Are Bio-Based Solvents Helpful in Controlling Environmental Pollution? Insights from The BRICS Countries

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***Abstract*—** Worldwide, certain countries require energy imports to meet energy requirements, but extreme weather events are increasing with climate change. Globally, most nations economically are shifting rapidly to zero-carbon energy. It is likely, however, that some newly developed renewable energy sources, such as biomass-based energy, will reduce the environmental stress imposed by fossil-based energy. Globally, nations are experiencing a transition fuelled by climate change, falling renewable energy costs, disruptive technologies, and more empowered consumers. This paper reviews the literature on bio-based solvents' application in separation processes. Separation of azeotropic and close-boiling mixtures is the only application covered. Challenges associated with implementing bioderived solvents at the industrial scale as alternatives to conventional solvents are discussed. Based on the study, comprehensive comments and recommendations were presented. According to the review, a biobased solvent-enhanced separation process should be considered a sustainable technology.

***Keywords*—** BRICS; Bio-derived solvent; Separation; Energy; Challenges.

I. Introduction

Brics Countries' economies are steadily growing due to their ongoing development and production. India and China have the biggest economies in the BRICS countries due to their high-tech, chemical, and petrochemical industries. The demand for their products has increased as their economies have grown, which has led to a rise in energy demand. In these developing economies, most processes employed by industries use large quantities of industrial solvents for purification purposes alongside energy demand concerns [1]. United Nations initiative requires all countries to achieve net-zero carbon footprints by 2050, including BRICS. Their products' increased demand increases the use of fossil fuel-based energy resources, increasing carbon emissions. This goes against UN policy. Approximately 80 to 90% of all solvents are used or produced in these industries, and 85% of the waste comes from industrial solvents. Undoubtedly, this issue directly or indirectly affects a significant portion of humans in various ways [2].

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Typically, industrial solvents are volatile organic solvents (VOCs), which cause severe and irreversible environmental problems due to their toxicity and volatility [3]. The majority of VOCs are mutagenic and carcinogenic so prolonged exposure can cause respiratory problems, reproductive issues, and skin irritation [4]. Without prior treatment, VOCs can pollute water bodies, soils, and airways [5].

It is crucial to transition towards sustainable and green solvents in BRICS countries to address the environmental concerns caused by the considerable consumption of environmentally unfriendly and highly toxic chemicals. These sustainable and green solvents must adhere to the 12 green principles of environmental chemistry [2]. Toxic and hazardous solvents cannot be used in the pharmaceutical and chemical industries. For production, these industries still require essential properties such as polarity and solubility of particular solvents. Pharmaceutical companies' Solvent Selection Guide recommends alternative solvents to replace toxic and environmentally harmful solvents [6].

In this context, this study reviews the progress of alternative solvents such as ionic liquids (ILs), deep eutectic solvents (DESs) and bio-based solvents (BBS). A focus will be placed on the positive applications of these solvents to industrial separation problems and the limitations that led to a shift to BBS from ILS and DESs. As a further step, the study will discuss the benefits of mixing BBS and DES precursors derived from biomass to form more bio-based DESs [7].

II. Ionic Liquids Application in Extraction Processes

Ionic liquids (ILs) are made entirely of cations and anions. Typically, they are in the liquid phase at temperatures below 100 °C or room temperature. The physicochemical properties of these solvents can be altered to meet certain desired applications [8]. Since their inception, ILs have been researched for various applications, including catalysis, electrochemistry, solvent chemistry, and extraction procedures. As a result of numerous industrial applications, ILs were given the phrase solvents of the future as they became substitutes for conventional solvents employed in industrial processes, which are highly flammable, hazardous, toxic and volatile. ILs possess low vapour pressures and

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contribute little to air pollution; therefore, they are considered green solvents. However, even though they are considered environmentally friendly solvents, IL production, usage and disposal must also be categorised as green solvents [9].

The first ionic liquid–based process was commercialised in 1996, and ever since, ILs have been employed in a broader range of applications. The aim of the section below highlights different processes where ILs are used to aid the extraction process.

A. Dissolution:

Plastic recycling is an issue being addressed by applying the ILs driven innovation. The convenient way to deal with the challenge posed by recycling plastics is that it is more cost-effective to incinerate or landfill the waste material. Since 2015, 6.3 billion tonnes of plastic waste has been produced; however, only 9% of the generated waste is recycled, 12% incinerated, and 79% disposed of through landfilling. Plastics usually comprise composite materials, frequently containing additives like pigments /plasticisers that need to be separated by chemical or mechanical processes [10]. Certain plastics have a chemical composition that facilitates easier chemical recycling, which involves breaking the polymer into monomers for further processing. A typical example of such plastic is PET (polyethylene terephthalate), popular in food packaging. It has been demonstrated that PET can be degraded by ionic liquids to release additives like pigments/plasticisers that need to be separated by employing chemical recycling [11].

B. Operating fluids:

Extensive research has been done on using ionic liquids as operating fluids rather than solvents, which operate as lubricants or heat transfer materials [12, 13]. Linde gas employed ILs as liquid pistons in an ionic compressor instead of traditional metal piston compressors. The IL was tuned to have low miscibility and low compressibility with the compressed gas. Additionally, the act of compressing gases to increase their pressure is exothermic, which necessitates heat transfer to take place. Heat exchangers directly outside the cylinder are customarily employed to eliminate unwanted heat. However, IL's high heat transfer capacity might also extract /remove heat directly inside the cylinder [8].

C. Dimerisation:

IFP (Institute Français du Pétrole) created a uniform IL-based catalyst to dimerise alkenes of lightweight. The co-catalyst of aluminium and soluble nickel salt was dissolved in hydrocarbon and introduced into the reactor system with alkene feed in the Dimersol process developed and made commercially in 1977 [14]. Upon the reaction completion, the reaction mixture had the catalyst, which was difficult to separate from the reaction mixture. Aqueous washing had to be employed to remove the catalyst, resulting in reduced yield and higher cost. To deal with the issues of low yield, reaction mixture and catalyst separation, the Dimersol process had to be improved into a biphasic Difasol. The biphasic Difasol process solubilises the nickel-based catalyst with the help of mildly acid imidazolium chloroaluminate IL [15]. The improved biphasic Difasol process allowed easy separation due to the low solubility of the dimerised products in the polar IL phase compared to the monomeric reactants, boosting yield, selectivity, and catalyst recyclability.

D. Micro-Extraction:

Microextraction is the extraction method where the volume of the extracting phase is small compared to the sample volume. Different microextraction techniques have employed ILsin immobilises and liquid form [16]. Sodium salicylate and coacervates developed via harmonious interchange of hydrotropic drug and ionic liquid have demonstrated exceptional capabilities to hide away dyes, curcumin and anti-cancer hydrophobic drugs; therefore, they can be used as effective micro extractants [17].

Yavir *et al.* [18] examined the effluence of IL properties as well as their structure on the variety of microextraction techniques such as single drop microextraction (SDME), solid-phase microextraction (SPME) and dispersive liquid-liquid microextraction (DLLME). Zhang *et al*. [19] combined high-performance liquid chromatography and dispersive liquid-liquid micro-extraction techniques for fast and effective separation of organophosphorus pesticide residue found in coarse cereals by using it as a dispersant and extracting medium. Zante *et al.* [20] reported on the success story of solvent extraction employing ionic liquids (1-butyl-3-methylimidazolium and tri-hexyl tetradecyl phosphonium chloride) for recovery of precious metals such as Co^{+2}, Li^{+2}, Mn^{+2} and Ni^{+2}. The concentration of Li^{+2} and Ni^{+2} ions in vegetable oil was successfully determined by merging flame atomic absorption spectroscopy as extraction solvent and dispersive liquid-liquid micro-extraction [21].

E. Biomass Extraction:

As a result of the demand for sustainable and clean energy sources, biomass extraction has gained attention. Biomass has high production, quick growth rate, and elevated photosynthetic efficiency, making macroalgae a significant renewable energy source [22]. Pretreatment technology based on ILs for processing macroalgae was examined by Gebrekrstos et al. [23]. In biomass extraction, ionic liquids can be utilised as extractive solvents. Microalgae biomass contains lipids, proteins, carbohydrates and bioactive compounds, which can be extracted using advanced technologies that use ionic liquids as extractive solvents. The extracted substances from microalgae (lipids, proteins, carbohydrates and bioactive compounds) are further used in producing biofuels, nutritive supplements and food additives [24]. Intracellular colorants can be recovered from natural producers of red colourants, such as Talaromyces amestolkiae, by using a series of imidazolium-based ILs such as 1-alkyl-3-methylimidazolium chloride ILs aqueous solutions [25].

Seaweeds contain primary and secondary metabolites that can be extracted using ionic liquids as extraction solvents. Primary and secondary metabolites could be helpful in the cosmetic and pharmaceutical industries [26].

F. Dissolution of Wood:

ILs are termed green solvents due to their fundamental properties: negligible vapour, solvation power, and the ability to dissociate cellulose and wood for manufacturing. Abushammala and Mao [27] emphasised the effect of IL structure on the dissolving of wood and the different challenges encountered during the dissolution process. ILs such as 1-butyl-3-methylimidazolium methyl sulfate and 1-butyl-3-methylimidazolium acetate are used to fractionate biomass from plants to produce lignin and cellulose [28]. Lignin from wood can be extracted effectively using protic ionic liquid 1-methylimidazolium chloride [29]. Lignocellulose and lignin are helpful bio-materials that can be dissolved and processed with ionic liquids to reduce reliance on fossil fuels [30].

III. DESs APPLICATION IN EXTRACTION PROCESSES

Shortcomings possessed by ILs have forced the development of DESs as their potential replacement. The study of DES has grown exponentially during the past two decades, yielding 2000 plus published papers in 2022. Eutectic solvents (ES) are liquid mixtures created by combining solid components, and as a result, the liquid phase is seen as solid-liquid equilibrium attained to form an eutectic system indicated by the decrease in melting points [31].

The terminology “deep” is emphasised when the depression on melting point is noticeably lower than that of estimated eutectic temperature when appropriate thermodynamic behaviour of liquid phase is assumed between DES precursors [32]. DESs and ILs have similarities stemming from their shared properties and characteristics. However, the DESs have the following advantages: biodegradability, non - flammability, non-toxicity, low volatility and chemical stability over ILs and conventional industrial solvents [33].

In recent years, DESs have displayed great promise for various industrial applications. Extensive research on the application of DESs has been done on carbon dioxide collection, separation and extraction processes, cosmetics, electrode position and biomass capture [34].

***Extraction Processes*:**

Sugars such as D-sorbital, D (+) xylose and xylitol, which act as HBD, and choline chloride, which act as HBA, are converted into DES by Carolina Gipiela Corrêa Dias *et al.* [35]. The thermophysical properties of DESs depend on temperature and the molar ratio of HBD:HBA. These sucrose-based DES can be employed in separation and food industries [2]. Recently developed DES were mostly Hydrophilic until Hydrophobic DES(HDES) were synthesised consisting of fatty acid and quaternary ammonium salt by Van Osch *et al.* [36].

As a result, a new application of DESs has emerged involving water-immiscible substances or solvents. Several Hydrophobic DES and their uses have been researched [37]. Recently, HDES have been employed for environmentally friendly extraction techniques. They have been used to extract heavy metals, isolate biomolecules and remove pesticides from agricultural run-offs [36]. Studies demonstrate that using HDES leads to enhanced extraction processes for several microanalytes due to the elevated solubility of hydrophobic solvents without causing reactive reactions.

HDES comprising of Thymol and tetra-decanol in the molar ratio of 2:1, respectively, was manufactured by Sai *et al.* [38] to extract biomass from furfural. In the enhancement of the sugar fermentation process, phenolic compounds are used. González *et al.* [39] employed menthol-derived HDESs to extract phenolic chemicals from aqueous effluent from biomass processing. The significant components of HDES are menthol and organic acids like octanoic acid, dodecanoic acid and decanoic acid. These organic acids have shown encouraging results in situations where HDESs can extract phenolic compounds more readily than glucose [2].

The concentration of micropollutants has increased due to the growing use of pesticides to combat insects [37]. Due to bio-accumulative action, micropollutants harm water bodies, living organisms and the soil [40]. In the quest to remove pesticides from water, Florindo *et al.* [41] used HDESs consisting DL-Methanol and tetrabutylammonium chloride salt as HBAs and different acids like pyruvic acid, acetic acid and levulinic acid as HBDs. Their competency compared to ILs often employed for similar purposes was evaluated.

The yield of 80% extraction efficiency was attained when water-stable DES that is based on DL-menthol and acids were employed for pesticides under study. The extraction of protein can be successfully achieved by using DESs derived from betaine and glycerol, urea, D-sorbitol, methyl urea and D(+)glucose as HBD. The demand for extracting components with medicinal properties and benefits has risen using cost-effective and little environmental risk methods. Flavonoids having antioxidants, anti-cancer and antimicrobial properties like robusta flavone and amentoflavone are also removed from Selaginella uncinata using DESs [2].

Thirty various DESs consisting of proline, lactic acid, betaine and choline chloride as HBDs with different HBAs such as malic acid, urea, sorbitol and glucose were synthesised by Liu *et al* [42] in order to extract robusta flavone and amentoflavone from S. uncinata. Additionally, to improve efficiency and reduce energy wastage and time consumption, the DESs-UAE based on ultrasound extraction techniques (DESs-UAE) were suggested over conventional extraction methods such as percolation, reflux and maceration. The results demonstrated that the combination of proline-lactic acid extracts robusta flavone and amentoflavone perfectly. The DESs-UAE approach achieved a high extraction rate, outperforming conventional techniques under optimised conditions and consuming less time [42].

IV. IONIC LIQUIDS AND DEEP EUTECTIC SOLVENTS SHORTCOMINGS

A. Ionic Liquids Shortcomings:

ILs were regarded as non-toxic compounds up until some studies revealed their toxicity. Due to their shallow vapour pressure, ILs were referred to as green solvents, becoming harmless compared to VOCs. IL evaporation into the environment has an insignificant effect. However, their solubility in water harms aquatic life. The high stability of ILs causes them to be less biodegradable [45]. ILs possess a certain level of toxicity due to their side chain length and the chosen cation and anions. Furthermore, ILs have a variety of additional shortcomings, such as high manufacturing cost, corrosivity, high viscosity, poor degradability, and restricted solute solubility. To address ILs' shortcomings, DES has been suggested as a suitable substitute [2].

B. DESs Shortcomings:

DES regeneration is a significant issue because DESs are mixtures instead of pure compounds [46]. Regeneration of two components combined at a specific ratio from a Liquid + DES mixture is extremely difficult and unfeasible. One of the hardest things to accomplish is the recovery of DESs from liquids; as a result, this hinders the employment of DESs in industrial sectors. Processes operated under atmospheric and ambient temperatures are preferred according to the sixth rule of green chemistry compared to those requiring high temperature and high pressure or vacuum conditions [47].

The low diffusion coefficient, high viscosity and low conductivity of some DESs and ILs need elevated temperatures in the practical uses of these solvents. However, increasing temperature and adding a non-viscous component (like water) to decrease viscosity would increase separation costs and energy [48].

V. BIO-BASED SOLVENTS:

Green and renewable substitutes that can be used instead of non-renewable organic /petroleum-based solvents are derived from biomass, casually referred to as bio-solvents, biomass-derived solvents or bio-based solvents. Aquatic plants, energy crops and forest products are typical examples of renewable biomass that can be used to generate bio-based solvents [43]. Bio-based solvents are considered greener alternatives to conventional solvents in research and industrial sectors as they offer some degree of sustainability. The desire to develop bio-based solvents is driven by the growing price of petroleum, the minimum carbon footprint of renewable feedstocks and the possibility of offering cleaner, less toxic and biodegradable solvents [43].

Bio-based solvents are accused of having volatile organic compounds as petroleum-derived compounds. Even after the alleged accusations,, these solvents are sustainable and play a massive role in legislative demands and consumer growth. In addition to NADESs (Natural DESs), other fascinating bio-based, non-toxic and biodegradable solvents have been assessed for different separation applications. Under separation applications, these bio-based solvents displayed characteristics similar to those of traditional solvents. Bio-based solvents are Cyrene, g-valerolactone, methyl (2,2-dimethyl-1,3-dioxolan-4-yl) methyl carbonate and 2-methyltetrahydrofuran to mention a couple [44].

CONCLUSIONS & RECOMMENDATIONS

Carefully selecting the solvent is essential according to the twelve principles of green chemistry. This means there must be a balance between solvent function, reduction of toxicity, biodegradability and the solvent must be produced from renewable resources. The shift from conventional solvents to ILS and then to NADESs, currently moving towards bio-based solvents, follows the twelve green chemistry principles. Therefore, bio-based solvents derived from biomass can control the pollution levels in BRICS countries as they are produced from renewable sources. These solvents pose little toxicity compared to VOCs, ILs and DESs. The important part when one develops a solvent-based separation is to make the recovery of the solvent easier. The extract phase can have energy levels that are pretty low due to the solvents being applied having excellent distribution coefficients. This leads to difficulty regenerating the solvents, necessitating a significant energy supply to separate the components.

When designing and developing a new solvent-based separation system, the designer should confirm that sustainability measures follow the green chemistry principles. The solvent-based separation process sustainability assessment considers the entire process, including the chemistry governing and applicable to solvent and solvent regeneration.

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CONFLICTS OF INTEREST

The authors declare no conflict of interest.

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