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## Review

## Techniques for recovery and recycling of ionic liquids: A review

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**Abbreviations:** 2-HEAF, 2-hydroxyethylammonium formate; AC, activated carbon; ACT, acteoside;  $Al_2(SO_4)_3$ , aluminium sulphate; APTS, aqueous two-phase system; BA, 3-amino-phenylboronic acid; CHI, chitosan; chol-IAA, choline-indole-3-acetate; CLSM, confocal laser scanning microscopy; CNFs, carbon nanofibers; CNG, cellulose nanocrystals; DNA, deoxyribonucleic acid;  $H_2C_2O_4$ , oxalic acid; IPA, isopropyl alcohol; JMs  $\oplus$  VPPs, Janus membranes with vertically penetrative pores;  $K_3PO_4$ , tripotassium phosphate;  $Na_2CO_3$ , sodium carbonate;  $Na_2SO_4$ , sodium sulphate;  $Na_3C_6H_5O_7$ , trisodium citrate;  $Na_3PO_4$ , trisodium phosphate; NaCl, sodium chloride;  $NaH_2PO_4$ , monosodium phosphate; NF, nanofiltration;  $[NH_4]_2SO_4$ , ammonium sulphate; NSAIDs, nonsteroidal anti-inflammatory drugs; PBMA, poly(butyl methacrylate); PMPC, poly(2-methacryloyloxyethyl phosphorylcholine); PTFE, polytetrafluoroethylene; RO, reverse osmosis; RS, rice straw; SBB, biochar derived from straw; SWCNT, single wall carbon nanotubes; TFP, 2,2,3,3-tetrafluoro-1-propanol; VPPs, vertically penetrative pores; [AMIM][Cl], 1-allyl-3-methylimidazolium chloride; [APPH][AC], 3-(aminopropyl)-2-pyrrolidonium acetate; [BMPYRR][CF<sub>3</sub>SO<sub>3</sub>], 1-butyl-1-methylpyrrolidinium triflate; [C<sub>2</sub>MIM][AC], 1-ethyl-3-methylimidazolium acetate; [C<sub>2</sub>MIM][BF<sub>4</sub>], 1-ethyl-3-methylimidazolium tetrafluoroborate; [C<sub>2</sub>MIM][CF<sub>3</sub>SO<sub>3</sub>], 1-ethyl-3-methylimidazolium triflate; [C<sub>2</sub>MIM][FSI], 1-ethyl-3-methylimidazolium bis(fluorosulfonyl)imide; [C<sub>2</sub>MIM][NTF<sub>2</sub>], 1-ethyl-3-methylimidazolium bistriflimide; [C<sub>2</sub>MIM][O<sub>2</sub>CH], 1-ethyl-3-methylimidazolium formate; [C<sub>4</sub>DMIM][NTF<sub>2</sub>], 1-butyl-2,3-dimethylimidazolium bistriflimide; [C<sub>4</sub>DMIM][OTF], 1-butyl-2,3-dimethylimidazolium trifluoromethanesulfonate; [C<sub>4</sub>MIM][AC], 1-butyl-3-methylimidazolium acetate; [C<sub>4</sub>MIM][ACE], 1-butyl-3-methylimidazolium acesulfamate; [C<sub>4</sub>MIM][BF<sub>4</sub>], 1-butyl-3-methylimidazolium tetrafluoroborate; [C<sub>4</sub>MIM][BR], 1-butyl-3-methylimidazolium bromine; [C<sub>4</sub>MIM][Cl], 1-butyl-3-methylimidazolium chloride; [C<sub>4</sub>MIM][HSO<sub>4</sub>], 1-butyl-3-methylimidazolium hydrogen sulphate; [C<sub>4</sub>MIM][NTF<sub>2</sub>], 1-butyl-3-methylimidazolium bistriflimide; [C<sub>4</sub>MIM][PF<sub>6</sub>], 1-butyl-3-methylimidazolium hexafluorophosphate; [C<sub>4</sub>MIM][TFO], 1-butyl-3-methylimidazolium trifluoromethanesulfonate; [C<sub>4</sub>PY][TFO], n-butylpyridinium trifluoromethanesulfonate; [C<sub>4</sub>MPYRR][NTF<sub>2</sub>], 1-alkyl-1-methylpyrrolidinium bistriflimide; [C<sub>6</sub>DIPA][IM], diisopropanolamine imidazole; [C<sub>6</sub>MIM][TFSA], 1-hexyl-3-methylimidazolium bis(trifluoromethylsulfonyl) amide; [C<sub>8</sub>MIM][Cl], 1-octyl-3-methylimidazolium chloride; [C<sub>8</sub>MIM][PF<sub>6</sub>], 1-octyl-3-methylimidazolium hexafluorophosphate; [C<sub>10</sub>MIM][Cl], 1-decyl-3-methylimidazolium chloride; [C<sub>12</sub>DMEA][IM], N,N-dimethylethylamine imidazole; [CO<sub>2</sub>ET], propionic acid; [(CH<sub>2</sub>)<sub>7</sub>COOHPYR][NTF<sub>2</sub>], pyridinium-based bistriflimide; [DBNH][AC], 1,5-diaza-bicyclo[4.3.0]non-5-enium acetate; [diBTMG][BR], hexalkyl-guanidinium bromine; [DMEA][HCOO], N,N-dimethyl-ethanolammonium formate; [EAPy][NTF<sub>2</sub>], 1-ethyl acetate-pyridine bistriflimide; [EIM][AC], 1-ethylimidazolium acetate; [EIM][Cl], 1-ethylimidazolium chloride; [HBTH][CF<sub>3</sub>SO<sub>3</sub>], benzothiazolium triflate; [HCO<sub>2</sub>ET], carboxylate; [MTBDH][AC], 7-methyl-1,5,7-triazabicyclo[4.4.0]dec-5-enium acetate; [MTBNH][AC], 5-methyl-1,5,7-triaza-bicyclo[4.3.0]non-6-enium acetate; [N<sub>2</sub>(2OH)(2OH)(2OH)][NTF<sub>2</sub>], N-ethyl-N,N,N-tris(2-hydroxyethyl) ammonium bistriflimide; [OHPy][NTF<sub>2</sub>], 1-hydroxyethyl-pyridine bistriflimide; [P<sub>4441</sub>][CH<sub>3</sub>SO<sub>4</sub>], tetrabutylmethyl-phosphonium methylsulphate; [P<sub>4448</sub>][BR], tetrabutylphosphonium bromide; [PYRR-EA][NTF<sub>2</sub>], N-(2-ethoxy-2-oxoethyl)-N-methyl-pyrrolidinium bistriflimide; [SUC][NTF<sub>2</sub>], succinyl bistriflimide; [TBHPP][Cl], phosphonium chloride; [TMG], 1,1,3,3-tetramethylguanidine; [TMGH][CO<sub>2</sub>ET], 1,1,3,3-tetramethylguanidine propionic acid.

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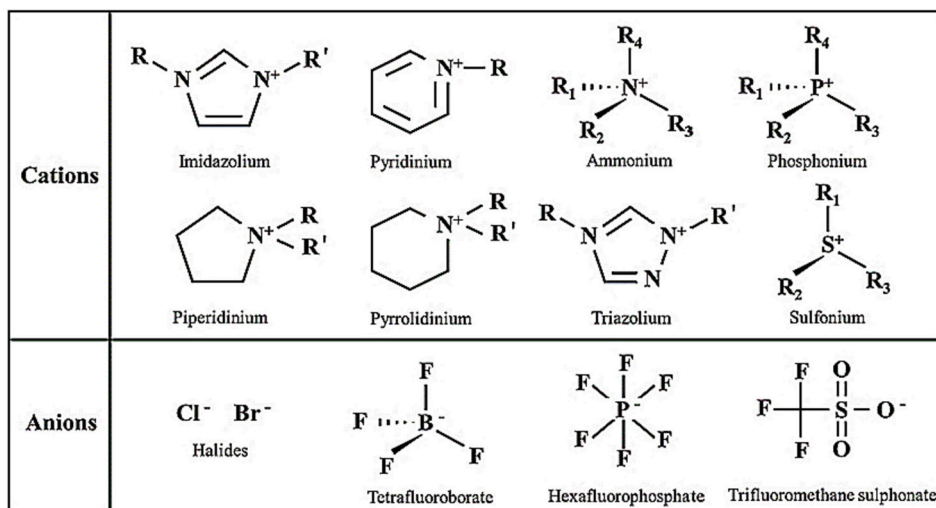
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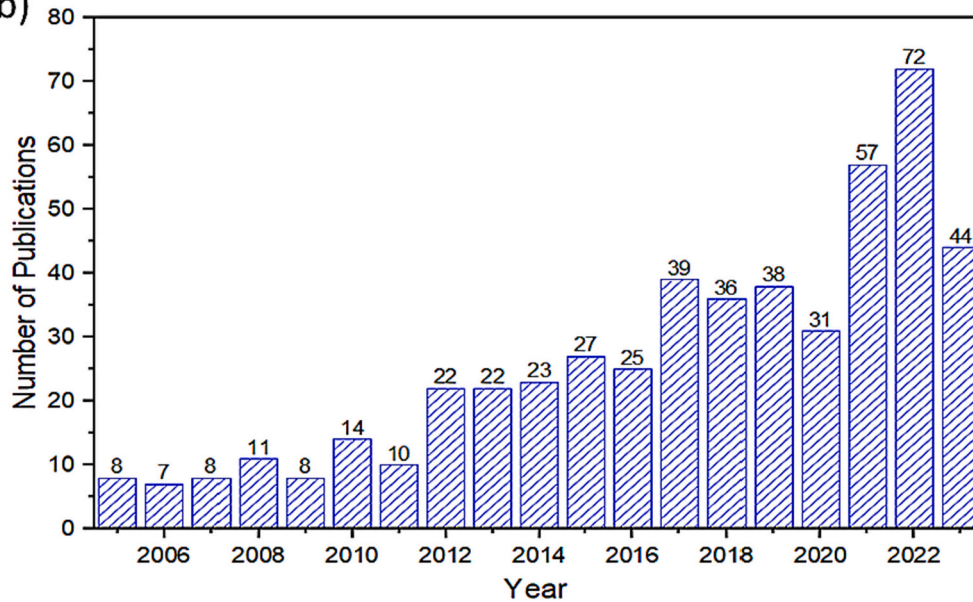


Fig. 1. (a) Common cations and anions of the ILs (Ren et al., 2023) and (b) number of relevant articles related to the recovery of ionic liquids published between 2005 and 2023. The data were obtained from Scopus. Search: ionic liquid recovery and recycling; Field: Article title, abstract, keywords).

## G R A P H I C A L A B S T R A C T

## H I G H L I G H T

- A comprehensive overview of ionic liquids recovery methods are reviewed.
- The properties of ionic liquids recovery techniques are summarized.
- Future perspectives of ionic liquids recycling methods are discussed.

## A R T I C L E I N F O

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## Keywords:

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Recovery and reuse  
Distillation  
Membrane separation  
Extraction  
Adsorption

## A B S T R A C T

Due to beneficial properties like non-flammability, thermal stability, low melting point and low vapor pressure, ionic liquids (ILs) have gained great interest from engineers and researchers in the past decades to replace conventional solvents. The superior characteristics of ILs make them promising for applications in fields as wide-ranging as pharmaceuticals, foods, nanoparticles synthesis, catalysis, electrochemistry and so on. To alleviate the high cost and environmental impact of ILs, various technologies have been reported to recover and purify the used ILs, as well as recycling the ILs. The aim of this article is to overview the state-of-the-art research on the recovery and recycling technologies for ILs including membrane technology, distillation, extraction, aqueous two-phase system (ATPS) and adsorption. In addition, challenges and future perspectives on ILs recovery are discussed. This review is expected to provide valuable insights for developing effective and environmentally friendly recovery methods for ILs.

## 1. Introduction

Over the past decades, there has been growing interest among researchers to investigate ionic liquids (ILs) (Kalb, 2020; Marcus, 2016) due to unique properties such as non-flammability, negligible vapor pressure and high thermal stability (Liu et al., 2020; Sowmiah et al., 2009). ILs, which consist of an organic or inorganic anion and an organic cation, exhibit melting points below 100 °C. As shown in Fig. 1(a), typical organic cations include ammonium (Bhatt et al., 2014; Sosnowska et al., 2020), sulfonium (Deyab, 2019; Sampaio et al., 2020a), imidazolium (Thakkar et al., 2016; Yang et al., 2019), triazolium (Brehm et al., 2020; Chu et al., 2020), pyridinium (Garcia et al., 2013), and phosphonium (Rodríguez-Escontrela et al., 2016; Sampaio et al., 2020b), while anions include  $\text{Cl}^-$ ,  $\text{Br}^-$ ,  $\text{NO}_3^-$ ,  $\text{BF}_4^-$ ,  $\text{PF}_6^-$ , and  $\text{SbF}_6^-$ , which can be chosen independently to design ILs based upon requirement. Notably, ILs are regarded as “solvents of the future” and “designer solvents” (Giernoth, 2010; Plechkova and Seddon, 2007), ascribed to the possibility to tailor and fine-tune the thermophysical characteristics to meet the practical demands of specific applications by varying the constituent cation and anion, substituent groups, functionalized alkyl groups as well as length of alkyl chain (Cagliero et al., 2020; Welton, 2018).

Unfortunately, one of the major drawbacks of adopting ILs in large-scale industrial processes is their price relative to conventional solvents (Hussain Solangi et al., 2023). It was also reported that the high production cost of ILs (\$50/kg) obstructed their commercial deployment for decades (Abouelela et al., 2022). Therefore, it is crucial to resolve the cost issue of ILs for example through improving the reusability. Recently, efforts have been directed to the recovery and recycling of ILs via several techniques such as distillation, membrane separation, extraction, adsorption, aqueous two-phase system (ATPS), crystallization, and so on. Fig. 1(b) shows the number of publications related to ILs recovery and recycling from year 2005 to 2023. The growth of research interest has been significant in the last 10 years and at least 30 papers has been published each year since year 2017. Although there is a significant amount of information regarding the positive impacts of these IL recycling methods reported in the scientific literature, the question yet to be answered is: Why are these methods seldom employed in the

industry? There are cases in which the researchers reported outstanding ILs recycling selectivity results via membrane separation, but the membrane permeate flux was adversely affected (Sosa et al., 2023a; Sosa et al., 2022). Also, despite distillation being a simple process to recover ILs, the anionic structure of the ILs could be destroyed as the pretreatment cycles increased.

Generally, distillation, especially vacuum distillation, is widely used and typically the first choice to recover ILs. Due to the low vapor pressure, the volatile substances can be vaporized from ILs. However, distillation is not preferred for thermal sensitive or non-volatile substances, in which case extraction is a more favourable method. The method used to recover ILs depends on the interaction between water and ILs, which can be affected by hydrophobicity/hydrophilicity properties of ILs, hydrogen bonding ability of both anions and cations, and size of ILs. For instance, the hydrophilic ILs can be recovered by adding salt (i.e., salting-out effect) via the ATPS method. In terms of membrane technology, high IL concentration can result in permeate flux deterioration (Sosa et al., 2023b) during the separation and recovery of the aggregated ILs (Mai et al., 2014).

Considering the increasing number of studies on the recycling and reuse of ILs, the aim of this review is to summarize the performance of IL after various recycling methods (e.g., distillation, membrane technology, adsorption, aqueous two phase system and extraction), the advantages and limitations of each methods, various challenges associated with their efficient utilization in various fields, IL degradation possibilities, future perspective as well as the economic feasibility of IL recycling. We hope that this review can provide valuable insights regarding designing and choosing suitable technologies for ILs recovery.

## 1.1. Application of ionic liquid

IL development has proliferated in diverse applications (Fig. 2), such as electrochemistry (Liu et al., 2021; Miao et al., 2021; Yi et al., 2021), organic synthesis (Vekariya, 2017; Yao et al., 2021; Zhang et al., 2011), analytical purification (Dharaskar et al., 2015; Paucar et al., 2021), food and pharmaceutical manufacturing (Gao et al., 2020a; Toledo Hijo et al., 2016; Zandu et al., 2019), environmental remediation (Krishnan et al., 2020; Lee et al., 2020; Sohaib et al., 2020), etc. For electrochemistry, ILs



with high ion conductivity and non-volatility properties have been treated as safe electrolytes to be used in various energy storage devices such as fuel cells, batteries, supercapacitors, solar cells, etc. Additionally, ILs attained from natural sources such as amino acids or choline can also be utilized in food industries to replace the conventionally used solvents. For instance, [De Souza Mesquita et al. \(2020\)](#) extracted carotenoids from fruit *Bactris gasipaes* by using ethanolic and ILs aqueous solution. Also, ILs have been utilized by pharmaceutical industries to enhance the drug delivery process. [Moshikur et al. \(2020\)](#) developed a *N*-methyl-2-pyrrolidone (NMP) cation-based active pharmaceutical ingredient IL for better drug delivery that improve drug accumulation in the skin. Moreover, ILs can act as solvent or stabilizer for nanoparticles synthesis. [Garg et al. \(2020\)](#) utilized choline-based ILs to synthesize palladium nanoparticles via hydrogenation. Besides, to circumvent the disadvantages of conventional catalyst in terms of reaction condition, selectivity, and yield, ILs are employed for better efficiency ([Kaur et al., 2022](#)). Furthermore, ILs play an important role in environmental remediation ([Ramos et al., 2020](#)). For instance, ILs are well-known for carbon capture and storage ([Hussain Solangi et al., 2023](#)). Also, nuclear wastes (actinides/lanthanides) can be treated via solvent extraction using ILs as extractant ([Iqbal et al., 2020](#)).

## 2. Recovery methods for ionic liquids (ILs)

### 2.1. Distillation

Distillation has been widely applied for separating liquid mixtures based on volatility differences of the components. To recover ILs, distillation is generally the first choice because of the high thermal stability (negligible vapor pressure) and low boiling point of ILs. Due to the non-volatility of ILs, the volatiles species in the mixture can be removed via the vacuum distillation method ([Zhou et al., 2018](#)). Specifically, the volatile liquid components are converted into gas phase under reduced pressure ([Fernandez et al., 2012](#)). Despite the high

energy consumption associated with distillation, it is generally applied as the final step in most of the published studies on recycling and recovering ILs ([Mai et al., 2014](#)).

[Xu et al. \(2017\)](#) utilized a rotary evaporator to recover 1-butyl-3-methylimidazolium acetate ([C<sub>4</sub>MIM][AC]) after the pretreatment of eucalyptus biomass. The used [C<sub>4</sub>MIM][AC] was introduced to the evaporator under reduced pressure at 80 °C over 6 h to eliminate the anti-solvent for the [C<sub>4</sub>MIM][AC] recovery process. The chemical structure of the recycled [C<sub>4</sub>MIM][AC] after the 4th cycle is similar to the fresh [C<sub>4</sub>MIM][AC], except that a new peak was detected at 4.04 ppm from the <sup>1</sup>H NMR spectra, which implies the accumulation of lignin fractions in the recycled [C<sub>4</sub>MIM][AC]. Nonetheless, the recycled [C<sub>4</sub>MIM][AC] was still capable of maintaining efficient enzymatic hydrolysis of around 72.8 %. In addition, the thermal stability of fresh and recycle [C<sub>4</sub>MIM][AC] was also assessed via TGA. The thermal stability of the recovered [C<sub>4</sub>MIM][AC] was basically unchanged compared to the fresh [C<sub>4</sub>MIM][AC]. However, there is a slight degradation located at 50 °C to 200 °C due to the absorbed water, dissolved lignin and carbohydrates products during the biomass pretreatment process.

[Huang et al. \(2020\)](#) utilized 1-butyl-2,3-dimethylimidazolium trifluoromethanesulfonate [C<sub>4</sub>DMIM][OTF] to enhance cellulose dehydration during the pyrolysis process to produce porous carbon. It was found that the sulfonate anions (OTF<sup>-</sup>) in the [C<sub>4</sub>DMIM][OTF] could lower the activation energy for dehydration and thus induce carbonization at low temperature. Upon pyrolysis, the carbon solid residue was filtered out while the filtrate mixture containing [C<sub>4</sub>DMIM][OTF] and acetone was evaporated at 55 °C under 100 mbar via a rotary evaporator for 3 h to eliminate acetone. Interestingly, [C<sub>4</sub>DMIM][OTF] can be recovered from the mixture with a recovery rate of >90 %. The authors proposed that some of the [C<sub>4</sub>DMIM][OTF] was lost to the gas phase during the volatilization during pyrolysis at 350 °C.

Most of the ILs are non-volatile. Nevertheless, according to [Earle et al. \(2006\)](#), ILs have low vapor pressure, and are detectable at high temperature (200 to 300 °C) instead of room temperature. [Earle et al.](#)

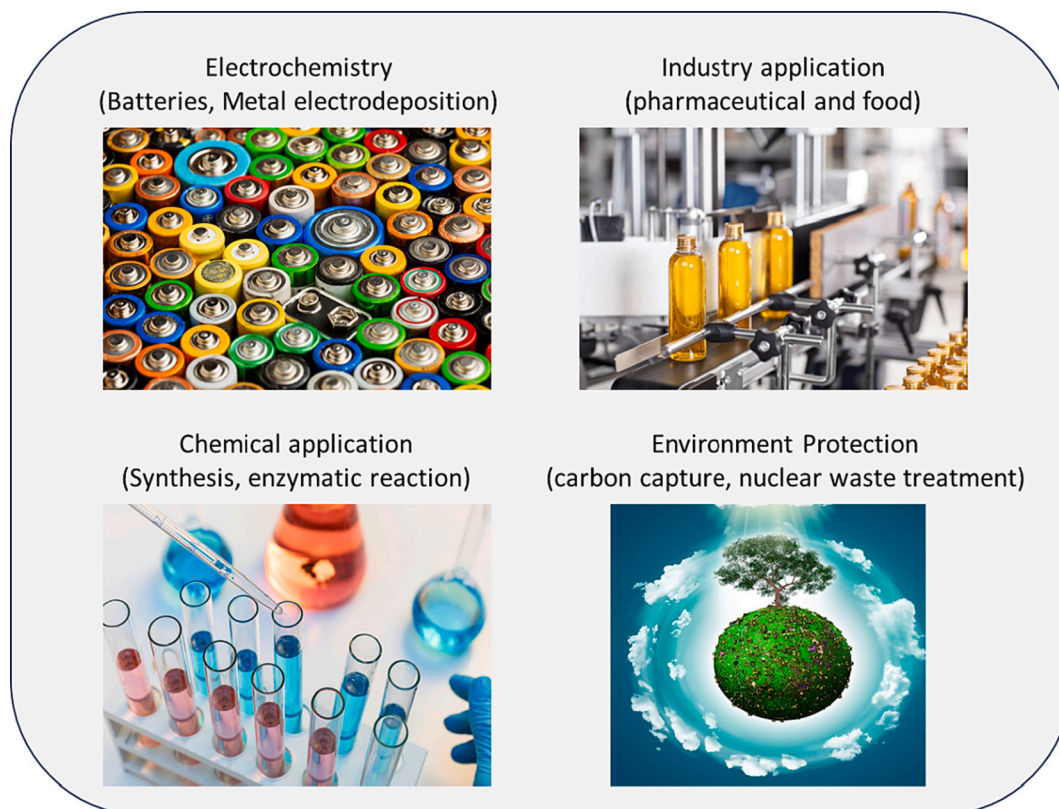


Fig. 2. Application of ILs for electrochemistry, industry applications, chemical applications and environmental protection.

(2006) demonstrated that the aprotic 1-butyl-2,3-dimethylimidazolium bistriflimide ([C<sub>4</sub>DMIM][NTF<sub>2</sub>]), 1-alkyl-1-methylpyrrolidinium bistriflimide ([C<sub>4</sub>MPYRR][NTF<sub>2</sub>]) and 1-butyl-3-methylimidazolium hexafluorophosphate ([C<sub>4</sub>MIM][PF<sub>6</sub>]) can be vaporized at 300 °C and 6 mbar with very little decomposition (<1.0 %), and then recondensed as pure ILs at a lower temperature.

Hossain et al. (2019) utilized the protic 1-ethylimidazolium chloride ([EIM][Cl]) and 1-ethylimidazolium acetate ([EIM][Ac]) to enhance the enzymatic hydrolysability of the pretreated biomass. The recycling of [EIM][Cl] was conducted by vaporizing the [EIM][Cl] at 200 °C for 1 h under vacuum condition and it was found that 93 % (by mass) of the original [EIM][Cl] was recovered. Furthermore, the recovered [EIM][Cl] showed no chemical structural change when compared to fresh [EIM][Cl], and the original acid:base ratio [EIM][Cl] employed was recovered in an identical ratio. Most importantly, the recovered [EIM][Cl] was distilled and reused three times to pretreat more wood with the same effectiveness. However, [EIM][Ac] was ineffective for biomass pretreatment, likely due to the weak acid and base properties of acetic acid and ethyl-imidazole, respectively, to form a true protic [EIM][Ac].

King et al. (2011) developed a new-generation acid-base conjugate 1,1,3,3-tetramethylguanidine ([TMG]) with propionic acid ([CO<sub>2</sub>ET]) to form [TMGH][CO<sub>2</sub>ET] for efficient biomass process by dissolving the cellulose rapidly. When [TMGH][CO<sub>2</sub>ET] was distilled at 200 °C over 30 min under 1.0 mmHg pressure, [TMGH][CO<sub>2</sub>ET] was able to dissociate into [TMG] and carboxylate ([HCO<sub>2</sub>ET]) during the distillation process and recombine to become [TMGH][CO<sub>2</sub>ET] with >99 % purity. This distillable characteristics is likely due to the free acid present in the reaction mixtures that allow the [TMG] dissociated into the neutral base and acid precursors at high temperature.

Recently, researchers are working towards new solvent which is more benign and greener for cellulose regeneration and fiber production, which fully depends on the solvent recyclability. This therefore shows regeneration of ILs in cellulose regeneration and fiber production process is a very important aspects in terms of sustainable manufacturing, circular economy, and commercialization. Elsayed et al. (2020) evaluated the recyclability of 7-methyl-1,5,7-triazabicyclo [4.4.0]dec-5-enium acetate ([MTBDH][Ac]) in comparison to 1,5-diaza-bicyclo[4.3.0]non-5-enium acetate ([DBNH][Ac]) after both were used as dissolution solvents for cellulose over 5 cycles. Firstly, a

centrifuge evaporator (62 °C, 250 mbar) was used to remove traces of water from the [MTBDH][Ac], followed by 1st stage and 2nd stage agitated thin-film evaporator with temperature and pressure ranging from 61 °C to 82 °C and 17 mbar to 20 mbar, respectively. Despite minor hydrolysis products detected at 2.78 ppm and 2.24 ppm for recovered [MTBDH][Ac], the <sup>1</sup>H NMR spectra were similar to the fresh [MTBDH][Ac] over 5 cycles (Fig. 3(a)). As evidenced in the SEM images in Fig. 3 (b–c), the recovered [MTBDH][Ac] at the 5th cycle exhibited full dissolution (99.99 %) of the 13 wt% cellulose, whereas only 76.5 % of cellulose dissolution was achieved using the single-cycle recovered [DBNH][Ac]. The deteriorated cellulose dissolution performance of recovered [DBNH][Ac] can be due to the high residual water content in the [DBNH][Ac] that hinder the cellulose dissolution ability and induce hydrolysis products.

Similarly, Parviainen et al. (2015) discovered that the hydrolysis products (3-(aminopropyl)-2-pyrrolidonium acetate, ([APPH][Ac])) generated during [DBNH][Ac] recovery process could diminish the cellulose dissolution capability. The authors reported that 41.5–54.9 mol% hydrolysis was found between the 5th and 6th of the [DBNH][Ac] recovery cycle after subjected to the vacuum rotary evaporator (1–3 mbar) at 60 °C for 2 h.

Brønsted acid-type 1-butyl-3-methylimidazolium hydrogen sulphate ([C<sub>4</sub>MIM][HSO<sub>4</sub>]) was utilized by Low et al. (2020) to synthesize crystalline cellulose nanoparticles (CNPs) from Microcrystalline Cellulose (MCC) via hydrolysis. Firstly, the mixing of acidic [C<sub>4</sub>MIM][HSO<sub>4</sub>] and MCC broke the inter and intramolecular hydrogen bonding of cellulose to produce the hydroxyl group. After adding DI water, the bonding of [C<sub>4</sub>MIM][HSO<sub>4</sub>] and cellulose was weakened, leading to the formation of high-crystallinity CNPs. The CNPs was obtained after centrifugation while the supernatant that contained [C<sub>4</sub>MIM][HSO<sub>4</sub>] was recovered via evaporation. Other than that, FTIR was used to analyze the chemical structure of recovered [C<sub>4</sub>MIM][HSO<sub>4</sub>], indicating almost similar peaks compared to pure [C<sub>4</sub>MIM][HSO<sub>4</sub>], except that the –OH group at peak 3440 cm<sup>-1</sup> was detected for recovered [C<sub>4</sub>MIM][HSO<sub>4</sub>]. This implies water was retained in the recovered [C<sub>4</sub>MIM][HSO<sub>4</sub>]. Overall, the [C<sub>4</sub>MIM][HSO<sub>4</sub>] recovery reached up to 90 % without producing any hazardous waste.

Zhang et al. (2022) proposed a new distillation and aeration apparatus (soxhlet extractor) to recycle 1-ethyl-3-methylimidazolium acetate

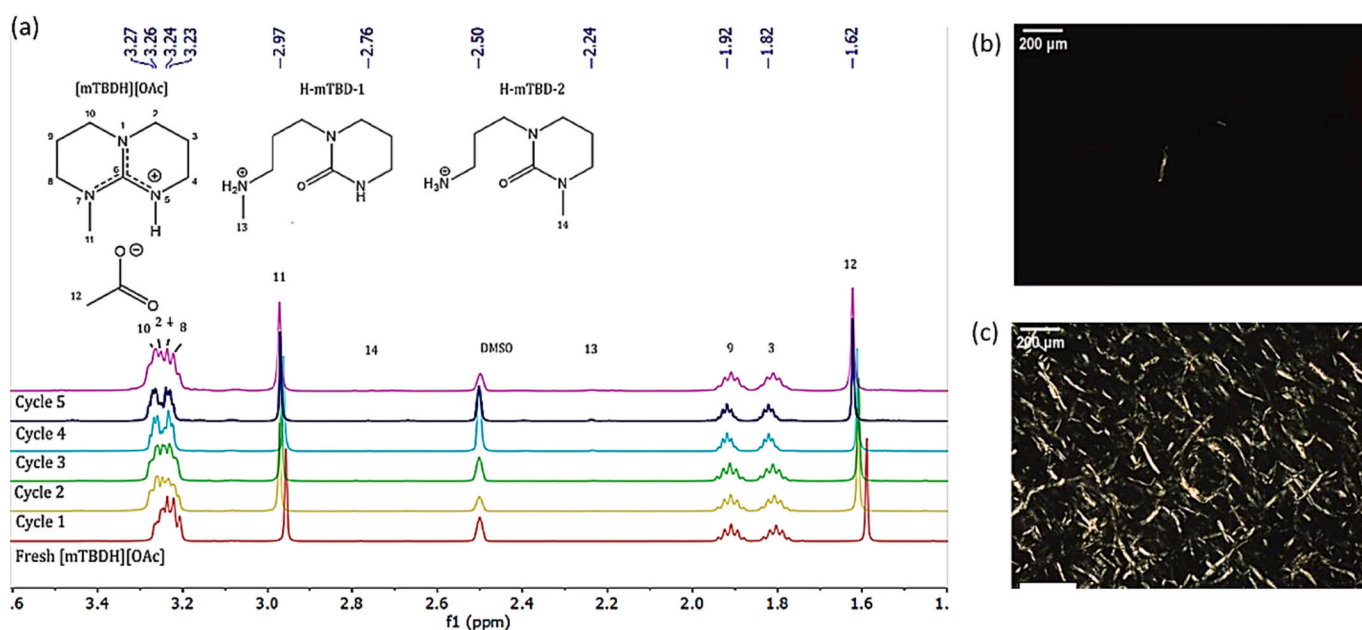


Fig. 3. (a) <sup>1</sup>H NMR peaks of fresh and recovered [MTBDH][Ac], and microscope image for the (b) dissolution of 13 wt% cellulose using recovered [MTBDH][Ac] (after 5th cycle) and (c) undissolved 13 wt% cellulose using recovered [DBNH][Ac] (after 1st cycle) (Elsayed et al., 2020).

([C<sub>2</sub>MIM][AC]) without drying or purification. Firstly, 0.3 g of a switchgrass sample was reacted with the fresh ([C<sub>2</sub>MIM][AC]) at 120 °C for 3 h, and the switchgrass slurry was washed with 19.2 g of water. The filtrate was then collected and treated as recycled ([C<sub>2</sub>MIM][AC]) to be used next cycle with 0.3 g of fresh switchgrass added. This new distillation apparatus was also important in removing water, and augmenting the interaction between ([C<sub>2</sub>MIM][AC]) and cellulose. X-ray diffraction (XRD) analysis and sugar yield performance to determine the performance of using recycled ([C<sub>2</sub>MIM][AC]). The XRD data show that all of the switchgrass treated with recycled ([C<sub>2</sub>MIM][AC]) gave higher pre-treatment efficiency compared to untreated switchgrass, with lower cellulose I structure detected at ~33.5°. Moreover, >80 % sugar yielded when using recycled ([C<sub>2</sub>MIM][AC]) (1st to 3rd cycles) to pretreat the switchgrass for delignification.

Jadhav et al. (2023) on the other hand reported the recovery of N-oxethylene substituted imidazolium-based zwitterions after cellulose dissolution. The cellulose dissolution with zwitterions was done at high temperature, therefore it generated metallic impurities, hemicellulose, degraded cellulose. These impurities must be eliminated in order to recycle and use the solvents in fiber processing. Firstly, the recovered zwitterions from cellulose dissolution will undergoes filtration and then enter the anion column to remove the hemicellulose/degraded cellulose, followed by the cation column to remove the metallic impurities. Evaporation steps is utilized to increase solvent concentration as per requirement and 99.6 % zwitterions solvent recovery rate was achieved. In this study, recovered solvent was again used to dissolve 10 % (w/w) cellulose ten times and the dope properties were shown to be comparable with the properties of dope produced initially with aqueous zwitterion. Table 1 summarized the ILs recovery findings via distillation process.

## 2.2. Membrane separation

Membrane technology such as reverse osmosis (RO) and nanofiltration (NF) is known as a low-energy, eco-friendly and high-efficiency method widely applied in purification and recovery in different sectors such as food (Bagci et al., 2019), pharmaceutical (Khoo

et al., 2022), wastewater treatment (Isoasaari et al., 2019), desalination and so on (Asadollahi et al., 2017; Khoo et al., 2021). Recently, membrane technology has been reported to be a promising method for ILs separation, especially NF, due to its ability to separate neutral and charged compounds, as well as monovalent and divalent ions (Mai et al., 2014; Zhou et al., 2018). Other than ionic separation, membrane is also more effective in separating low volatile components than that of other separation techniques such as distillation (Hinchliffe and Porter, 2000; Kuzmina, 2016; Mai et al., 2014). For instance, Fernández et al. (2011) attempted to recover the hydrophobic 1-hexyl-3-methylimidazolium bis (trifluoromethylsulfonyl) amide ([C<sub>6</sub>MIM][TFSA]) residues by concentrating and promoting the formation of a new phase of [C<sub>6</sub>MIM][TFSA] via a commercial NF270 membrane. Owing to the nucleation-growth mechanism, the new phase of [C<sub>6</sub>MIM][TFSA] formed when the [C<sub>6</sub>MIM][TFSA] concentration in the retentate was above the [C<sub>6</sub>MIM][TFSA] solubility during the filtration. It was reported that small drops of the new liquid phase can be observed, and the IL phase can be separated by sedimentation after 2 days. Further investigation showed that the NF270 membrane exhibited remarkable [C<sub>6</sub>MIM][TFSA] recovery rate and permeate flux of 75.6 % and 33.1 L/m<sup>2</sup>·h, respectively, using a dead-end stirred cell at 35 bar. Membrane pore size and the ions sizes of ILs are important criteria that need to be taken into account for membrane separation. Wang et al. (2016) investigated the use of commercially available NF membrane, namely, NF90 and NF270, to recycle 5 wt% of 1-butyl-3-methylimidazolium chloride ([C<sub>4</sub>MIM][Cl]), 1-butyl-3-methylimidazolium tetrafluoroborate ([C<sub>4</sub>MIM][BF<sub>4</sub>]) and [AMIM][Cl] aqueous solution. It was reported that, due to the smaller membrane pore size of NF90, higher retention was obtained for [C<sub>4</sub>MIM][Cl] at 96 % compared to that of NF270 membrane at 90 %. Moreover, the excellent [C<sub>4</sub>MIM][Cl] retention was also mainly due to the larger [C<sub>4</sub>MIM]<sup>+</sup> cation.

Instead of using the commercially available NF membrane, Avram et al. (2017) incorporated 3-amino-phenylboronic acid (BA) into the membrane PA layer during interfacial polymerization (IP) of the NF membrane in order to separate sugar and [C<sub>4</sub>MIM][Cl]. Some of the studies reported that the hydroxyl groups of BA are capable to form bonding with hydroxyl groups of sugar (Liu et al., 2012; Zhao and

**Table 1**  
Summary of ionic liquids recovery findings via distillation process.

Ionic liquids	Pressure, mbar	Temperature, °C	Time, h	Key Findings	Ref
[EIM][Cl]	8–10	200	1	93 % of [EIM][Cl] was recovered. The recycled [EIM][Cl] can maintain the biomass pre-treatment performance throughout 4 cycles distillation recovery.	(Hossain et al., 2019)
[AMIM]Cl and [C <sub>4</sub> MIM][AC]	–	80	6	Both [AMIM]Cl and [C <sub>4</sub> MIM][AC] showed admirable recyclability with 92 % recovery yield for the first 4 cycles. Yield 90 % recovery after reused for 4 cycles.	(Xu et al., 2017)
[C <sub>4</sub> DMIM][OTF]	100	55	3	97 wt% of [C <sub>4</sub> DMIM][OTF] was recovered.	(Huang et al., 2020)
[DMEA][HCOO]	0.75	122	–	99 % of [DMEA][HCOO] was recovered.	(Idris et al., 2014)
[MTBDH][AC]	250 20 17	62 61 82	– – –	Fed into centrifuge evaporator at 400 rpm, 50 kg/h. Fed into agitated thin-film evaporator (TFE-1) at 400 rpm, 3.5 kg/h. Fed into agitated thin-film evaporator (TFE-2) at 400 rpm, 3.5 kg/h. The [MTBDH][AC] yield a recovery value of 95.7 % in the concentrate stream after reused at 5th cycle.	(Elsayed et al., 2020)
[C <sub>4</sub> MIM][BR]	Low P Low P	70 90	–	Ethanol was evaporated at 70 °C in low pressure. Water was evaporated at 90 °C in low pressure. The recovery rate of [C <sub>4</sub> MIM][BR] was recorded as 82.48 % after 5 run.	(Chen et al., 2017)
[C <sub>2</sub> MIM][AC]	–	65	–	The sugar yield between 57.31 and 45.88 mg after [C <sub>2</sub> MIM][AC] was reused for 5 cycles.	(Chuetor et al., 2022)
[C <sub>2</sub> MIM][AC]	–	120	3	The residual (diluted [C <sub>2</sub> MIM][AC]) was reused for another 3 cycles. The sugar yield approximately 90 % after 0.5 g of fresh [C <sub>2</sub> MIM][AC] was topped up at 3rd cycle.	(Zhang et al., 2022)
[C <sub>2</sub> MIM][AC]	–	130	3	The [C <sub>2</sub> MIM][AC] recovery achieved 81 % after reuse for 3 cycles.	(Yuan et al., 2017)
[C <sub>4</sub> MIM][AC]	–	75	–	The products yield reduced from 92.5 % to ~70 % when the [C <sub>4</sub> MIM][AC] was reused for 4 cycles.	(Trinh et al., 2018)



Shantz, 2011), thus inhibiting the sugar passage while enhancing  $[C_4MIM][Cl]$  diffusivity. When 0.1 wt% of the BA was added, the rejection of glucose reached as high as 97.8 % while only 19.4 % of  $[C_4MIM][Cl]$  was rejected with permeability at 2.31  $L/m^2 \cdot h \cdot bar$ . This indicates that more  $[C_4MIM][Cl]$  was allowed to pass through the membrane to the permeate side.

Besides membrane pore size, the pore structure also plays an important role in recovering ILs, especially oil-like ionic liquids (ILs) in water emulsion. Yang et al. (2021) demonstrated that the Janus membrane with vertically penetrative pores (VPPs), which are less tortuous than the sponge-like pores (SPs) of conventional membranes, were more suitable for high-viscosity ILs separation. As shown in Fig. 4(a), 1-ethyl-3-methylimidazolium bistriflimide ( $[C_2MIM][NTF_2]$ ) exhibited higher permeability through the IL-phobic side to the IL-philic side of VPP-1 membrane than that of SP membrane, owing to the shorter path lengths. Other than that, the IL permeance and recovery ratio were further analyzed by using different ILs with diverse viscosities, as depicted in Fig. 4(b). Results indicate that the recovery ratio declined sharply from 95.6 % to 80.1 % with increasing viscosity for  $[C_2MIM][NTF_2]$  ( $\mu = 28.4$  mPa·s) and 1-octyl-3-methylimidazolium hexafluorophosphate ( $[C_8MIM][PF_6]$ ) ( $\mu = 694.5$  mPa·s). It was suggested that the high mass transfer resistance of the highly viscous ILs extended the retention time within the membrane channels, resulting in reduced IL recovery. The IL-in-water emulsions separation was examined, as presented in Fig. 4(c), the results showing that the emulsion phenomenon was hardly found after the separation process, which confirms the successful IL separation from water.

Recently, Sosa et al. (2022) utilized the commercial pressure-driven NF and RO membranes (namely, NF270 and BW30LE-RO) for the recovery of two superbase ILs (namely,  $[MTBDH][AC]$  and 5-methyl-1,5,7-triaza-bicyclo[4.3.0]non-6-enium acetate ( $[MTBNH][AC]$ )). Due to the smaller pore size of the RO membrane, the BW30LE-RO membrane achieved >80 % ILs retention, which is higher than the 68 % of the NF270 membrane, after being subjected to 5 wt% of  $[mTBDH][OAc]$  and  $[mTBNH][OAc]$  at 40 bar. It must be pointed out that the  $[MTBDH]$

$[AC]$  and  $[MTBNH][AC]$  retention performance diminished with increasing concentration. As the  $[MTBDH][AC]$  and  $[MTBNH][AC]$  concentration increased up to 20 wt%, no flux and retention were observed for the BW30LE-RO membrane. From these results, it was concluded that higher  $[MTBDH][AC]$  and  $[MTBNH][AC]$  concentrations caused an IL film atop the membrane layer and gradually neutralized the membrane charges, and thus the  $[MTBDH][AC]$  and  $[MTBNH][AC]$  rejection rate deteriorated as a result of weak electrical repulsion forces.

Chang et al. (2019) introduced hydrophilic poly(2-methacryloyloxyethyl phosphorylcholine) (PMPC) and IL-philic poly (butyl methacrylate) (PBMA) polymer networks, which are capable of locking  $[C_4MIM][PF_6]$  or water to form hydrogel film and ionogel film, respectively (Fig. 5(a)), onto the porous stainless steel membranes (PMs). As presented in Fig. 5(b), PBMA-PM exhibited good affinity towards  $[C_4MIM][PF_6]$  but repelled water upon the formation of the ionogel film.  $[C_4MIM][PF_6]$  hardly passed through the PMPC-PM membrane and only water selectively diffused through. Fig. 5(c) and (d) shows different separation efficiencies for different ILs when using PBMA-PM and PMPC-PM, respectively. Notably, ILs viscosity plays a significant role that affect the flux passing through the membrane. PBMA-PM gave lower flux while maintaining higher ILs separation efficiencies at the permeate side (>98 %), whereas PBMA-PM gave higher water flux (ranging from 30,000 to 55,000  $L/m^2 \cdot h$ ) with separation efficiencies consistently >98 % at the feed side.

Other than pressure-driven membrane-filtration, pervaporation is also a membrane-based separation technology with either vacuum or sweep gas at the permeate side to produce a chemical potential differential for permeate to be removed in the vapor state (Baker, 2012; Vane, 2005). Pervaporation offers the advantage in separating azeotropic mixtures with close boiling points (whereby distillation is ineffective), organic-organic mixtures, thermally sensitive compounds as well as dilute organics from aqueous solutions (Jiang et al., 2009; Mujiburohman and Feng, 2007; Ong et al., 2011; Semenova et al., 1997; Smitha et al., 2004). Sun et al. (2017) reported that up to 99.95 % of  $[C_2MIM]$

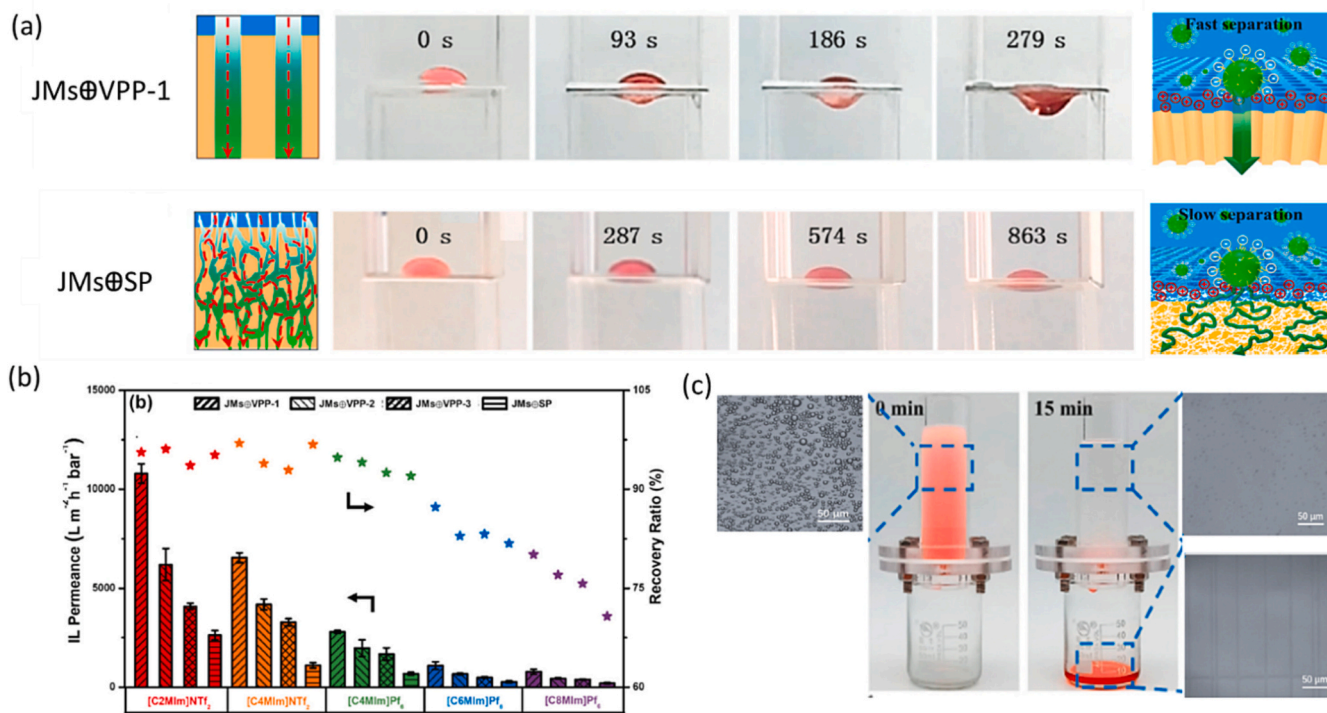


Fig. 4. (a) Schematic diagram (left and right) and image view (middle) of 20  $\mu L$  IL droplet transportation underwater from the from the IL-phobic side to the IL-philic side of JMs @ VPP-1 and JMs @ SP, (b) various type of ILs recovery ratio and permeances in water emulsions of different membranes, (c) photographs of the  $[C_2MIM][NTF_2]$  water emulsion separation using JMs @ VPP-1 with the optical microscopic image before and after separation (Yang et al., 2021).

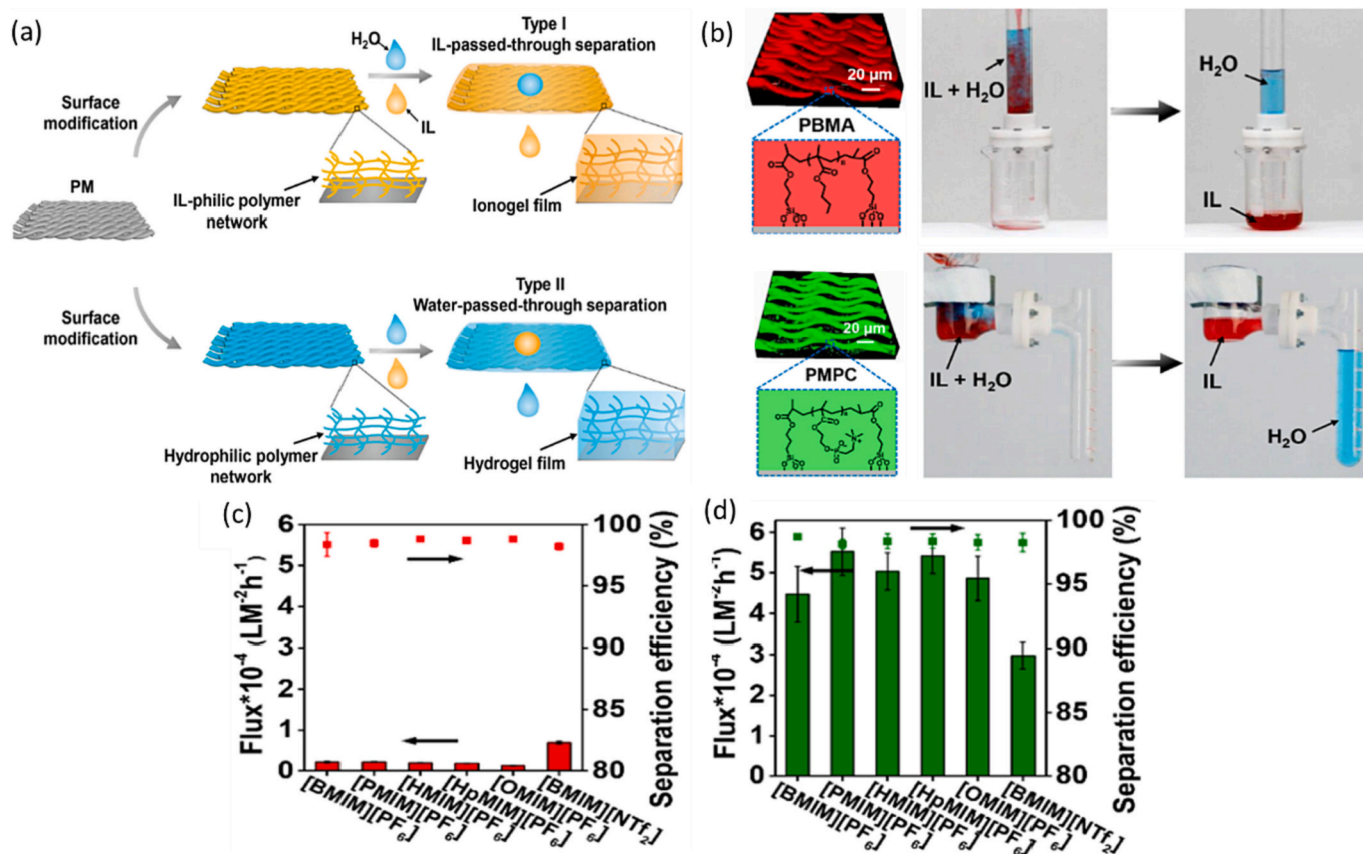


Fig. 5. (a) Schematic diagram of the IL-philic ionogel and hydrophilic hydrogel film design concept, (b) confocal laser scanning microscopy (CLSM) images of PBMA-PM (top) and PMPC-PM (bottom) and the corresponding separation performance, flux and separation efficiency of ionic liquid from water mixtures using (c) PBMA-PM and (d) PMPC-PM (Chang et al., 2019).

[AC] can be recovered at a constant flux of 0.47 L/m<sup>2</sup>·h after 50 g of [C<sub>2</sub>MIM][AC] containing 52 wt% of water was introduced to the pervaporation (PV) process at 80 °C over 60 days. Further experiments were carried out to investigate the biomass pretreatment efficiency of recycled [C<sub>2</sub>MIM][AC]. Compared to the fresh [C<sub>2</sub>MIM][AC], the 5th recycled [C<sub>2</sub>MIM][AC] performed well and yielded comparable glucose level (>90 %) after 80 wt% of [C<sub>2</sub>MIM][AC] was added to the biomass pretreatment process at 160 °C for 1 h. Table 2 summarized the ILs recovery findings via membrane technology.

### 2.3. Extraction

Other than distillation and membrane technology, extraction have gained high interest since it is considered as an energy-efficient technology that involves two immiscible phases (conventionally, an aqueous phase and organic phase). Recently, ILs have been used as an ideal substitute due to its nonvolatility, adjustable miscibility and stability properties. The extraction of thermal sensitive or non-volatile products from ILs has been facilitated via liquid-liquid extraction to allow the reuse and recycling of ILs (Zhao et al., 2005). For instance, water can be used to extract hydrophilic products from hydrophobic ILs. When the products exhibit immiscibility with water and mixtures with ILs, it is more suitable to apply organic solvents such as hexane (Earle et al., 1999; Fukuyama et al., 2002), toluene (Luo et al., 2018), cyclohexane (Titze-Frech et al., 2013), ethyl acetate (Mukesh et al., 2013; Singh et al., 2017), diethyl ether (Earle et al., 1999; Handy and Zhang, 2001; Kralisch et al., 2007; Moreau et al., 2006) and so on to extract products from ILs. ILs have been considered as unique solvents that have been used protein stabilization due to their remarkable solvation characteristics (Fujita et al., 2005).

Mukesh et al. (2013) utilized a bio-based choline-indole-3-acetate (chol-IAA) to dissolve 3.5 % w/w deoxyribonucleic acid (DNA) from salmon for a dissolution duration of up to 6 h. It was reported that DNA can be regenerated from chol-IAA by adding ice-cold isopropyl alcohol (IPA), followed by introducing ethyl acetate to regenerate chol-IAA with a recovery rate of 90–95 %. The recovered chol-IAA was recycled and reused for three consecutive cycles (fresh DNA dissolution), and it was found out that no degradation of the DNA molecules occurred.

Similarly, Singh et al. (2017) dissolved 25 % w/w of salmon DNA using 2-hydroxyethylammonium formate (2-HEAF) in order to preserve the DNA over long durations. As shown in Fig. 6(a), the DNA was mixed with 2-HEAF and stirred for 12 h at 25 °C to complete the DNA dissolution process. Ice-cold ethanol was used to regenerate the DNA (99.8 % of DNA recovery) while ethyl acetate was added to recover 2-HEAF (93.11 % of 2-HEAF recovery). Interestingly, 24.5 % w/w of fresh DNA can be dissolved after subjected to the recycled 2-HEAF.

Peng et al. (2022) attempted to recover 2,2,3,3-tetrafluoro-1-propanol (TFP) by using 1-ethyl acetate-pyridine bistriflimide ([EAPy][NTF<sub>2</sub>]) and 1-hydroxyethyl-pyridine bistriflimide ([OHPy][NTF<sub>2</sub>]), via liquid-liquid extraction (Fig. 6(b)). After the extraction process, the extracted ILs was placed in the oven at 80 °C for 24 h to eliminate the water contents for future reuse purpose. FTIR results in Fig. 6(c) shows that the recovered [EAPy][NTF<sub>2</sub>] had consistent peaks with the fresh [EAPy][NTF<sub>2</sub>]. Fig. 6(d) compares the extraction efficiency of TFP using fresh and recovered [EAPy][NTF<sub>2</sub>] and [OHPy][NTF<sub>2</sub>]. The 4th-cycle recycled [EAPy][NTF<sub>2</sub>] and [OHPy][NTF<sub>2</sub>] shows outstanding extraction performance (>85 %) almost similar to the fresh [EAPy][NTF<sub>2</sub>] and [OHPy][NTF<sub>2</sub>].

Apart from DNA dissolution application, the extraction of metal ions using ILs has gained much attention recently because of the potential to

**Table 2**  
Summary of ionic liquids recovery via membrane technology.

Ionic liquids	Membrane type	Pressure, bar	Permeability, L/m <sup>2</sup> ·h·bar	Key Findings	Ref
[C <sub>4</sub> MIM][Cl]	NF	6.9–17.25	2.31	19.4 % of [C <sub>4</sub> MIM][Cl] was rejected at feed side when 10 % of [C <sub>4</sub> MIM][Cl] was used as feed.	(Avram et al., 2017)
[C <sub>6</sub> MIM][TFSA]	NF270	35	0.95	[C <sub>6</sub> MIM][TFSA] achieved a recovery rate of 75.6 %	(Fernández et al., 2011)
[C <sub>4</sub> MIM][PF <sub>6</sub> ]	Stainless steel porous membrane	–	–	98 % of [C <sub>4</sub> MIM][PF <sub>6</sub> ] was recovered in permeate side.	(Zhang et al., 2017)
[MTBNH][AC]	NF270	40	~2	~60 % of [MTBNH][AC] was retained in the feed side after tested using 5 wt% [MTBNH][AC]. Water was filtered out at permeate site.	(Sosa et al., 2022)
[MTBDH][AC]	NF270	40	~1.75	~68 % of [MTBDH][AC] was retained in the feed side after tested using 5 wt% [MTBDH][AC]. Water was filtered out at permeate site.	(Sosa et al., 2022)
[MTBNH][AC]	BW30LE-RO	40	~0.55	~80 % of [MTBNH][AC] was retained in feed side after tested using 5 wt% [MTBNH][AC]. Water was filtered out at permeate site.	(Sosa et al., 2022)
[MTBDH][AC]	BW30LE-RO	40	~0.45	~85 % of [MTBDH][AC] was retained in the feed side after tested using 5 wt% [MTBDH][AC].	(Sosa et al., 2022)
[C <sub>4</sub> MIM][Cl]	NF90	25	2.124	The retention of [C <sub>4</sub> MIM][Cl] was up to almost 96 %.	(Wang et al., 2016)
[C <sub>4</sub> MIM][PF <sub>6</sub> ]	PBMA-PM	–	–	Obtained lower flux (~2000 L/m <sup>2</sup> ·h) but higher separation efficiency (>98 %).	(Chang et al., 2019)
[C <sub>4</sub> MIM][PF <sub>6</sub> ]	PMPC-PM	–	–	[C <sub>4</sub> MIM][PF <sub>6</sub> ] was retained at feed side while water allowed to pass through membrane. Obtained higher flux (~45,000 L/m <sup>2</sup> ·h) and higher separation efficiency (>98 %).	(Chang et al., 2019)
[C <sub>2</sub> MIM][NTF <sub>2</sub> ]	Polydopamine/poly(diallyldimethylammonium chloride) modified JMs ⊕ VPPs	–	>10,000	[C <sub>2</sub> MIM][NTF <sub>2</sub> ] recovery ratio of JMs ⊕ VPP-1 reached 95.6 ± 1.6 %.	(Yang et al., 2021)
[C <sub>2</sub> MIM][AC]	PTFE membrane	0.051	~353	Water was evaporated out while [C <sub>2</sub> MIM][AC] was retained at feed side with concentration increased from 5 % to 45 %.	(Lynam et al., 2016)
[C <sub>2</sub> MIM][O <sub>2</sub> CH]	PTFE membrane	0.051	~343	[C <sub>2</sub> MIM][O <sub>2</sub> CH] was retained at feed side with concentration increased from 5 % to 60 %.	(Lynam et al., 2016)

be a “green solvent” for preconcentration, separation and extraction of heavy metal ions from aqueous phase (Cocalia et al., 2006; Mancini et al., 2013; Visser et al., 2001; Zhao et al., 2005). Wang et al. (2021a) investigated gold (Au(III)) recovery from electronic waste by using hydrophobic succinyl bistriflimide [SUC][NTF<sub>2</sub>]<sub>2</sub>. Au(III) was almost completely stripped out from the loaded [SUC][NTF<sub>2</sub>]<sub>2</sub> phase by oxalic acid after extraction and [SUC][NTF<sub>2</sub>]<sub>2</sub> was then recycled back. The stripping raffinate containing [NTF<sub>2</sub>]<sup>-</sup> anion was then mixed with the stripping raffinate consist of [SUC]<sup>2+</sup>, with [Li][NTF<sub>2</sub>] added to ensure the formation of regenerated [SUC][NTF<sub>2</sub>]<sub>2</sub>. It was also reported that the extraction efficiency of Au(III) in each cycle decreased gradually, dropping from ~98 % to 87.6 % after 5 cycles. The deteriorated Au(III) extraction efficiency using the recycled [SUC][NTF<sub>2</sub>]<sub>2</sub> can be due to the loss of [SUC][NTF<sub>2</sub>]<sub>2</sub> during the transfer process.

Likewise, Liu et al. (2023) utilized N-(2-ethoxy-2-oxoethyl)-N-methyl-pyrrolidinium bistriflimide [PYRR-EA][NTF<sub>2</sub>] to extract Au(III) from the discarded CPU. Aqua regia was used as leaching agent while [PYRR-EA][NTF<sub>2</sub>] was used to extract Au(III) from leachate. It was reported that 99.39 % of Au(III) extraction efficiency was achieved after extraction using [PYRR-EA][NTF<sub>2</sub>]. Afterwards, oxalic acid (H<sub>2</sub>C<sub>2</sub>O<sub>4</sub>) was used as the stripping agent to recover [PYRR-EA][NTF<sub>2</sub>], and almost 100 % of [PYRR-EA][NTF<sub>2</sub>] was regenerated. It must be pointed out that [PYRR-EA][NTF<sub>2</sub>] was turned into [PYRR-EA][Cl] after reacting with H<sub>2</sub>C<sub>2</sub>O<sub>4</sub>, while NTF<sub>2</sub><sup>-</sup> was transferred to the water phase. In order to obtain the regenerated [PYRR-EA][NTF<sub>2</sub>], the stripping solution and raffinate were mixed together, followed by DI water rinsing to remove the residual H<sub>2</sub>C<sub>2</sub>O<sub>4</sub>. The regenerated [PYRR-EA][NTF<sub>2</sub>] was then reused for the extraction process and the Au(III) extraction efficiency decreased slightly from 95.8 % to 84.3 %.

Pei et al. (2018) developed a novel oil-water phase separation via an IL-based microemulsion reversible switching mechanism that composed of n-pentanol, water and CO<sub>2</sub>-responsive N,N-dimethylethylamine imidazole ([C<sub>12</sub>DMEA][IM]). The monophasic microemulsion separated

into two phases after CO<sub>2</sub> bubbling and reverted back to microemulsion monophase after the removal of CO<sub>2</sub>. Benzaldehyde and malononitrile were added into n-pentanol/[C<sub>12</sub>DMEA][IM]/H<sub>2</sub>O microemulsions to produce 2-benzylidenemalononitrile. Upon bubbling of CO<sub>2</sub> for <10 min at 25 °C, the product (benzylidenemalononitrile) precipitated in the water phase and was filtered out (97.2 % of benzylidenemalononitrile yielded). The microemulsion phase was then regenerated after CO<sub>2</sub> was eliminated by N<sub>2</sub> bubbling. The reversible switching mechanism was mainly ascribed to the reversible formation of carbonate and bicarbonate from the reaction between CO<sub>2</sub> and anions, resulting in the increase of ionic strength in the mixtures, and caused salting-out effect to occur during microemulsion (Mercer and Jessop, 2010). Additionally, benzylidenemalononitrile yield was slightly dropped from 97.2 % to 94.5 % after [C<sub>12</sub>DMEA][IM] was reused for 3 cycles in the reaction of benzaldehyde and malononitrile.

In order to alleviate energy consumption and excessive cost during microalgae lipid extraction and separation, Tang and Row (2020) synthesized a novel CO<sub>2</sub>-induced switchable diisopropanolamine imidazole ([C<sub>6</sub>DIPA][IM]) with reversible hydrophobic-hydrophilic conversion. The microalgae lipid was extracted and dissolved in hydrophobic [C<sub>6</sub>DIPA][IM]. Upon CO<sub>2</sub> bubbling, the hydrophobic [C<sub>6</sub>DIPA][IM] was transformed into hydrophilic [C<sub>6</sub>DIPA][IM] and the lipids was separated with the aqueous phase ([C<sub>6</sub>DIPA][IM] + CO<sub>2</sub>/water). To recover the hydrophobic [C<sub>6</sub>DIPA][IM], the switching process can be achieved easily by removing CO<sub>2</sub> at 40 °C for 20 min. The [C<sub>6</sub>DIPA][IM] phase transition can be attributed to the interaction force between N-containing heterocyclic anion and CO<sub>2</sub>. The recovered [C<sub>6</sub>DIPA][IM] was then reused for 5 extractions, and it was found that the recycling ratio of [C<sub>6</sub>DIPA][IM] was >93.7 % for each cycle.

Moreover, supercritical fluids such as supercritical carbon dioxide (scCO<sub>2</sub>) are also considered green solvents and adopted in a range of processes, including extraction, due to their unique properties such as non-toxic, chemically inert, cheap, non-flammable, readily available,



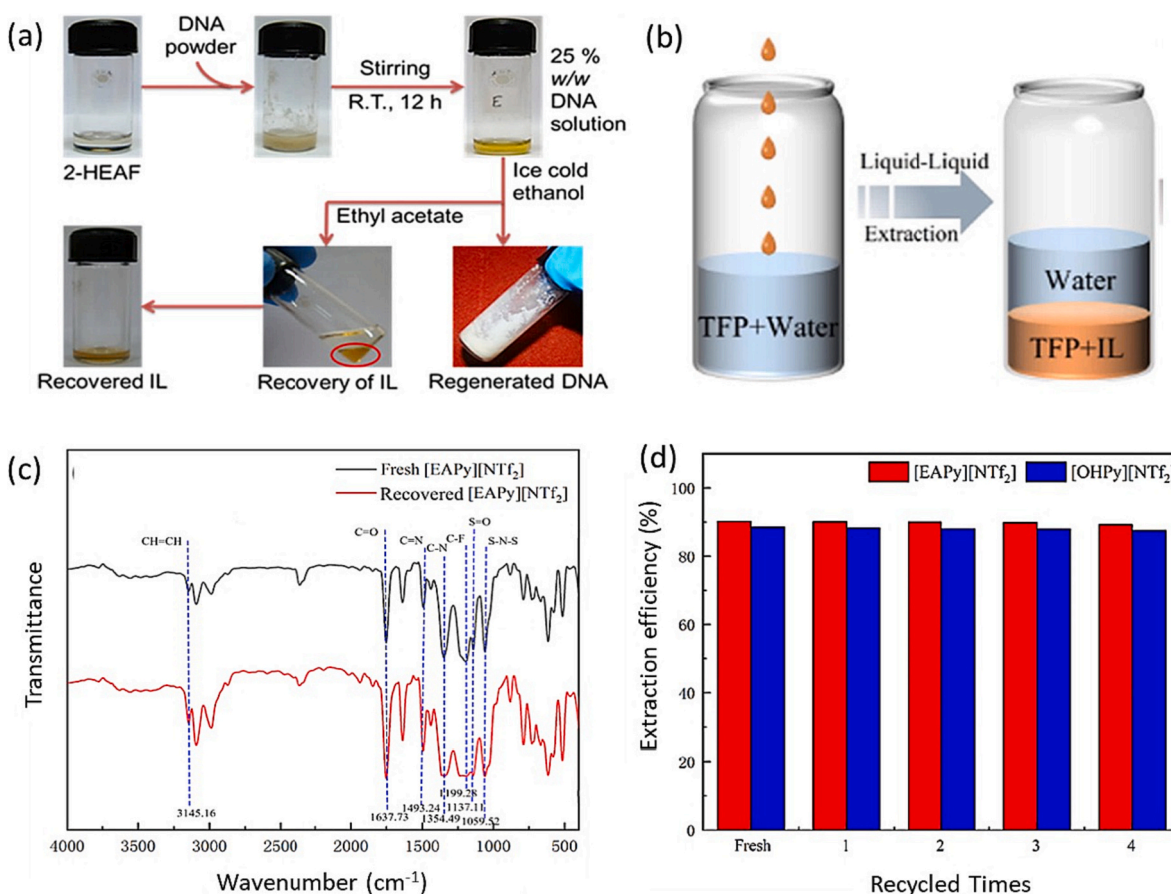


Fig. 6. (a) DNA dissolution and regeneration (Singh et al., 2017). (b) Schematic illustration of TFP extracted from water via [EAPy][NTF<sub>2</sub>] and [OHPy][NTF<sub>2</sub>], (c) FTIR spectra of fresh and recovered [EAPy][NTF<sub>2</sub>] and [OHPy][NTF<sub>2</sub>], (d) the extraction efficiency of recycled [EAPy][NTF<sub>2</sub>] and [OHPy][NTF<sub>2</sub>] (Peng et al., 2022).

and relatively low critical temperature and pressure (e.g. Pc = 73.8 bar; Tc = 31.0 °C) (Migowski and Lozano, 2023; Vladić et al., 2023). Also, the pure [C<sub>4</sub>MIM][PF<sub>6</sub>] is easy to be recovered after extraction when the dissolved CO<sub>2</sub> in the [C<sub>4</sub>MIM][PF<sub>6</sub>] phase was removed by depressurization. Blanchard and Brennecke (2001) investigated the removal of aromatic and aliphatic solutes (etc., benzene and hexane) from [C<sub>4</sub>MIM]

[PF<sub>6</sub>] by using scCO<sub>2</sub> at 40 °C and 138 bar. It was reported that low-polarity solutes gave higher solubility in CO<sub>2</sub>, while high-polarity and aromatic solutes gave higher affinity to the [C<sub>4</sub>MIM][PF<sub>6</sub>]-rich liquid phase. Overall, 95 % of the solutes were recovered from [C<sub>4</sub>MIM][PF<sub>6</sub>]. Table 3 summarized the ILs recovery findings via extraction process.

**Table 3**  
Summary of ionic liquids recovery findings via extraction process.

Ionic liquids (extractant)	Raffinate	Extracted component	Key Findings	Ref
[(CH <sub>2</sub> ) <sub>7</sub> COOHPIR][NTF <sub>2</sub> ]-[C <sub>4</sub> MIM][NTF <sub>2</sub> ]	Al(III)/Gd(III)	Al(III)	Extraction efficiency of [(CH <sub>2</sub> ) <sub>7</sub> COOHPIR][NTF <sub>2</sub> ]-[C <sub>4</sub> MIM][NTF <sub>2</sub> ] for Al(III) still >97.8 % after recycling for 8 cycles. 99.4 % of Al(III) removal was achieved after three-stage continuous extraction.	(Hu et al., 2022)
[C <sub>2</sub> MIM][PF <sub>6</sub> ]	Baicalin and baicalein	Baicalein	73.4 % of baicalein was yielded after using recycled [C <sub>2</sub> MIM][PF <sub>6</sub> ] at 4th cycles when compared to the fresh [C <sub>2</sub> MIM][PF <sub>6</sub> ].	(Wang et al., 2021a)
[C <sub>2</sub> MIM][FSI]	Water and 1000 mg/L phenolic compounds	Phenolic compounds	The regenerated [C <sub>2</sub> MIM][FSI] was reused for 3 times without any significant capacity loss. Phenolic extraction efficiency was at 90 %.	(Sas et al., 2018)
[EAPy][NTF <sub>2</sub> ]	Water and TFP	TFP	The recovered [EAPy][NTF <sub>2</sub> ] can maintain similar extraction efficiency compared to fresh [EAPy][NTF <sub>2</sub> ] after 4 runs at 90 %.	(Peng et al., 2022)
[diBTMG][BR]	Tea tree oil	Terpinen-4-ol	Terpinen-4-ol extraction rate decreased from 90 % to 87 % after the [diBTMG][BR] was reused for 7 cycles.	(Li et al., 2021a)
2-HEAF	-	Salmon DNA	93.11 % of 2-HEAF was recovered from DNA/2-HEAF solution using ethyl acetate.	(Singh et al., 2017)
[SUC][NTF <sub>2</sub> ] <sub>2</sub>	Metal ions solutions	Au(III)	The extraction efficiency decreased from >98 % to >87.6 % after using recycled [SUC][NTF <sub>2</sub> ] <sub>2</sub> for 5 runs.	(Wang et al., 2021b)
[C <sub>12</sub> DMEA][IM]	Aldehyde and malononitrile in n-pentanol/[C <sub>12</sub> DMEA][IM]/H <sub>2</sub> O solution	Benzaldehyde	Benzaldehyde yield decreased from 97.2 % to 94.5 % after [C <sub>12</sub> DMEA][IM] was reused for 3 cycles.	(Pei et al., 2018)
[C <sub>6</sub> DIPA][IM]	Wet microalgae	Lipid	Recycling ratio of [C <sub>6</sub> DIPA][IM] obtained after 5 cycles was >93.7 %.	(Tang and Ho Row, 2020)

## 2.4. Aqueous two-phase system (ATPS)

Aqueous two-phase system, also known as aqueous biphasic system, forms when two incompatible aqueous phases, such as IL/salt, salt/polymer (e.g., citrate, sodium sulphate, potassium phosphate and polyethylene glycol (PEG)) (Wu et al., 2017), polymer/polymer (e.g., PEG and dextran), are in contact with each other at a particular temperature above the critical concentration (Torres-Acosta et al., 2018). When compared to liquid-liquid extraction, in which hazardous and volatile chemicals are applied, ATPS is a much more sustainable and compatible method due to its non-toxicity and scalability. ATPS has been also widely used for biological production due to their low interfacial tension, high water content and relatively biocompatible environment for stabilizing cells (Ooi et al., 2011).

The partitioning of solute in one phase of the ATPS system can be affected by several parameters such as the constituent and concentration of the phases (Freire, 2016). Yang et al. (2018) used imidazolium chloride salts ([C<sub>n</sub>MIM][Cl]) as adjuvant to study the ATPS system composed of PEG and sodium salts. Parameters of the ATPS system such as type of salt, molecular weight of polymer and length of the [C<sub>n</sub>MIM][Cl] cationic carbon chain were evaluated. It was found that phase separation was enhanced when the molecular weight of PEG increased from 400 to 2000 g/mol. Also, trisodium citrate (Na<sub>3</sub>C<sub>6</sub>H<sub>5</sub>O<sub>7</sub>) showed strongest salting-out tendency due to C<sub>6</sub>H<sub>5</sub>O<sub>7</sub><sup>3-</sup> anions and relative ease to form ATPS. Moreover, it was reported that phase separation improved when the [C<sub>n</sub>MIM][Cl] alkyl side chain length was greater than six carbon atoms (etc., [C<sub>10</sub>MIM][Cl] and [C<sub>8</sub>MIM][Cl]), especially with PEG at higher molecular weight.

Wei et al. (2021) employed ATPS based on [C<sub>4</sub>MIM][Cl] and tripotassium phosphate (K<sub>3</sub>PO<sub>4</sub>) solution for rice straw (RS) pretreatment.

K<sub>3</sub>PO<sub>4</sub> solution acted as an antisolvent agent, and was able to precipitate the regenerated RS from the [C<sub>4</sub>MIM][Cl] phase to form a new phase in between [C<sub>4</sub>MIM][Cl] and K<sub>3</sub>PO<sub>4</sub> phases, as shown in Fig. 7(a). The recovered [C<sub>4</sub>MIM][Cl] was then recycled and reused up to 8 cycles to pretreat the RS. From the SEM image shown in Fig. 7(b), the pretreated rice straw using the 8th-cycle [C<sub>4</sub>MIM][Cl] showed more impurities accumulated on the RS sample, while firm and continuous surfaces were observed for RS using fresh [C<sub>4</sub>MIM][Cl]. As shown in Fig. 7(c), the yield of reducing sugar increased from 92.9 % to 105.2 % from the 1st to 7th recycles of [C<sub>4</sub>MIM][Cl], and then slightly decreased to 98.9 % after using 8th recycle due to the accumulation of impurities.

In addition, Li et al. (2010b) studied the 1-butyl-3-methylimidazolium tetrafluoroborate ([C<sub>4</sub>MIM][BF<sub>4</sub>]) recovery from water by using various kosmotropic salt (etc., trisodium phosphate (Na<sub>3</sub>PO<sub>4</sub>), sodium carbonate (Na<sub>2</sub>CO<sub>3</sub>), sodium sulphate (Na<sub>2</sub>SO<sub>4</sub>), monosodium phosphate (NaH<sub>2</sub>PO<sub>4</sub>), sodium chloride (NaCl)) at different mass fractions. Due to the salting-out effect, the [C<sub>4</sub>MIM][BF<sub>4</sub>] recovery efficiency was found to increase when the mass fraction of salts increased, giving 98.77 % when the mass fraction of Na<sub>2</sub>CO<sub>3</sub> was 0.1694. The recovery of [C<sub>4</sub>MIM][BF<sub>4</sub>] also rely on type of salts used. [C<sub>4</sub>MIM][BF<sub>4</sub>] recovery was the best in the presence of Na<sub>3</sub>PO<sub>4</sub>, followed by Na<sub>2</sub>CO<sub>3</sub>, Na<sub>2</sub>SO<sub>4</sub>, NaH<sub>2</sub>PO<sub>4</sub>, and finally NaCl. This can be attributed to the salt anions (i.e., PO<sub>4</sub><sup>3-</sup>, CO<sub>3</sub><sup>2-</sup>, SO<sub>4</sub><sup>2-</sup>, H<sub>2</sub>PO<sub>4</sub><sup>-</sup>, Cl<sup>-</sup>) that demonstrate strong interaction with water molecules and induce the salting-out effect for better [C<sub>4</sub>MIM][BF<sub>4</sub>] recovery efficiency.

The cation alkyl side chain and the functional groups attached to the cation also play an important role to create the ATPS system. Neves et al. (2019) separated the carbohydrate and antioxidants from a vanillin-rich pudding by using cholinium-derived N-ethyl-N,N,N-tris(2-hydroxyethyl) ammonium bistriflimide ([N<sub>2</sub>(2OH)(2OH)(2OH)] [NTF<sub>2</sub>]). The used

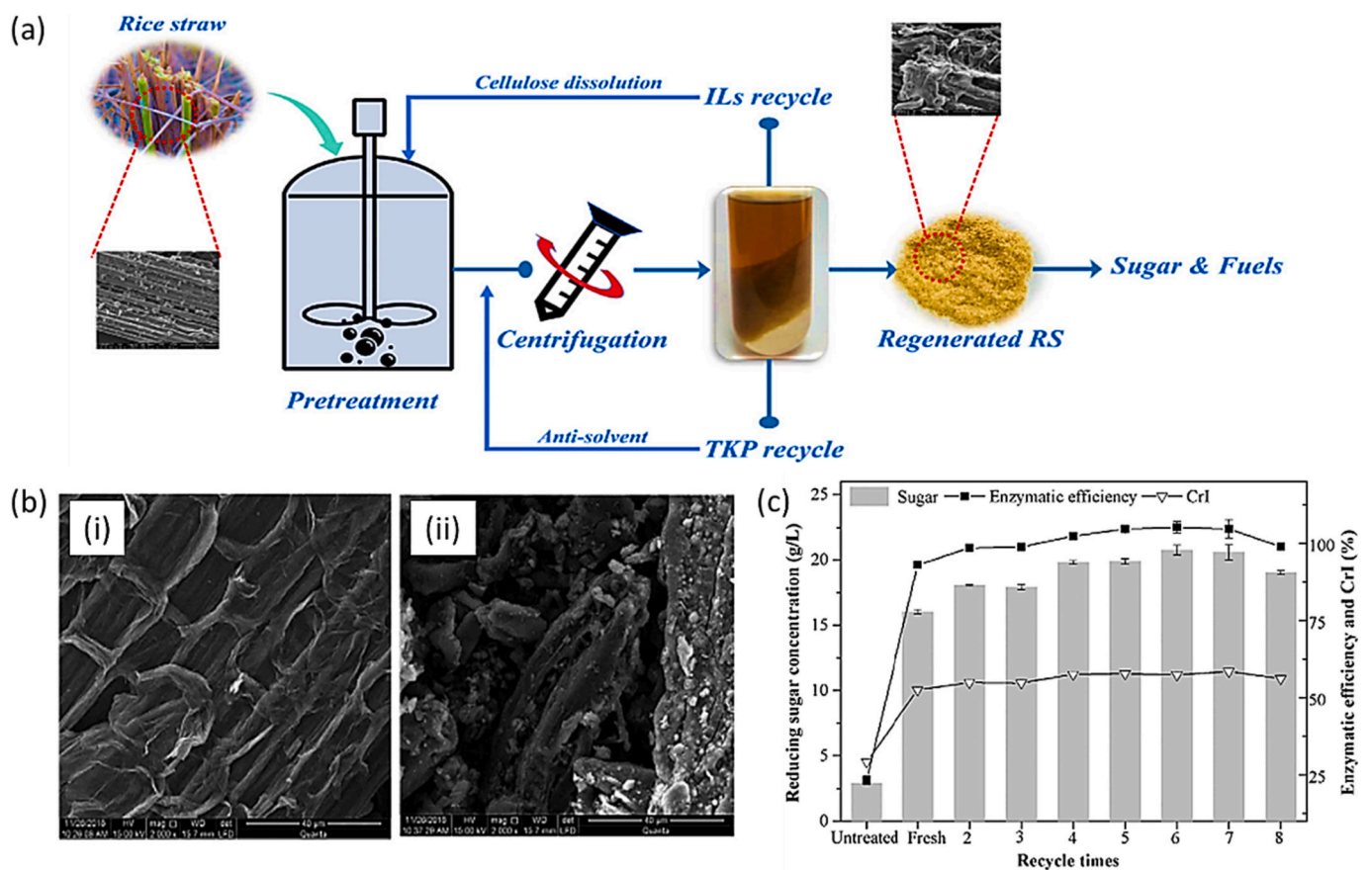


Fig. 7. (a) Schematic diagram of the [C<sub>4</sub>MIM][Cl] pretreatment and biorefinery process, (b) SEM images of rice straw pretreated with (i) fresh [C<sub>4</sub>MIM][Cl] and (ii) 8th recycle [C<sub>4</sub>MIM][Cl], (c) enzymatic hydrolysis performances for pretreated RS with [C<sub>4</sub>MIM][Cl] over multiple cycles (Wei et al., 2021).

$[N_2(2OH)(2OH)(2OH)][NTF_2]$  was recycled and reused for 3 cycles, with results indicating that 85 wt% of  $[N_2(2OH)(2OH)(2OH)][NTF_2]$  can be recovered for each recycling assay, and able to partition the antioxidant contents between 72 % to 82 % from carbohydrate due to the salting-out effect.

ATPS has also been employed in wastewater treatment plants to remove the pharmaceutical ingredients. Almeida et al. (2016) utilized IL-based ATPS composed of 1-ethyl-3-methylimidazolium triflate ( $[C_2MIM][CF_3SO_3]$ ) and salting-out agent, aluminium sulphate ( $Al_2(SO_4)_3$ ), in order to extract the fluoroquinolones (FQs). 40 wt% of  $K_3PO_4$  was added to the system to increase the pH to 7.2 to precipitate ciprofloxacin. The  $[C_2MIM][CF_3SO_3]$  recycling process involved only 2 steps, specifically, precipitation of FQs in the  $[C_2MIM][CF_3SO_3]$  phase by adding  $Al_2(SO_4)_3$  and water, followed by removing the FQs from  $[C_2MIM][CF_3SO_3]$  using vacuum filtration to recover the  $[C_2MIM][CF_3SO_3]$ -rich phase. It was reported that the recycled  $[C_2MIM][CF_3SO_3]$  was able to maintain outstanding extraction efficiencies of 91.2 %, 90.9 % and 95.3 % for ofloxacin, norfloxacin and ciprofloxacin, respectively, in four consecutive cycles. Also, the reusability of  $[C_2MIM][CF_3SO_3]$  was >97 wt% for the four consecutive cycles.

Similarly, Almeida et al. (2017) applied the ATPS system composed of  $Al_2(SO_4)_3$  salt and tributylmethyl-phosphonium methylsulphate  $[P_{4441}][CH_3SO_4]$  to separate nonsteroidal anti-inflammatory drugs (NSAIDs), such as diclofenac, ibuprofen, naproxen, and ketoprofen. After the NSAIDs was extracted to the  $[P_{4441}][CH_3SO_4]$  phase, water (which acted as an antisolvent) was introduced to precipitate NSAIDs, followed by vacuum filtration to obtain the precipitated NSAIDs. The recovered  $[P_{4441}][CH_3SO_4]$  was reused for 4 cycles and >94 wt% of  $[P_{4441}][CH_3SO_4]$  was recovered. Table 4 summarized the ILs recovery findings via ATPS.

## 2.5. Adsorption

Adsorption has been widely used as a robust method to recover ILs from aqueous solutions. To date, numerous adsorbents are known to recover ILs from the aqueous solution, such as activated carbons (AC), ion exchange resins, biosorbents, etc. (Anthony et al., 2001; Farooq et al., 2012; Hassan et al., 2014; Lemus et al., 2012, 2017; Palomar et al., 2009; Sui et al., 2018; Ushiki et al., 2017). The recovery of  $[C_4MIM][PF_6]$  from wastewater was first proposed by Anthony et al. (2001) by using AC. Both anions and cations were removed from the aqueous solution, but AC exhibited a lower efficiency for removing  $[C_4MIM][PF_6]$ . Compared to toluene removal, the amount of AC needed for  $[C_4MIM][PF_6]$  was >50 times more. The results indicate that AC gave better removal efficiency for small nonpolar compounds than for polar or ionic compounds.

**Table 4**  
Summary of ionic liquids recovery findings via aqueous two-phase system.

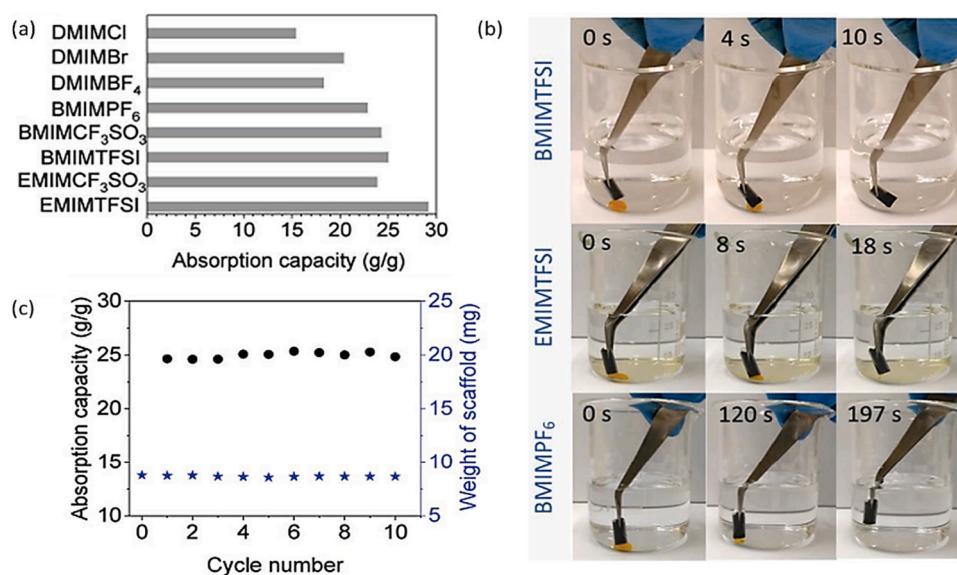
Ionic liquid	Salt used	Product or extractant	Key findings	Ref
$[C_4MIM][Cl]$	$K_3PO_4$	Reducing sugar	98.9 % of reducing sugar yielded after the $[C_4MIM][Cl]$ was recycled and run for 8 cycles.	(Wei et al., 2021)
$[P_{4448}][BR]$	$K_3PO_4$	Astaxanthin	The $[P_{4448}][BR]$ recovery and extraction efficiency of astaxanthin remained at >90 % after recycling over three times.	(Gao et al., 2020b)
$[N_2(2OH)(2OH)(2OH)][NTF_2]$	Carbohydrates	Antioxidant	85 wt% of $[N_2(2OH)(2OH)(2OH)][NTF_2]$ was recovered in each recycling assay. IL was recycled two times with the same separation performance.	(Neves et al., 2019)
$[P_{4441}][CH_3SO_4]$	$Al_2(SO_4)_3$	NSAIDs	The extraction efficiency for the NSAIDs remained almost similar after $[P_{4441}][CH_3SO_4]$ was recycled and reused in four consecutive cycles (>99 %).	(Almeida et al., 2017)
$[C_2MIM][CF_3SO_3]$	$K_3PO_4$ and $Al_2(SO_4)_3$	FQs	The extraction efficiency of FQs was >90 % when using recycled $[C_2MIM][CF_3SO_3]$ for 4 cycles, with no losses compared to fresh $[C_2MIM][CF_3SO_3]$ .	(Almeida et al., 2016)
$[C_4MIM][ACE]$	$Na_2CO_3$	Glauicine	90 % of $[C_4MIM][ACE]$ was recovered.	(Keremedchieva et al., 2015)
$[C_4MIM][TFO]$	$NaH_2PO_4$	Lactoferrin	>99 % of $[C_4MIM][TFO]$ can be recycled.	(Alvarez-Guerra et al., 2014)
$[C_2MIM][AC]$	$K_3PO_4$	Miscanthus	Recycled $[C_2MIM][AC]$ took 48 h for full cellulose conversion while fresh $[C_2MIM][AC]$ required <25 h for complete cellulose conversion.	(Shill et al., 2011)

In addition, the hydrophobicity and hydrophilicity nature of IL as well as the AC surface chemistry play a significant role in affecting the adsorption capacity. The adsorption of hydrophobic ILs onto AC was considered reasonable. As for hydrophilic ILs, adsorption could be improved by enhancing the surface hydroxyl groups of AC. When the hydroxyl group on the adsorbent increases, the hydrogen bonding interactions with the hydrophilic ILs increases and thus improves the adsorption performance. However, the functionalization of AC to enhance its adsorption affinity to ILs might be costly. Therefore, Shi et al. (2016) introduced straw- and wood-based biochars to enhance the adsorption efficiency for the removal of  $[C_2MIM][BF_4]$  from wastewater. The typical biochar contained less carbonized structure and more surface polar functional groups than AC (Azargohar and Dalai, 2006; Qi et al., 2013); therefore, it can be used for the adsorption of polar ionic compounds. Overall, both the straw- and wood-based biochars exhibited excellent  $[C_2MIM][BF_4]$  adsorption efficiency due to the abundant oxygen-containing groups and also high percentage of micropores. More importantly, the adsorbed  $[C_2MIM][BF_4]$  can be desorbed upon the introduction of HCl solution at a pH of 0.5, and the biochars can be reused up to 10 adsorption-desorption cycles without jeopardizing the adsorption efficiency.

Guan et al. (2019) developed a superhydrophobic 3D aerogel composed of carbon nanofibers (CNFs) and chitosan modified with 1,6-hexamethylene diisocyanate to absorb various types of ILs. As presented in Fig. 8(a), the CNF-based 3D aerogels demonstrated superior ILs adsorption capacity ranging from 15 to 30 g/g, which is higher than that of AC (0.18 to 0.46 g/g). As shown in Fig. 8(b), the adsorption of ILs occurred promptly within a few seconds to hundreds of seconds depending on the viscosity of ILs (etc.,  $[C_4MIM][NTF_2]$  = 56.4 cP,  $[C_2MIM][NTF_2]$  = 70 cP,  $[C_4MIM][PF_6]$  = 263.7 cP). After the adsorption process, acetone was applied to wash out ILs from the CNF-based 3D aerogels, which were reused up to 10 cycles at the same capacity for  $[C_4MIM][NTF_2]$  (Fig. 8(c)).

Sui et al. (2018) ascertained the adsorption and desorption of  $[C_4MIM][BF_4]$  and  $[C_2MIM][BF_4]$  from aqueous solution using sulfonic acid cation-exchange resin (Amberlite IR 120Na). When the ion-exchange resin was added to the  $[C_2MIM][BF_4]$  solution, the  $H^+$  of the resin sulfonic group exchanges with the  $[C_2MIM][BF_4]$  cationic part, thus enabling the  $[C_2MIM][BF_4]$  cations to adsorb to the ion-exchange resin. Overall, up to 95 % and 90 % of  $[C_4MIM][BF_4]$  and  $[C_2MIM][BF_4]$ , respectively, were adsorbed by the resins after subjected to 20 °C for 30 min. When the  $[C_2MIM][BF_4]$ -loaded ion-exchange resin was in contact with 15 wt% of HCl, the  $H^+$  of HCl replaces the  $[C_2MIM][BF_4]$  cations that adsorbed onto the resin. Consequently, the ion-exchange resin can be regenerated and returned to its original acidic form. The reutilization of regenerated ion-exchange resin was investigated in three





**Fig. 8.** (a) CNF-based 3D aerogels absorption capacity towards different ILs, (b) CNF-based 3D aerogel (diameter = 4.5 mm, length = 11 mm) adsorption performance to different type of ILs as a function of time, (c) absorption capacity of re-used CNF-based 3D aerogels for [C<sub>4</sub>MIM][NTF<sub>2</sub>] (Guan et al., 2019).

adsorption cycles, and it was found that the adsorption performance slightly declined. Nonetheless, the adsorption ratio of [C<sub>4</sub>MIM][BF<sub>4</sub>] still remained over 90 %.

He et al. (2017) utilized ion-exchange resins to recover benzothiazolium triflate [HBTH][CF<sub>3</sub>SO<sub>3</sub>] from coexisting glucose. It was reported that the gel-type ion-exchange resin with H<sup>+</sup> ionic form (732(H)) exhibited higher [HBTH][CF<sub>3</sub>SO<sub>3</sub>] adsorption efficiency than macroporous and Na<sup>+</sup>-ions formed ion-exchange resins. Moreover, an acidic HCl-EtOH-H<sub>2</sub>O mixture (18:8:74, V/V/V) was employed in this study for the desorption process to recover the adsorbed [HBTH][CF<sub>3</sub>SO<sub>3</sub>] and reuse the resin. Consequently, about 98.60 % of [HBTH][CF<sub>3</sub>SO<sub>3</sub>] desorption was achieved. The desorbed resin was further reused for 5 cycles, through which the [HBTH][CF<sub>3</sub>SO<sub>3</sub>] adsorption deteriorated from 98.6 % to 79.4 %.

Lemus et al. (2012) carried out an extensive analysis of the separation of 1-octyl-3-methylimidazolium hexafluorophosphate ([C<sub>8</sub>MIM][PF<sub>6</sub>]) from aqueous solution using commercial and chemical-treated AC. The separation of [C<sub>8</sub>MIM][PF<sub>6</sub>] was strongly dependent on the chemical and physical properties of both adsorbent and solute. It was reported that narrow mesoporous or microporous AC with high densities of pores (diameter <8 nm) exhibited the highest adsorption capacities.

Due to the non-polar nature of AC, the increasing hydrophobicity of [C<sub>8</sub>MIM][PF<sub>6</sub>] implies higher affinity. Acetone was used to regenerate AC after [C<sub>8</sub>MIM][PF<sub>6</sub>] adsorption and <sup>1</sup>H NMR analysis was carried out to evaluate the recovered [C<sub>8</sub>MIM][PF<sub>6</sub>].

In addition, Yu et al. (2016) demonstrated a novel biosorbents-based corncob-cellulose nanocrystals for the removal of [C<sub>4</sub>MIM][Cl] from aqueous solution. The surface of epoxy cellulose nanocrystals (CNG) was further modified with diethylenetriaminepentaacetic acid and sulfosalicylic acid, termed CNGD and CNGS, respectively. CNGS showed more favourable [C<sub>4</sub>MIM][Cl] adsorption (0.499 mmol/g at 25 °C) than CNGD (0.473 mmol/g at 65 °C) per the Langmuir model. This can be possibly due to the π-π stacking interactions between CNGS and [C<sub>4</sub>MIM][Cl]. Also, ethanol was used for the desorption of [C<sub>4</sub>MIM][Cl] from CNGS and CNGD. Overall, CNGD and CNGS can be regenerated effectively and reused for another 5 runs without significant adsorption capacity reduction. Table 5 summarized the ILs recovery findings via adsorption method.

**Table 5**  
Summary of ionic liquids recovery findings via adsorption process.

Ionic liquids	Adsorbents	Adsorbate	Key Findings	Ref
[C <sub>4</sub> MIM][BF <sub>4</sub> ]	SWCNT	Acteoside	ACT was separated from [C <sub>4</sub> MIM][BF <sub>4</sub> ] via SWCNT. The separation of ACT from [C <sub>4</sub> MIM][BF <sub>4</sub> ] can reach >95 %.	(Xu et al., 2021)
[C <sub>4</sub> MIM][NTF <sub>2</sub> ]	CNF-based 3D aerogel	[C <sub>4</sub> MIM][NTF <sub>2</sub> ]	[C <sub>4</sub> MIM][NTF <sub>2</sub> ] was absorbed by CNF-3D aerogel and the absorption capacity was maintained at around 24 g/g after 10 cycles. 99.5 % separation efficiency was obtained by 22 mm - long CNF-based 3D aerogels.	(Guan et al., 2019)
[C <sub>4</sub> MIM][BF <sub>4</sub> ]	Ion-exchange resin	[C <sub>4</sub> MIM][BF <sub>4</sub> ]	95 % of [C <sub>4</sub> MIM][BF <sub>4</sub> ] can be adsorbed by the resins.	(Sui et al., 2018)
[C <sub>2</sub> MIM][BF <sub>4</sub> ]	Ion-exchange resin	[C <sub>2</sub> MIM][BF <sub>4</sub> ]	90 % of [C <sub>2</sub> MIM][BF <sub>4</sub> ] can be adsorbed by the resins.	(Sui et al., 2018)
[HBTH][CF <sub>3</sub> SO <sub>3</sub> ]	732 cation exchange resin	[HBTH][CF <sub>3</sub> SO <sub>3</sub> ]	The adsorption amount of [HBTH][CF <sub>3</sub> SO <sub>3</sub> ] using 732 resin is >300 mg/g. 732 resin maintained the adsorption efficiency (>93.4 %) for [HBTH][CF <sub>3</sub> SO <sub>3</sub> ] after reused for 3 cycles	(He et al., 2017)
[C <sub>4</sub> MIM][Cl]	CNGS	[C <sub>4</sub> MIM][Cl]	The [C <sub>4</sub> MIM][Cl] adsorption capacity using CNGS reached 0.499 mmol/g. CNGS can be reused at least 5 times to adsorb [C <sub>4</sub> MIM][Cl].	(Yu et al., 2016)
[C <sub>4</sub> MIM][Cl]	CM <sub>600-2-2</sub>	[C <sub>4</sub> MIM][Cl]	The [C <sub>4</sub> MIM][Cl] adsorption capacity reached 0.183 mmol/g. Adsorption capacity of CM <sub>600-2-2</sub> reduced to 0.135 mmol/g after reused for 3 cycles.	(Wang et al., 2015)
[C <sub>2</sub> MIM][BF <sub>4</sub> ]	SBB	[C <sub>2</sub> MIM][BF <sub>4</sub> ]	Adsorbents can be reused up to ten adsorption-desorption cycles without losing adsorption efficiency.	(Shi et al., 2016)

### 3. Advantages and disadvantages of different ILs recovery methods

The recovery and recycling of ILs play a significant role in the commercialization of applicable technologies, and it is important too for environmental conservation as well as effective resources consumption. Due to the different natures of ILs (e.g., hydrophilic and hydrophobic), different recovery and purification technologies, such as distillation, membrane technology, adsorption, extraction, ATPS system, etc. are required. Accordingly, it is vital to understand the advantages and disadvantages of the different recovery techniques for ILs (see Table 6).

### 4. Ionic liquid degradation

The IL degradation possibility is evaluated in this section. It is worth noting that although ILs are generally considered to be thermally stable, the IL stability can be hindered by IL thermal degradation and impurities introduced during the IL regeneration process (Li et al., 2010a; Shi et al., 2013; Xu et al., 2017). For instance, Xu et al. (2017) reported that the anionic structure of the 4th recycled [C<sub>4</sub>MIM][AC] after evaporation recovery was unrepairable or destroyed as the number of biomass pretreatment cycles increased, with eucalyptus hydrolysis rate of 72.8 %.

**Table 6**  
Advantages and limitations of different IL recovery methods.

Methods	Advantages	Disadvantages
Distillation	<ul style="list-style-type: none"> <li>Simple method (Mai et al., 2014)</li> <li>Safe process and can be carried out without the addition of toxic agents (de Jesus and Maciel Filho, 2022)</li> <li>Suitable for recover high concentration ILs (Zhou et al., 2018)</li> </ul>	<ul style="list-style-type: none"> <li>IL can be thermally decomposed during distillation (Earle et al., 2006)</li> <li>Energy-intensive process (de Jesus and Maciel Filho, 2022)</li> <li>Emission of volatile organic compounds to the atmosphere through leakage losses (Jessop, 2011)</li> </ul>
Membrane	<ul style="list-style-type: none"> <li>Eco-friendly and energy-saving (Li et al., 2021b)</li> <li>Large-scale separation application (Othman et al., 2022)</li> <li>Simple operation (Song et al., 2022)</li> <li>Cost effective (Kavitha et al., 2022; Zhang et al., 2015)</li> </ul>	<ul style="list-style-type: none"> <li>Molecular diffusion deteriorated for high viscosity ILs (Kuzmina, 2016; Sanni et al., 2022)</li> </ul>
Extraction	<ul style="list-style-type: none"> <li>Simple operating method (involve organic and aqueous solution conventionally)</li> <li>Suitable to extract non-volatile or thermal sensitive species (Mai et al., 2014)</li> <li>Energy-efficient technology (Zhao et al., 2005)</li> </ul>	<ul style="list-style-type: none"> <li>Environmental pollution (consume large amount of hazardous, flammable and volatile organic compounds (VOCs) (Nasirpour et al., 2020)</li> <li>Disposal of diluents and spent extractants will cause further destructive damage (Huddleston et al., 1998)</li> </ul>
Aqueous two-phase extraction	<ul style="list-style-type: none"> <li>Non-toxic system (Tang et al., 2014)</li> <li>Large scale production (Yau et al., 2015)</li> <li>Widely used for separation of biological products and low molecular weight compounds</li> </ul>	<ul style="list-style-type: none"> <li>Expensive for commercial scale (Sanford et al., 2016)</li> <li>High salt concentrations needed (Torres-Acosta et al., 2018)</li> </ul>
Adsorption	<ul style="list-style-type: none"> <li>Non-destructive mechanism (Guan et al., 2019)</li> <li>Eco-friendly method (Sui et al., 2018)</li> <li>Cost-effective (Sui et al., 2018)</li> <li>Recover IL of different nature (Lemus et al., 2017)</li> </ul>	<ul style="list-style-type: none"> <li>Slow IL diffusion during adsorption (Guan et al., 2019)</li> <li>Subsequent reutilization (Guan et al., 2019)</li> <li>Inability to recover the anions of the original ionic liquid (Corley and Iacono, 2019)</li> </ul>

This phenomenon can be attributed to the contamination of [C<sub>4</sub>MIM][AC] by the water and lignin residue during pretreatment. Other than that, Shi et al. (2013) observed that after the [C<sub>2</sub>MIM][AC] was recovered via filtration and vacuum evaporation, impurities (lignin or sugar residues contained in the recycled [C<sub>2</sub>MIM][AC]) were detected. When compared with the pristine [C<sub>2</sub>MIM][AC], the reused [C<sub>2</sub>MIM][AC] showed slight decrease in the yield of glucose (6.5 % reduction) and xylose (10.8 % reduction). Despite the operational simplicity of distillation, it is unable to remove those non-volatile impurities (e.g., inorganic ions) in the IL, and this might affect the quality of the recycled IL (Pan et al., 2024). Furthermore, the IL thermal stability can be affected by the alkyl chain length of IL itself. It is reported that the bond between the cation and alkyl chain such as ammonium and imidazolium can be weakened with increasing alkyl chain length, causing it to be more vulnerable to attack and therefore more likely thermally decomposed (Xu and Cheng, 2021). For membrane recovery, Pan et al. (2024) utilized a recycler consisting of extraction and IL purification steps to remove the impurities and recover the desired IL, respectively. During the second step, the ILM membrane (membrane prewet by hydrophobic IL) in the recycler was used to recover 1-butyl-1-methylpyrrolidinium triflate [BMPYRR][CF<sub>3</sub>SO<sub>3</sub>] after platinum nanoparticles synthesis. It was reported that some of the impurities such as ethylene glycol and platinum (518 ppm) persisted in the recycled [BMPYRR][CF<sub>3</sub>SO<sub>3</sub>]. The particle accumulation can jeopardize the membrane separation efficiency of the used [BMPYRR][CF<sub>3</sub>SO<sub>3</sub>], leading to degradation of the quality of recycled [BMPYRR][CF<sub>3</sub>SO<sub>3</sub>]. Moreover, the water content in the phosphonium chloride [TBHPP][Cl] can affect the removal efficiency of dibenzothiophene during the extractive desulfurization process (Moghadam et al., 2017). When the water content increased from 0 to 5 %, the dibenzothiophene extraction efficiency was reduced from 68 % to 59 %. After 5 cycles of dibenzothiophene extraction, [TBHPP][Cl] was saturated and lost its ability in the desulfurization process. For [C<sub>2</sub>MIM][AC] recycling via ATPS process, it must be pointed out that despite the addition of K<sub>3</sub>PO<sub>4</sub> being able to induce a phase separation of [C<sub>2</sub>MIM][AC] and desired cellulose (product), the basicity of the K<sub>3</sub>PO<sub>4</sub> solution can lead to the partial cleavage of lignin during the biomass pretreatment process, allowing it to remain soluble in [C<sub>2</sub>MIM][AC] (Shill et al., 2011). However, the impurities such as lignin after the biomass pretreatment have to be removed in order to prolong the lifetime of the recycled [C<sub>2</sub>MIM][AC]. With regards to the adsorption method, Sui et al. (2018) utilized the sulfonic acid cation-exchange resin to regenerate [C<sub>4</sub>MIM][BF<sub>4</sub>] after the extraction of oil sand. However, the existence of natural ions (e.g., CaCl<sub>2</sub>) in [C<sub>4</sub>MIM][BF<sub>4</sub>] during the washing step after oil sand extraction deteriorated the adsorption efficiency of the resin, with the adsorption ratio of [C<sub>4</sub>MIM][BF<sub>4</sub>] decreasing from 95.32 % to 79.13 % when the Ca<sup>2+</sup> ions elevated from 0 to 1000 ppm. The suppressed adsorption can be attributed to the strong electrostatic attraction of Ca<sup>2+</sup> towards ion-exchange resins, which compete against the [C<sub>4</sub>MIM] cations for binding sites. Overall, the IL recycling quality can be greatly affected by the impurities during the reactions. Therefore, it is recommended to filter out the impurities prior to IL recycling to ensure the purity of the regenerated IL.

### 5. Challenges and future perspective for ILs recovery

Distillation is a mature and relatively simple process without the need for toxic agents. It uses heat to vaporize volatile solvents or water, and non-volatile ILs are left as residues. It is also suitable to recover high concentration ILs solution to achieve high purity level. However, distillation is energy-intensive and thus increases the operating costs. Moreover, thermal decomposition may possibly occur. To address the challenges associated with distillation, membrane-based separation is considered as a simple, cost effective, eco-friendly and energy-efficient method which has been used to recover ILs. The pressure-driven membranes such as NF and RO have been used to separate both hydrophilic and hydrophobic ILs, which can be recovered in either the feed or

permeate side. However, the membrane flux can be diminished due to the inevitable concentration polarization or fouling. Also, the high-viscosity ILs could lead to flux deterioration. Notably, concentrated ILs can be obtained but not pure IL. Therefore, it is necessary to combine with post-treatments such as distillation or adsorption to further concentrate the ILs. Extraction is an energy-efficient and simple technology that is promising for extracting non-volatile or thermal-sensitive ILs. Organic solvents have been commonly used in the extraction system to separate either hydrophilic or hydrophobic ILs. However, the use of hazardous organic solvents could lead to cross contamination and adverse environmental impact. To this end, aqueous two phase system (ATPS) is an environmentally friendly method that is non-toxic, in which organic solvents are not needed to recover hydrophilic ILs from aqueous solutions. By adding the inorganic salts or carbohydrates, the salt-out mechanism is capable of inducing the phase separation of ILs and water. However, some inorganic ions are retained in the aqueous solution. Regarding the adsorption method, the possibility for adsorption and desorption renders it an environmental friendly and effective way to recover ILs. This method is suitable to recover low-concentration or diluted ILs. However, the recovery of ILs is dependent on the physical and chemical properties of adsorbents, such as pore size and functional groups. For instance, activated carbon (AC) is favourable for hydrophobic ILs, while ion exchange resin attracts the cations but not the anions of ILs (Zhou et al., 2018).

As opposed to the traditional solvent (e.g., volatile organic compounds), ILs are more environmentally friendly (Forsyth et al., 2004). Despite the fact that ILs are designated 'green' solvents, some of the anions such as  $[SbF_6]$ ,  $[BF_4]$ , and  $[PF_6]$  can hydrolyzed into water fluoride forms, which are toxic to aquatic organisms (Cho et al., 2021). Recent studies have also revealed that the cations and structure of ILs can influence toxicity (Cho et al., 2021; Fan et al., 2019; Radošević et al., 2013). Therefore, to enhance the practical feasibility of ILs, research efforts should be intensified to address these problems by introducing biodegradable ILs such as biochar, or plant-based adsorbents such as corncob-cellulose nanocrystals, as aforementioned. A life cycle assessment (LCA) of ILs is essential to analyze the environmental impact and performance of IL-based processes, as well as weigh the improvements possible by IL usage. On top of the well-reported recovery techniques for ILs, solar evaporation may be a potential energy-efficient technology that has not yet been much explored.

## 6. Economy feasibility of IL recycling methods

Despite ILs possessing many advantages such as negligible vapor pressure, non-flammable, and having high thermal stability, the major issue in utilizing and scaling the ILs lies in the high cost, which exceeds more than \$800/kg (Tullo, 2020), resulting in many industrial-scale applications being untenable. Since the commercially available ILs are more expensive than the traditional organic solvents, IL recycling and reuse seems to be the most promising method for more economically feasible large-scale applications. For a practical process, the ILs should be recovered >80 % or 90 %, or more preferably >99 % from the reaction mixture. Numerous IL recycling techniques, such as distillation, membrane separation, ATPS, extraction and adsorption, have been introduced to recycle ILs. All of these ILs recovery methods have their own pros and cons. For instance, membrane separation is an energy efficient and space-efficient method with low capital cost to recover ILs, especially via nanofiltration membranes (Sosa et al., 2023a). Nevertheless, large amounts of permeate demands could restrict its application in industry due to the flux limitation (Chen et al., 2023). Currently, extraction and distillation are the most commonly adopted separation methods for the IL recovery process at both industrial- and lab-scales (Chen et al., 2023; Zhou et al., 2018). However, distillation is energy-intensive and costly on the commercial scale (Liu et al., 2010). In terms of extraction, it requires the use of particular apparatus and organic solvents, which not only hinders the commercial-scale

implementation, but also generates undesirable solvent waste (Liu et al., 2010). In order to obtain a cost-effective IL recycling process, Chen et al. (2023) introduced a practical and cost-effective hybrid recycling process that composed of ATPS, membrane separation and distillation methods to recover 10 wt% n-butylpyridinium trifluoromethanesulfonate  $[C_4PY][TFO]$ . Interestingly, the hybrid process with the use of  $[NH_4]_2SO_4$  salting-out agent to recycle 10 wt%  $[C_4PY][TFO]$  could reduce the energy consumption and total annual cost by 91 % and 57 %, respectively, compared to distillation. This indicates that hybrid technologies for IL recycling is promising for industry implementation.

## 7. Conclusion

This present review details the various recovery and recycling techniques of ILs reported in the past decades, as well as the challenges and future perspectives of ILs recovery. Although ILs are known as "designer solvents", the toxicity and biodegradable level of ILs should be taken into account rigorously as some hazardous ILs might threaten the environment especially aquatic life. Overall, the ILs recovery and purification is vital for industrial applications. Membrane-based technology seems to be a promising method to recover ILs as commercial membranes such as NF270 and NF90 showed reasonable IL separation and purification. Perhaps more efforts should be directed to membrane modification to enhance the recovery efficiency of ILs.

## CRediT authorship contribution statement

**Ying Siew Khoo:** Conceptualization, Formal analysis, Writing – original draft. **Tommy Chandra Tjong:** Writing – original draft. **Jia Wei Chew:** Project administration, Supervision, Writing – review & editing. **Xiao Hu:** Project administration, Supervision, Validation, Writing – review & editing.

## Declaration of competing interest

The authors declare no conflicts of interest.

## Data availability

The data that has been used is confidential.

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