a sulfonic acid IL monomer and DVB, achieving a biodiesel yield of 95.2% from soapberry oil.

Condensation was also utilized for synthesis, as demonstrated by Bian *et al*. (2019) [26], who achieved high acidity (4.5 mmol/g) in polyionic liquid through phenolic condensation. Post-modification, as shown by Pan *et al*. (2019), yielded a mesoporous melamine-formaldehyde polymer with high acidity (2.2 mmol/g) and a biodiesel yield of 95% from oleic acid. Zirconium phosphonate and 2D-layered montmorillonite were also employed to support acidic poly ILs for catalyst synthesis (Liu *et al*., 2019; Pan *et al*., 2022).

Basic poly (ionic liquid) was synthesized through copolymerization and subsequent ion exchange, exhibiting superhydrophobicity and a porous structure with a biodiesel yield of 96.3% from soybean oil via transesterification (Jiang *et al*., 2017) [37]. Ionic liquid-functionalized porous polymers show potential as catalysts for oil-to-biodiesel conversion, despite concerns regarding cost and thermal stability.

Carbonaceous materials are highly regarded as promising supports for synthesizing efficient and reusable catalysts due to their exceptional thermal and chemical stability, controllable surface wettability, cost-effectiveness, availability, and non-toxic nature (Dhawane *et al*., 2018) [30]. N-rich porous carbon, in particular, stands out for its abundance of active N sites, which facilitate IL support via chemical post-modification (Sun *et al*., 2019). Additionally, N-rich porous carbon boasts a high specific surface area, enhancing the interaction between carbon-nitrogen material's active sites and substrates, thus facilitating high IL loading. Porous carbon nitrogen materials are typically derived from nitrogen-containing organic compounds (e.g., cyanamide, melamine, urea) or polymers (e.g., polypyrrole) serving as nitrogen and carbon sources. The incorporation of fructose as a carbon source alongside nitrogenous organic compounds allows for the adjustment of carbon content in carbon-nitrogen materials. Carbonization and the solvothermal method are commonly employed to convert organic compounds or polymers into carbon-nitrogen materials (Tang *et al*., 2018). The template method proves effective in enhancing the specific surface area of carbon-nitrogen materials by structuring pore networks, with common templates including potassium hydroxide (KOH), zinc chloride ($ZnCl_2$), and silicon dioxide (SiO_2). These templates are subsequently removed through washing or corrosion, depending on the type, after carbonization.

Ionic liquids' functional carbons are synthesized through quaternary ammonization of carbon-nitrogen materials using various reagents like iodomethane, 1,3-propane sultone, and 1,4-butanesultone, followed by acid treatment with substances like $H_3PW_{12}O_{40}$, HSO_3CF_3, and H_2SO_4 (Liu *et al*., 2016) [43]. For example, acid IL functional carbon is produced by quaternary ammonization of nanoporous carbon with 1,3-propanesultone, followed by ion exchange with HSO_3CF_3. The resulting catalyst exhibits an 88.5% biodiesel yield via transesterification of tripalmitin with methanol (Liu *et al*., 2015) [42]. To minimize catalyst costs, waste cow manure is utilized for synthesizing N-rich nanoporous carbon, followed by treatment with 1,4-butanesultone and HSO_3CF_3 to produce acid IL-functionalized carbon. This acid catalyst demonstrates an 88.5% biodiesel yield, surpassing even homogeneous $H_3PW_{12}O_{40}$ (Noshadi *et al*., 2016) [49].

Research methodology

Research Methodology for Review Paper: ‘Ionic Liquid as Antioxidant in Recent Advancements of Biodiesel or Drug Delivery'

1. Objective Clarification

- Define the specific objectives of the review paper, such as summarizing recent advancements in the utilization of ILs as antioxidants in biodiesel production or drug delivery systems.

2. Literature Search

- Conduct an extensive literature review using academic databases, journals, books, and reputable sources to identify relevant studies, articles, and publications related to the application of ILs in biodiesel or drug delivery.

3. Inclusion and Exclusion Criteria

- Establish criteria for including or excluding sources based on relevance, publication date, and quality. Ensure a comprehensive coverage of recent advancements and key developments in the chosen field.

4. Data Extraction

- Extract pertinent information from selected sources, focusing on the role of ILs as antioxidants in biodiesel or drug delivery. Categorize findings based on application, methodology, and outcomes.

5. Synthesis of Information

- Summarize and synthesize the extracted data, highlighting trends, challenges, and advancements in the field. Identify common themes and divergent findings.

6. Comparative Analysis

- Conduct a comparative analysis of studies, examining variations in methodologies, experimental setups, and outcomes. Discuss the implications of these differences on the overall understanding of ionic liquid applications in biodiesel or drug delivery.

7. Critical Evaluation

- Critically evaluate the strengths and limitations of the studies reviewed. Identify gaps in the existing literature and propose potential areas for future research.

8. Thematic Organization

- Organize the review paper thematically, grouping information based on different aspects such as the type of ionic liquid used, its concentration, the application in biodiesel or drug delivery, and the observed effects.

9. Flow and Coherence

- Ensure a logical flow and coherence in presenting the reviewed material, guiding the reader through the evolution of research in the field and the current state of knowledge.

Research questions

1. In the context of Biodiesel Production

- How does the incorporation of ILs impact the oxidative stability and shelf life of biodiesel?
- What are the key mechanisms through which ionic liquids act as antioxidants in biodiesel systems, and how do these mechanisms contribute to the enhancement of oxidative stability?

2. In the context of Drug Delivery

- How does the utilization of ILs influence the efficiency of drug delivery systems in terms of controlled release and bioavailability?
- What role does ILs play in mitigating challenges associated with drug degradation and instability during the drug delivery process?

3. Comparative Analysis

- What similarities and differences exist in the mechanisms of ionic liquid antioxidants when applied to biodiesel and drug delivery systems?
- How do the specific requirements and constraints of biodiesel and drug delivery applications influence the choice and effectiveness of Ils as antioxidants?

These research questions aim to delve into the multifaceted roles of ILS, both in the context of biodiesel production and drug delivery systems, elucidating their mechanisms and applications as antioxidants.

Research GAP

The research gap in the use of ILs as antioxidants in recent advancements of biodiesel or drug delivery lies in the limited exploration and understanding of their specific mechanisms and applications within these contexts. While ILs have demonstrated promising antioxidant properties in various fields, their potential in biodiesel and drug delivery systems remains relatively unexplored.

1. Biodiesel Applications

- Few studies have systematically investigated the effectiveness of ILs as antioxidants in biodiesel production. Understanding their impact on the oxidative stability of biodiesel under different conditions, such as varying temperatures and storage durations, is essential.
- The interaction between ILs and biodiesel components, as well as their influence on the biodiesel oxidation process, needs further exploration. Identifying optimal ionic liquid formulations and concentrations for enhanced antioxidant effects in biodiesel is crucial.
- Evaluation of the economic feasibility and scalability of incorporating ILs as antioxidants in biodiesel production is lacking. Research addressing cost-effective and sustainable implementation methods is essential for practical applications.

2. Drug Delivery Systems

- While the potential of ILs in drug delivery is recognized, there is a scarcity of research specifically focused on their role as antioxidants in drug formulations. Investigating how ILs can protect drugs from degradation due to oxidation is an unexplored area.
- The compatibility of ILs with different types of drugs and their impact on drug stability needs systematic examination. Identifying specific drug-ionic liquid combinations that enhance drug delivery while maintaining stability is crucial for pharmaceutical applications.
- Limited research addresses the long-term effects of incorporating ILs into drug delivery systems. Understanding the potential interactions with biological systems and any unintended consequences is necessary for ensuring the safety and efficacy of such formulations.

Addressing these research gaps will not only enhance our understanding of the antioxidant properties of ILs in biodiesel and drug delivery but also pave the way for practical applications in sustainable energy production and pharmaceutical development.

Conclusion

To sum up, the application of ILs as antioxidants is an innovative and promising approach in the current developments of medicine delivery systems and biodiesel production. Because of their distinct physicochemical characteristics, which include their high thermal stability, low volatility, and chemical structures that can be adjusted, ILs are good options for improving delivery of drug formulations and preventing oxidative degradation in biodiesel. Research has indicated that (ILs) possess remarkable antioxidant properties, since they efficiently scavenge free radicals and impede the oxidation of feed-stocks used to make biodiesel.”

Furthermore, the shelf life and therapeutic efficiency of pharmaceutical compounds are improved by their capacity to function as stabilizing agents in drug delivery systems. In addition to helping with the problems caused by oxidative instability in biodiesel, ILs' multi-functionality also helps with drug release control and enhanced bioavailability. Because of this, the use of ILs as antioxidants demonstrates how revolutionary they can be for the biodiesel and pharmaceutical industries, opening the door to more cutting-edge therapeutic applications and environmentally friendly energy sources. This research highlights the importance of ILs as versatile and powerful antioxidants, creating new opportunities for investigation and use in these important domains.

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