



Selective recovery of rare earth elements from e-waste via ionic liquid extraction: A review

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ABSTRACT

Increasing generation of electronic waste (e-waste) is a major concern for emerging rare earth elements (REEs) pollution if not appropriately disposed of. Nevertheless, e-waste could be considered as a secondary source for critical REEs mining. Under the context of circular economy, this review focuses on selective recovery of REEs from e-waste using ionic liquid (IL) extraction. ILs demonstrate a sustainable alternative to organic volatiles used in traditional solvent extraction process. Despite insufficient literature on applications of ILs for REEs extraction from limited categories of real e-waste, the review comprehensively summarizes the latest studies on recovery of REEs from e-waste (mostly uncontaminated) using IL extraction and the existing data indicate encouraging outcomes in terms of extractability and selectivity. Roles of ILs in REEs recovery have been critically analyzed. The review has systematically illustrated the fundamental design of ILs for selective extraction of REEs, including bifunctional and task-specific ILs, incorporation of dilutant/extractant, non-fluorinated ILs, etc. This fundamental knowledge could greatly facilitate cost-effective and selective extraction of REEs from e-waste. ILs must be completely assessed in terms of extractability, selectivity, and reusability. Synergistic extraction based on quaternary phosphonium salts are discussed to realize excellent extraction of REEs into IL phase leaving non-REEs in the aqueous phase for easier separation. Overall, regeneration of spent ILs and subsequent separation of extracted REEs from ILs are also crucial to selective recovery of REEs from e-waste using novel ILs. Nevertheless, industrialization of this technology would require more fundamental data on extraction efficiency and recovery rate of REEs from real e-waste.

1. Introduction

Selective recovery of rare earth elements (REEs) from natural ores and secondary resources is essential for wide applications, especially for green energy transition. Due to distinct advantages over chemical precipitation and ion exchange, solvent extraction (SX) has been developed as the predominant separation technology to obtain high-purity forms of REEs. Nevertheless, SX still has some drawbacks in terms of adverse effects on human health and surrounding environment resulting from the use of highly volatile, toxic, and flammable organic solvents (e.g., extractants and diluents). It is interesting to note that ionic liquids (ILs), which are not only environmentally benign but also can dramatically increase extraction efficiency, have been regarded as a prospective “greener alternative” to conventional molecular solvents used for REEs

extraction via SX.

1.1. Mining of electronic waste

Currently, there is a progressive shift towards circular economy (CE) and researchers have been focusing on promoting sustainable reuse and recycling of waste streams [1]. Precious and valuable metals could be recovered from these waste materials if appropriate extraction and management are implemented. Being alternative source to natural ores, waste materials demonstrate great potential to achieve additional economic and sustainable growth [2]. Electronic waste (e-waste) is one of the largest and fastest-growing waste streams worldwide with a total generation of ca. 45 million tons in 2014 and an annual growth rate of 3–5 % thereafter (Fig. S1) [3]. In the European union (EU), Waste

Abbreviations: Bif-ILs, Bifunctional ionic liquids; CE, Circular economy; DES, Deep eutectic solvents; EOL, End-of-life; EU, European union; HBD, Hydrogen bond donor; HSAB, Hard–Soft Acid–Base; HREs, Heavy rare earth elements; IL-ABSs, IL-based aqueous biphasic systems; ILs, Ionic liquids; LREs, Light rare earth elements; REEs, Rare earth elements; SX, Solvent extraction; TSILs, Task-specific ILs; VOCs, Volatile organic compounds; WEEE, Waste electrical and electronic equipment.

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Table 1

Six categories of e-waste according to EU WEEE (Directive 2012/19/EU).

No.	Category	Examples
1	Temperature Exchange Equipment	Refrigerators & Freezers, Air conditioning equipments
2	Screens and Monitors	CRT monitors, Flat Panel Displays
3	Lamps	LED, Fluorescent tubes
4	Large Equipment	Dishwashers, Ovens, Washing machines
5	Small Equipment	Vacuum cleaners, Hi-Fi equipments, Kitchen appliances, power tools
6	Small IT and Telecommunication Equipment	Printers, other small IT, Phones, computers & Laptops

electrical and electronic equipment (WEEE) Directive and revised CE performance are important policy tools to make provision for this shift in EEE sector [4]. Introduced in 2003, WEEE Directive was later amended in 2012 to account the rapid growth of EEE markets and reduce cycles of innovation [5]. This directive followed the principle of waste disposal and prioritized e-waste prevention, after which it could be reused and recycled (Fig. S2) [5]. Finland, as an EU member state, is also actively involved in promoting e-waste recovery and the recycling rate of e-waste has increased steadily since 2010, from 28.7 % to 49.2 % (Fig. S3) [6]. Overall, e-waste could be designated to any waste which falls under the dominion of European WEEE directive (Directive 2012/19/EU) and can be classified into six categories (Table 1). Different disposal techniques are applicable according to materials composition and quality of e-waste, with reusing and recycling as materials being the predominant manner. The raw material group has a substantial recycling potential, where critical and valuable elements could be recovered with an estimated value up to 55 billion euros [7]. Due to higher concentrations of precious metals, including rare earth elements (REEs), than those present in primary ores, e-waste could be regarded as secondary ores. Furthermore, the cost of mining from ores could be 13 times higher than that of e-waste recycling [8]. Thus, recycling could be more lucrative as compared to traditional mining. Other advantages include less demand of natural ores, as well as less adverse environmental effect resulting from e-waste, dangerous substances and water usage [9]. Nevertheless, although REEs are among the most critical elements for modern technologies, their commercial recycling is still limited. Only ca. 1 % of REEs are recycled from end-of-life (EOL) waste, the rest is wasted and removed from the cycle of materials [8]. Due to inefficient collection, technological difficulty, and lack of incentives, their recycling rates are relatively low. The recycling rate of REEs, however, is much higher for some categories of e-waste with more efficient collection and easier processing.

1.2. Rare earth elements: Sources and uses

The 17 elements known as REEs are metals with similar properties, such as Scandium (Sc), Yttrium (Y), Samarium (Sm), Terbium (Tb) and all lanthanides (Fig. 1). The latter corresponds to the elements with

atomic numbers ranging from 57 {lanthanum (La)} to 71 {lutetium (Lu)} in the periodic table [10]. According to their atomic numbers, they have been divided into two types, i.e., “light rare earths elements” (LREEs) and “heavy rare earths elements” (HREEs). Due to similar chemical properties, ‘Y’ is frequently associated with HREEs. The elements from ‘Sm’ to ‘Tb’ are sometimes considered as “middle rare earth elements” [11].

Because of their extraordinary properties, REEs are employed in a variety of high-tech applications, such as lasers, permanent magnets [12,13], energy storage, etc., which are also regarded as technology-critical metals. Their supply has become important due to the ever-increasing global demand for these metals, especially from diverse supplies. Besides, they are frequently mined as by-products or co-products of other metals. Some well-known minerals as primary source of ores include bastnäsite, monazite, parisite, lanthanite, ancylite, chevkinite, allanite, loparite, cerite, stillwellite, britholite, fluocerite, and cerianite [14]. Phosphate or apatites are other minerals that could also be exploited for mining and processing [15,16]. Alternative sources must be investigated due to the crucial demand for these resources, particularly in the context of CE [17,18]. Exploitation of secondary sources is becoming increasingly vital with regards to economic and environmental concerns [19]. Due to high abundance of REEs in e-waste, selective recovery therein could be promising [18,20,21].

Recently, many studies have been conducted on EOL items containing large quantities of REEs, including magnets [22,23] and fluorescent lights [24,25]. Usually, one or more acid components, such as nitric acid (HNO₃), sulphuric acid (H₂SO₄), hydrochloric acid (HCl), phosphoric acid (H₃PO₄) or their combination, are used in chemical attack or leaching process (e.g., aqua regia). Subsequently, these leachates are extracted and separated using SX, ion exchange, and/or precipitation [26]. SX, also known as liquid–liquid extraction, is utmost widely employed method for recovery of REEs from aforementioned sources. Despite its widespread usage on a commercial scale, SX has some drawbacks in terms of huge volume of hazardous and/or inflammable solvents, thereby posing a threat to human health and the environment [27]. For instance, aliphatic diluents are considered as highly inflammable liquids and smokes with safety concerns. Evaporation is also problematic when these organic solvents are used during hydrometallurgy process. One of the key solutions for metallurgical processes is to eliminate or substitute these harmful solvents. Due to low volatile and non-flammable properties, ionic liquids (ILs) are considered as biodegradable and eco-friendly alternative to organic solvents [28].

Therefore, in this review, we will focus on the application of a new generation of ILs with modified chemical structures for selective separation of REEs. We will update the latest research on ILs used to extract and separate REEs from various e-waste feedstocks, including tailoring ILs for a specific task, elucidation of extraction mechanism, improved extraction efficiency and selectivity, combination with membrane technique and electrodeposition, and perspectives on its industrialization.

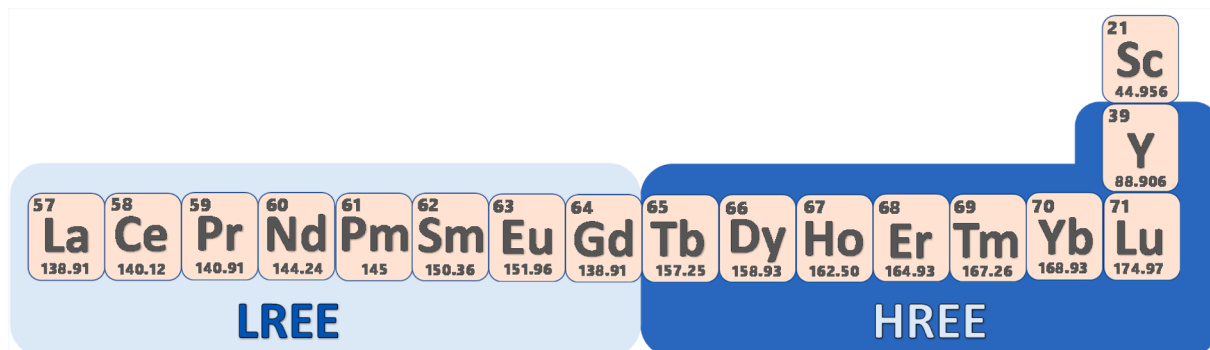


Fig. 1. Rare earth elements sectioned as light rare earth elements (LREE) and heavy rare earth elements (HREE).

2. Ionic liquids and their applications in e-waste processing

ILs could be defined as solvents comprising of organic moiety and ions with low melting point ($<100\text{ }^{\circ}\text{C}$) [29]. Ever since their introduction in 1914, they have been attracting significant interest from researchers in wide applications due to their eco-friendly nature and environmental benefits as alternatives to organic solvents for various synthetic chemical and electrochemical reactions. They could be divided into two categories, namely ionic solvents, and deep eutectic solvents (DES). Ionic solvents consist of an organic cation (imidazolium, pyridinium, triazolium, ammonium, phosphonium, pyrrolidinium, piperidinium) and an organic (acetate, bis(trifluoromethyl) sulfonyl imide, triflate, etc.) / inorganic anion (chloride, bromide, hexafluorophosphate, tetrafluoroborate, etc.), while DES consist of combinations of two or three components which form hydrogen bond interactions with each other to make a eutectic mixture [30–32]. Apposite cation–anion combination factors in the modification of physicochemical properties of the ILs include viscosity, solubility, density, and hydrophobicity. They could be considered as “designer solvents” because they may be well modified according to the conditions and selectivity, and serve better than traditional organic solvents [33]. In orthodox approach used for metal extraction, ILs are usually designed in a way that they are usually in hydrophobic nature to avoid their miscibility with the aqueous phase. Their hydrophobicity is associated with the length of alkyl chain present in the organic moiety of the cation as well as types of the combined anion (hexafluorophosphate $[\text{PF}_6]^-$ or bis(trifluoromethyl)sulfonyl imide $[\text{NTf}_2]^-$). The density and viscosity of ILs are important characteristics to consider when implementing an extraction procedure since they could influence mass transfer (e.g., diffusion) [34]. Longer alkyl chain length will lead to less density and high viscosity, which might change autonomously for varied anions in preference to hydrophobicity [35]. Viscosity behavior also depends upon the chemical structure of ILs and may also be affected due to hydrogen bonding and van der Waals interactions therein [36]. Furthermore, ILs with less or non-coordinating ligand as anion tend to show lower viscosity than those possessing strong ligand as anion for identical cation. Metal ions also play a determining role in the solubility of an IL. The Hofmeister series (or the lyotropic series) bring a broad suggestion to the hydrophobicity of an anion with an ascending order: $[\text{SO}_4]^{2-} < \text{Cl}^- < \text{Br}^- < [\text{NO}_3]^- < \text{I}^- < [\text{ClO}_4]^- < [\text{SCN}]^- < [\text{NTf}_2]^-$. This order indicates the increasing charge density and hydration numbers for various anions [37,38]. Metal salts comprising hydrophilic anions with high charge density are implausible to mix in poorly coordinating hydrophobic ILs, e.g., 1-butyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide, $[\text{C}_4\text{mim}][\text{NTf}_2]$. Therefore, addition of a ligand or a functional group increases the solubility of metals in ILs. The strength of metal–ligand interaction could also be evaluated using the qualitative Pearson’s Hard–Soft Acid–Base (HSAB) theory. HSAB principle is used to categorize Lewis acid/base as hard, soft, or borderline on the basis of their charge density, polarizability, and oxidation state. According to “like attracts like” deduced from the HSAB principle, it could be classified that cations of high charge (e.g., lanthanides) act as hard acids whilst many oxygen bearing functional groups (e.g., phosphates, carbonates, nitrates, or carboxylates) act as hard bases. Researchers have previously reviewed the use of ILs for metal separation [28,39–44], while the use of the immobilized IL in metal extraction has been reviewed elsewhere [40,45,46]. Multiple research groups have already investigated the use of ILs for metal electro-deposition [31,43,47–49]. Some studies have also investigated a thorough explanation on the essential leading ideologies dictating metal extraction using ILs [50,51], as well as detailed applications for particular types of ILs, such as acidic ILs [52] or temperature receptive ILs [53].

E-waste production is growing globally at an exponential rate as a consequence of population growth, economic growth, and ever-increasing demand for electronic equipment. CE encompasses the concept that waste stream always constitutes an inherent value to be

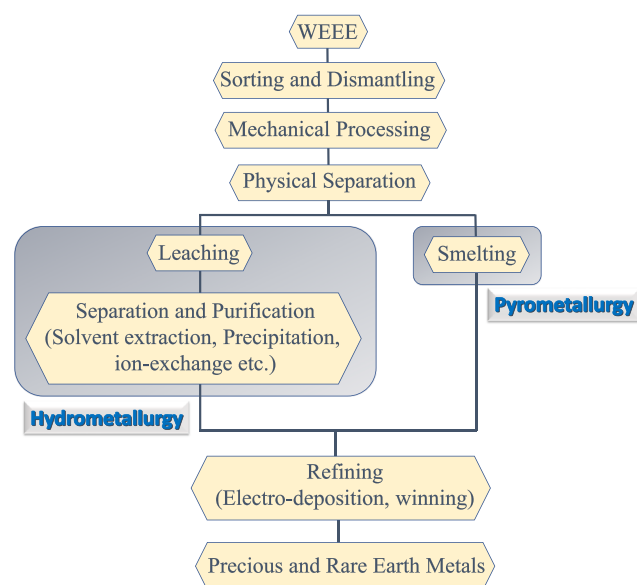


Fig. 2. Simplified flowchart representation of the main phases in WEEE reprocessing.

properly circulated, which could offer us both economic and environmental benefits. E-waste processing entails three major steps: 1) disassembling (removal of hazardous inorganic substances); 2) upgrading (improving metal content); and 3) refining (extraction of final products) (Fig. 2). Upgrading can be achieved by mechanical and/or metallurgical processing, while refining is to produce final metallic products by processing recovered materials [54–58]. Recent research in metal extraction processing comprises the use of non-toxic lixiviants, including ILs, amino acids and chelators. Metallurgical process consists of hydro- and pyro-metallurgy, but hydrometallurgy is more favored over pyrometallurgy for e-waste treatment. Pyrometallurgy tends to consume high energy, which may emit lethal volatiles and gases, and generate a large quantity of slags with probable loss of valuable metals therein. Conversely, hydrometallurgy exhibits distinct advantages in terms of less energy demand, less dust, higher recovery rate, and easier application circumstances. Furthermore, this technique can be applied to recycle different metals from diverse sources, such as low-grade ores, various metal ores and other secondary sources, especially at room-temperature [21,59]. Moreover, hydrometallurgy can be integrated with a variety of processes (e.g., membranes, solvent impregnated resins, absorption, and SX) to recover critical and precious metals. In particular, SX provides a continuous operation with good selectivity and extraction of a wide range of lanthanide concentrations from aqueous leachate solutions [60].

During hydrometallurgy processing of e-waste, ILs have broadly replaced classical organic solvents and been used as green solvents or extractants for the recovery of metals [61,62]. SX or liquid–liquid extraction system composes two immiscible liquid phases. The organic medium generally constitutes lipophilic complexing agents which help with the distribution of an analyte in the organic phase via formation of ionic pairs. Classical liquid–liquid extraction schemes are based on the usage of volatile organic compounds (VOCs), such as chloroform (CHCl_3), tetrachloromethane (CCl_4), benzene (C_6H_6), etc., as the organic liquid phase, during which metals can be recovered [12]. Nevertheless, excessive amounts of contaminated diluents are generated from traditional SX process. Thus, VOCs have been replaced by greener diluents (e.g., ILs and DES) to prevent environmental risk during SX and recycling processes. ILs have been used in a variety of industrial innovations and goods [70]. If appropriately chosen, ILs could provide an ecologically friendly alternative to current hydrometallurgical e-waste recycling. They could minimize waste generation and acid consumption while

achieving selective extraction [63]. In this review, ILs are mostly limited to recover value from printed circuit boards, fluorescent phosphor waste, nickel metal hydride batteries, and spent permanent magnets. Studies have been more focused on these waste types due to their inherent metallic value.

3. Design of ILs for critical metals recovery using solvent extraction process

Apart from the leaching process, the most common method for recovering REEs is solvent or liquid–liquid extraction process. Ever since the pioneer work by Dai et al. [64] that highlighted the potential of using ILs for SX to recover metals, extensive studies have demonstrated advantages of ILs over traditional organic solvents [65]. SX can achieve excellent solute re-distribution in biphasic system with dissolved REEs in the hydrophobic ILs and non-REEs in the aqueous phase [45]. Hydrophobic ILs can be water-miscible in the SX process by introducing kosmotropic ions (e.g., potassium phosphate, K_3PO_4 , potassium carbonate, K_2CO_3 , and sodium thiosulfate, $Na_2S_2O_3$) in the aqueous solution. As a consequence, a biphasic system can be formed due to enhanced stability and hydrogen bonding within the water molecules. Subsequently, these water-miscible ILs can be isolated from the aqueous phase and reused [66,67]. Hence, IL-based aqueous biphasic systems (IL-ABSs) have been considered for the extraction and separation process for metals recovery [68].

ILs are more thermodynamically favorable than traditional organic solvents in metal-extractant binding, which could improve the extraction efficiency and selectivity from the aqueous leaching solutions containing precious metals [69]. As a result, IL-based extraction systems have been developed by employing a combination of extractants or synergistic systems diluted in ILs, or task-specific ILs (TSILs) that include the extracting moieties into cation or anion of ILs [70].

Extraction process using ILs is generally different from that using traditional organic diluents [44,69]. ILs may dissolve neutral or charged compounds, while traditional solvents can only extract neutral species. Electroneutrality underpins these extraction types and procedures. Consequently, charged species must be counterbalanced in the aqueous

phase if they are extracted in the IL phase. Typical mechanisms could be elaborated according to leaching solutions (aqueous phase), extracting nature and types of applied ILs [71,72]. Three representative mechanisms are neutral exchange cationic exchange, and anionic exchange (Fig. 3).

For neutral exchange, the process is similar to that considered for traditional organic solvents, where IL acts as an alternative to the organic media and extracts neutral species from aqueous phase [73]. In cationic exchange, cationic species in the aqueous phase bind themselves with anionic species in the IL phase, whereas cationic species in IL phase are transferred to the aqueous phase in order to maintain electroneutrality [74], *vice-versa* for anionic exchange [74,75]. Equations in Table 2 illustrate general extraction mechanisms to maintain electroneutrality. In extraction Eq. (a), a neutral metal bonded with ligand is extracted from the aqueous phase into the IL phase through the deprotonation of ligand. Cation of IL (X^+) in Eq. (b) or anion (Y^-) in Eq. (c) is released to the aqueous phase to make up the incoming charge of the metal complex in extraction Eqs. (b) and (c), which represents a specific ion exchange mechanism (Table 2).

The fundamental disadvantage of IL-based extraction technique is that a portion of the IL is generally decomposed in the aqueous phase, which can be termed “sacrificial agent”. To determine the exchange process, the solubility of an IL towards aqueous medium is crucial because hydrophobic cations can attenuate these exchanges but hydrophilic anions exacerbate the phenomena [76,77]. For instance, improving the hydrophobic nature of an IL can transform anionic

Table 2

General mechanisms for IL extraction.

S.N.	General extraction equation	Exchange mechanism
(a)	$[REE]_{aq}^{n+} + nHL_{IL} \rightarrow \{ [REE] \bullet L_n \}_{IL} + nH_{aq}^+$	Neutral
(b)	$[REE]_{aq}^{n+} + nL_{aq} + mX_{IL}^+ \rightarrow \{ [REE] \bullet L_n \}_{IL}^{n+} + mX_{aq}^+$	Cationic
(c)	$[REE]_{aq}^{n+} + nL_{aq} + mY_{IL}^- \rightarrow \{ [REE] \bullet L_n \}_{IL}^{n+} + mY_{aq}^-$	Anionic

Notes: REE, rare earth element; H, proton; L, ligand; aq, aqueous phase; IL, ionic liquid phase; X^+ , cation of IL; Y^- , anion of IL.

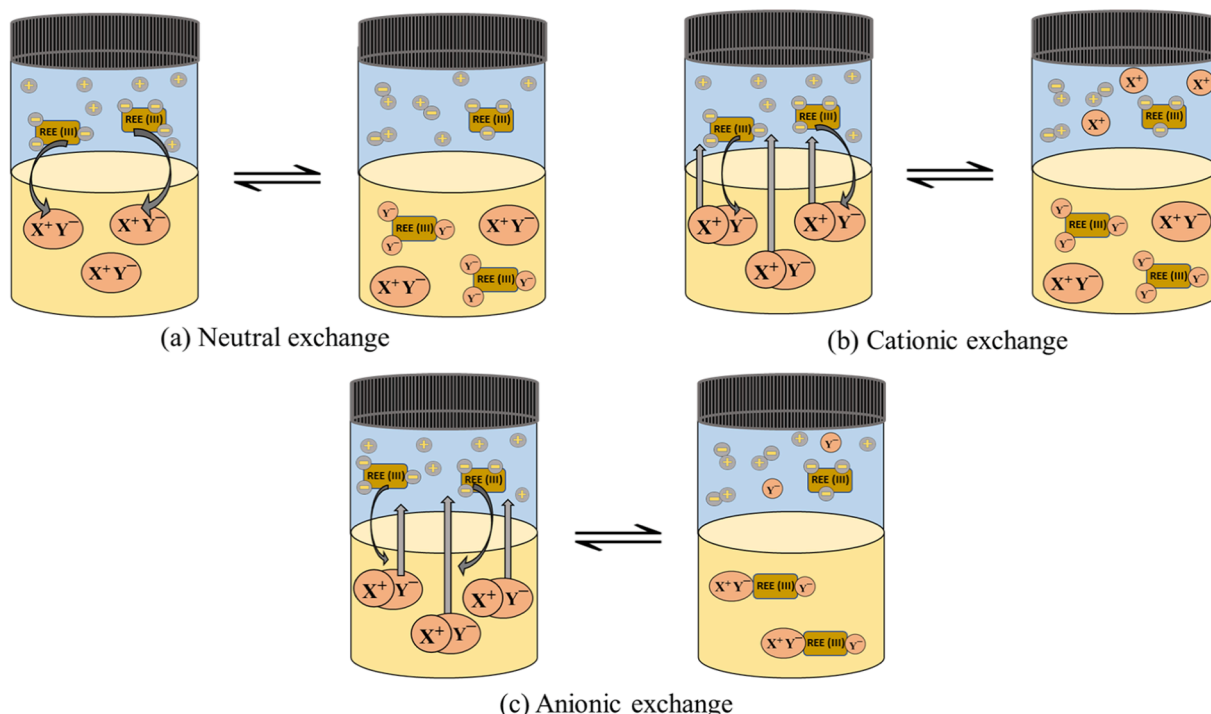


Fig. 3. Schematic representation for the solvent extraction process. (X^+Y^- : Ionic liquid).

Table 3

Summary of ionic liquids used for REEs recovery from e-waste via SX.

Ionic liquid	Extractant	Waste substrate	Leaching agent	REEs recovered (%)	Ref.
[A336][P204], [A 336][P507]	–	Phosphor fluorescent powder	HNO ₃	Y, Eu, Gd, Ce, Tb; (95.2)	[87]
[C ₄ mim][Tf ₂ N]	DODGAA	Phosphor fluorescent powder	HNO ₃ / HCl/ H ₂ SO ₄	Y, La, Ce, Eu, Tb	[88]
[A336][NO ₃]	TBP	Nd-Fe-B magnet	HNO ₃	Pr, Nd, Dy	[89]
[P66614][NO ₃]	EDTA	Nd-Fe-B magnet	HNO ₃	Nd (99.6), Dy (99.8)	[97]
[P66614][SCN]	–	SmCo magnet, Ni-H, Lamp phosphor	HNO ₃ / HCl	La (88), Sm (50), Eu (64)	[98]
R ₄ NCy, R ₄ ND	–	Nd-Fe-B magnet	HCl	Nd (98.7), Pr (99.02)	[99]
Primene 81R•Cyanex 572	–	Nd-Fe-B magnet	HCl	Nd, Tb, Dy; (99)	[105]
Primene 81R•D2EHFA, Primene 81R• Cyanex 572	–	Red phosphor (Y ₂ O ₃ : Eu ³⁺ - YOX)	HCl	Ce, Eu, Y (99.9)	[108]
Cyanex 272	–	SmCo magnet	HCl	Sm (99.4)	[109]
[A336][SCN], [A336][NO ₃], [A336][Cl]	–	SmCo magnet	HCl	Sm	[110]
[C101][SCN], [A336][SCN]	–	waste fluorescent lamp phosphor	HCl	Y (98.2), Eu (98.7)	[113]
[P66614][NO ₃]	DEHEHP	waste fluorescent lamp phosphor	HNO ₃	Y, Ce, Eu, Tb	[115]
Cyphos IL 104	TOPO	waste tube light phosphor powder	HCl	Y, Eu	[116]
[N444Bn] ₂ [SA]	–	SmCo magnet	HOAc	Sm, Nd, La	[117]

exchange process to neutral exchange process [78]. Acidity of the aqueous phase also plays an important role in ion-exchange process [79]. DES could be an alternative to organic solvents for metals recovery as well. Depending on the hydrogen bond donor (HBD) selection and the quaternary ammonium salt to HBD mole ratio, DES exhibit unique solvent characteristics in terms of their low cost of synthesis, minimal environmental toxicity, and solubility for a variety of metal oxides [80,81]. However, DES have been widely studied for the leaching process but less for the extraction process. Hydrophobic DESs also showed inspiring results [82–86] when they are used for the extraction and separation of metals. Nevertheless, unlike ILs, DES are completely soluble in water, therefore, a biphasic extraction is not a suitable process in this case.

In the next section, the use of IL as a solvent for liquid–liquid extraction will be examined in the context of e-waste recycling (Table 3). It is worth noting that not all the experiments have employed real waste. Instead, synthetic solutions of metal oxides are mainly used. This practice may overlook interactions in multifaceted system of e-waste leaching solutions and ignore the importance of trace pollutants that might be produced or present in the waste substrates during the extraction process. Then, the extraction efficiency could drop when ILs are applied in a real waste system. Generally, extraction efficiency (E), distribution ratio (D), stripping efficiency (S), and separation factors (β) are calculated using Eqs. (1)–(4) during the experimental studies.

$$\text{Extraction efficiency (E)} : E = \frac{M_i - M_f}{M_i} \times 100\% \quad (1)$$

$$\text{Distribution ratio (D)} : D = \frac{M_i - M_f}{M_f} \quad (2)$$

$$\text{Stripping efficiency (S)} : S = \frac{M_{aq}}{M_{ILs}} \times 100\% \quad (3)$$

$$\text{Separation factor (\beta)} : \beta = \frac{D_a}{D_b} \quad (4)$$

where $[M]_i$ and $[M]_f$ are the initial and final concentrations of REEs in the aqueous phase, respectively; $[M]_{aq}$ represents the equilibrium concentration of REE ions in stripping acid; $[M]_{ILs}$ is the initial concentration of loaded REE ions in the extracting phase; D_a and D_b are the distribution ratio for REE ion a and b, respectively.

4. ILS for recovery of REEs from e-waste

Recovery of REEs from e-waste was initially reported by Yang et al. in 2012, where bifunctional ionic liquids (Bif-ILs) were prepared by means of methyltriethylammonium chloride {Aliquat336 (A336)} and commercially available acidic organophosphorus extractants: diisooctyl

phosphate, P204 and 2-ethylhexyl phosphoric acid-2-ethylhexyl ester, P507 (humic acids, HA) (Fig. 4). These Bif-ILs were used to extract REEs from waste fluorescent powder [87]. Selection of acidic organophosphorus anions instead of neutral ones was associated with their internal synergistic and steric effects. Thus, this Bif-IL offered higher extractability than that of A336 or HA extractant alone. Despite many studies on REE recovery using neutral organophosphorus extractants (e.g., tributyl phosphate, TBP; tri-*n*-octylphosphine oxide, Cyanex923; di-(1-methylheptyl)methyl-phosphonate, P350; etc.), there is still little research concerning the use of Bif-ILs for removal / recovery of REEs. The only drawback of using [A336][P204] and [A336][P507] extractants was the formation of third phase that should be further removed using 10 % isopropanol. The recovery rate for REEs was 95 % after 5–7 stages using a crosscurrent abstraction procedure at a $V_o:V_w$ ratio of 4:1 and pH of 0.56. The extraction efficiency for acidic extractants was compared with neutral extractants (e.g., P350, TBP and Cyanex923) and the extractability was in the order of Cyanex923 > [A336][P204] = [A336][P507] > P350 > TBP because both [A336][P204] and [A336][P507] possess binary functional groups. The Bif-ILs exhibited very stable reusability even after multiple loading/stripping experiments, where the loss of extractants was negligible.

It is difficult to find an extractant readily soluble in IL. For instance, organophosphorus extractants, containing 2-ethylhexylphosphonic acid mono-2-ethylhexyl ester (PC-88A) and di(2-ethylhexyl) phosphoric acid (D2EHFA) and known as an extractant for the recovery of REEs, exhibit poor solubility in ILs. However, in 2012, Yang et al. successfully confirmed the solubility of *N*, *N*-dioctyldiglycol amic acid (DODGAA) (Fig. 5) in ILs. Due to the solubility of DODGAA towards ILs, it was used for recovery of REEs from phosphor powders in waste fluorescent lamps with IL [C₄mim][Tf₂N] as extracting solvent [88]. An effective recovery of REEs (e.g., Y, Eu, La and Ce) from the metal impurities (Fe, Al and Zn) was accomplished in acidic leaching solution of phosphor powders using [C₄mim][Tf₂N] based IL containing DODGAA as novel extractant at different pH levels. DODGAA showed a higher selectivity for heavier REEs (Y, La, Ce, Eu and Tb) as compared to commercial extractant PC-88A. Stability and reusability of extractants is another important aspect for their industrial use. The loss of this extractant was negligible with only slight decrease in extractability after five cycles. However, this hydrophilic IL should be redesigned to be more hydrophobic. After all, this system is selective for recovery of REEs.

Owing to selective properties and cost-effective value, A336-type ILs have been attracting great attention as TSILs for SX in recent years. For example, tricaprylmethylammonium nitrate ([A336][NO₃]) was used as a diluent IL with TBP (Fig. 6) as an extractant for the recovery of P [89]. During their experiments, the addition of sodium nitrate (NaNO₃) into aqueous media dramatically enhanced the extraction of REEs due to the presence of NO₃[−]. Nd(III) was enriched in the IL phase after continuous extraction process in Eq. (5).

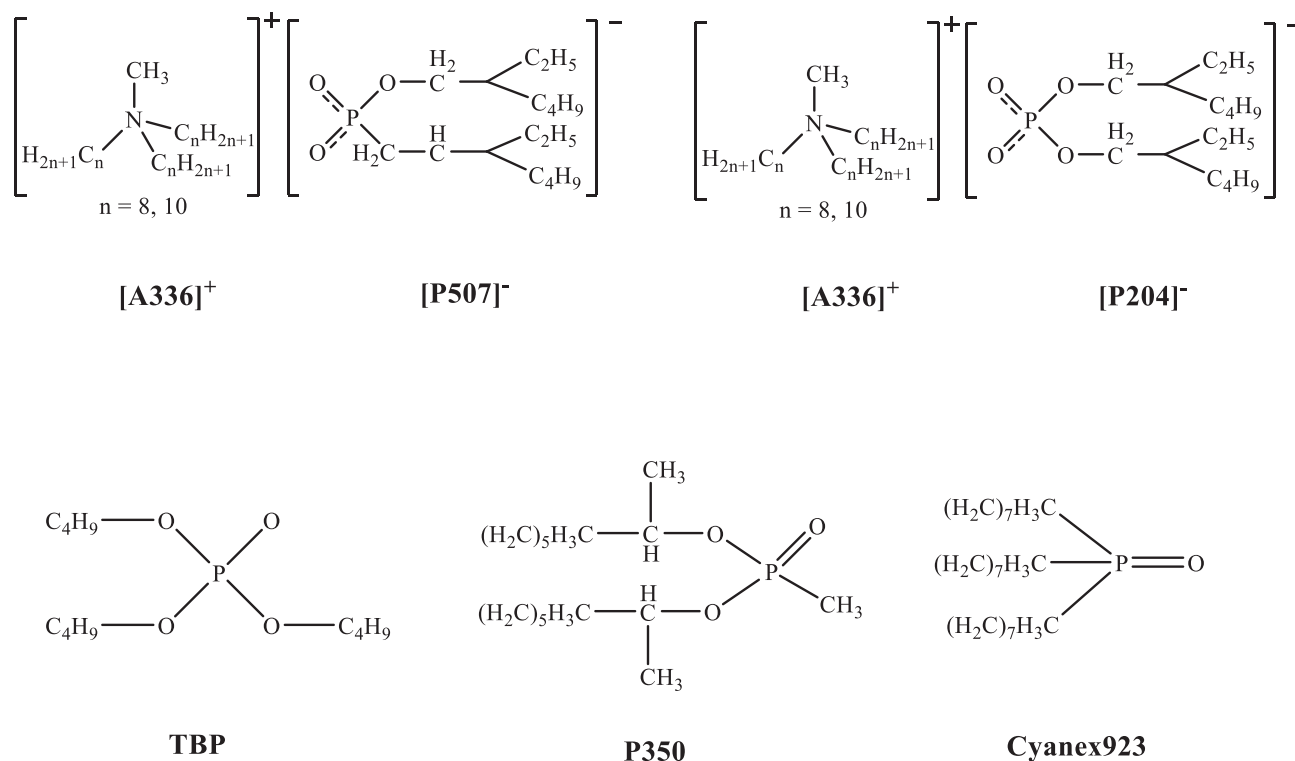


Fig. 4. Chemical structures of the ILs and extractants used in Ref. [87].

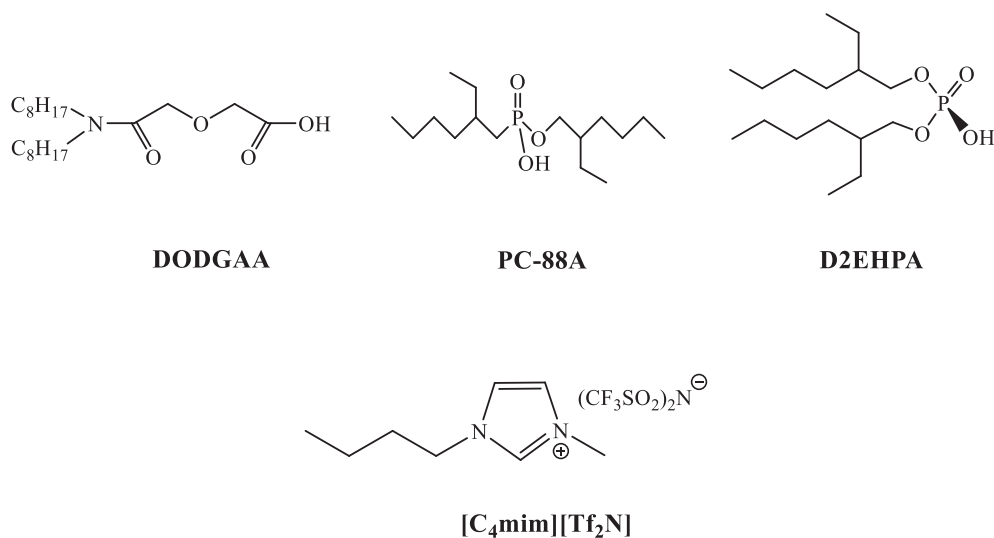


Fig. 5. Chemical structures of the ILs and organic extractants used in Ref. [88].

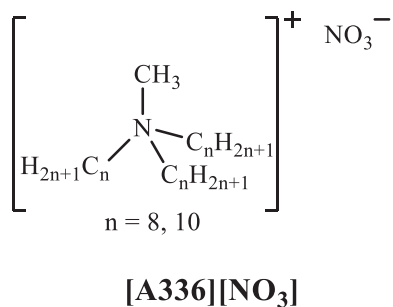
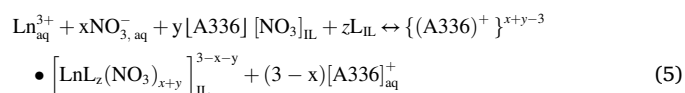


Fig. 6. Chemical structure of the ILs used in Ref. [89].



Moreover, the extractability for REEs was considerably enhanced with increased concentration of nitrate ions (1.0 to 5.0 M) in the aqueous phase, achieving a nearly 100 % extraction efficiency at 3.0 M NO_3^- . Seven recycling experiments were performed with almost negligible loss of extractant and a slightly reduced E value.

Binnemans et al. have successfully synthesized and employed ILs for the recovery of both transition and rare earth metals from e-waste and synthetic solutions [38,90–96]. Particularly, undiluted non-fluorinated IL, trihexyl(tetradecyl)phosphonium nitrate ([P66614][NO₃]) (Fig. 7)

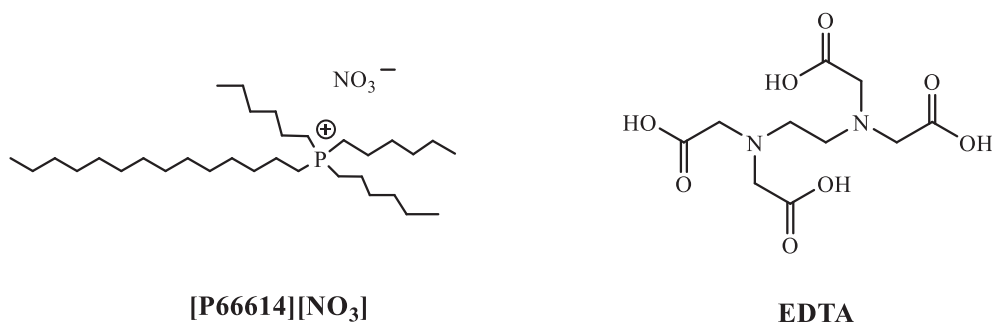


Fig. 7. Chemical structures of the ILs and extractants used in Ref. [97].

was successfully applied for the separation and recovery of REEs from NdFeB magnet [97]. Iron free leachate was obtained using nitric acid followed by SX at pH of 2. Cobalt was separated using [P66614][NO₃] alone, while Nd and Dy were extracted using this IL with ethylenediaminetetraacetic acid (EDTA) as a selective complexing agent. Overall, the process led to recovery of metal oxides with high purities, e. g., CoO (99.8 %), Nd₂O₃ (99.6 %), Dy₂O₃ (99.8 %), which would benefit industrial processes.

Fluorinated ILs are not considered ideal because of the presence of hydrophilic 1-alkyl-3-methylimidazolium cations. Thus, it is necessary to replace them with hydrophobic quaternary ammonium or phosphonium cations with long alkyl chains in ILs, for instance, Aliquat 336 and [P66614] cations. Owing to lower solubility in water, [P66614] based IL is more advantageous over Aliquat 336. Based on this notion, [P66614][SCN] (Fig. 8) was employed as an extractant in SX for the separation of the pairs of Ni(II)/La(III), Co(II)/Sm(III) and Zn(II)/Eu(III) [98]. Unfortunately, REEs showed lower affinity in comparison to transition metal ions towards [P66614][SCN] during the extraction process. The strong affinity of transition metals towards the IL could be attributed to strong metal–ligand binding (σ -bonding and π -back bonding) between SCN[−] anion and 3d transition metal ions as compared to their coordination with rare earth ions. But this difference in extraction behavior was useful in separating REEs from transition metals.

Comparative studies have also been conducted for the recovery of Nd and Pr from NdFeB magnets using ILs [99]. This work has investigated the extraction efficiency of different Bif-ILs towards REEs. The results demonstrated that Bif-ILs, i.e., trioctylmethylammonium bis(2,4,4-trimethylpentyl)phosphate (R₄NCy) and trioctylmethylammonium di(2-ethylhexyl)phosphate (R₄ND), performed better than conventional extractants, including Aliquat 336, bis(2,4,4-trimethylpentyl) phosphinic acid Cyanex 272 and D2EHPA, under similar experimental conditions (Fig. 9). At pH 2.5 and A:O ratio of 1:1, the synthesized ILs were compared with neutral extractants (Aliquat 336, Cyanex 272, and D2EHPA) for Nd and Pr extraction in HCl media. The extraction efficiency for Aliquat 336-Cyanex 272 and Aliquat 336-D2EHPA was substantially higher than that for [Cyanex 272] and [D2EHPA] alone. When compared with R₄NCy and R₄ND, their *E* values were higher than that for commercially available extractants, with R₄ND being even superior to R₄NCy. A two-stage counter current modeling investigation revealed extraction efficiency of 98.97 % for Nd and 99.02 % for Pr. When loaded organic phase was stripped using H₂SO₄ acid, 98.11 % and 98.75 % of

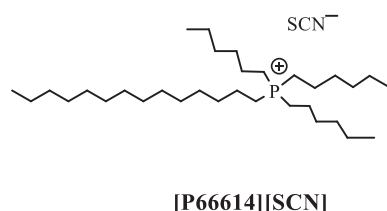


Fig. 8. Chemical structure of the ILs used in Ref. [98].

stripping efficiency was found for Nd and Pr, respectively.

In another work, recovery of Nd/Tb/Dy from NdFeB magnet wastes was investigated using cationic extractants in SX. It has been confirmed that the extractability of extractants for recovery of Nd and Pr was in the order: Cyanex 272-Aliquat 336 IL > D2EHPA-Aliquat 336 IL > Cyanex 272 > D2EHPA > Aliquat 336 [99]. Another commercially used extractant, i.e., Cyanex 572 (Cy572, a mixture of Cy272 and PC88A), is specifically designed for the extraction and purification of REEs [100]. Owing to the strong bond created by REEs, combination of Cy272 and PC88A can achieve better stripping of REEs using lower concentration of acid rather than strong acid [100–102]. Wang et al. recovered Th with purity of 95 % and 99.9 % from low and high Th concentrations, respectively, through a series of extraction, scrubbing, and stripping steps [103]. Given that REEs making up 30 % of NdFeB magnets [18,104] and Nd, Dy, and Tb being essential REEs in clean energy market [20], this work focused on how to recover and extract these REEs from magnet wastes. SX approach was used to test the selectivity of extractants, such as Cyanex 272 and Cyanex 572, in separating REEs. However, fluctuation of pH level is problematic in cationic extraction. Thus, Primene 81R•Cyanex 572 (Fig. 10) was synthesized to recover the REEs with 99 % purity from an aqueous mixture of Nd/Tb/Dy in chloride medium [105]. When REEs were extracted with this IL, pH value was maintained because there was no proton exchange in the aqueous phase.

ILs containing quaternary ammonium or phosphonium salts were mainly used for REEs separation [106,107]. However, Pavón et al. have investigated recovery of REEs from red phosphors (Y₂O₃:Eu³⁺ - YOX) using a different IL with a primary amine (Primene 81R) because of its high basic nature in comparison to secondary and tertiary amines [108]. Fluorescent lamp wastes were selected for the recovery capability of REEs therein. REE(III) oxides were obtained by precipitating and calcination of YOX leachate with oxalic acid. To study the effectiveness of REEs separation in chloride medium, Cyanex 572, D2EHPA, and ILs Primene 81R•Cyanex 572 and Primene 81R•D2EHPA were used. Yt, Eu, and Ce were separately recovered using the Primene 81R•D2EHPA IL and Primene 81R•Cyanex 572 IL as extractants after a four-stage crossflow SX procedure. Their goal was to separate Yt, Eu, and Ce from leachates using cationic commercial extractants D2EHPA and Cy572 or IL extractants including Primene81R•D2EHPA IL (P81R•D2EHPA IL) and Primene 81R•Cyanex 572 IL (P81R•Cy572 IL). This study has not only selected commercial cationic or IL extractants based on their selectivity and behavior in the Ce/Eu/Y separation, but also investigated separation of REEs from Ce/Eu/Y mixture. When P81R•D2EHPA IL and P81R•Cy572 IL were used as extractants, Ce(III), Eu(III), and Y(III) with purity of 99.9 % can be obtained. Hence, Ce/Eu/Y could be selectively separated from the leachate of real fluorescent lamp waste when these ILs were used in a crossflow SX.

Furthermore, SmCo magnets were used to construct a solvometallurgical recycling method for the recovery of Sm, Co, Cu, and Fe [109]. In the process, crushed and milled SmCo magnets were leached in HCl medium and Aliquat 336 was used as an extractant to recover

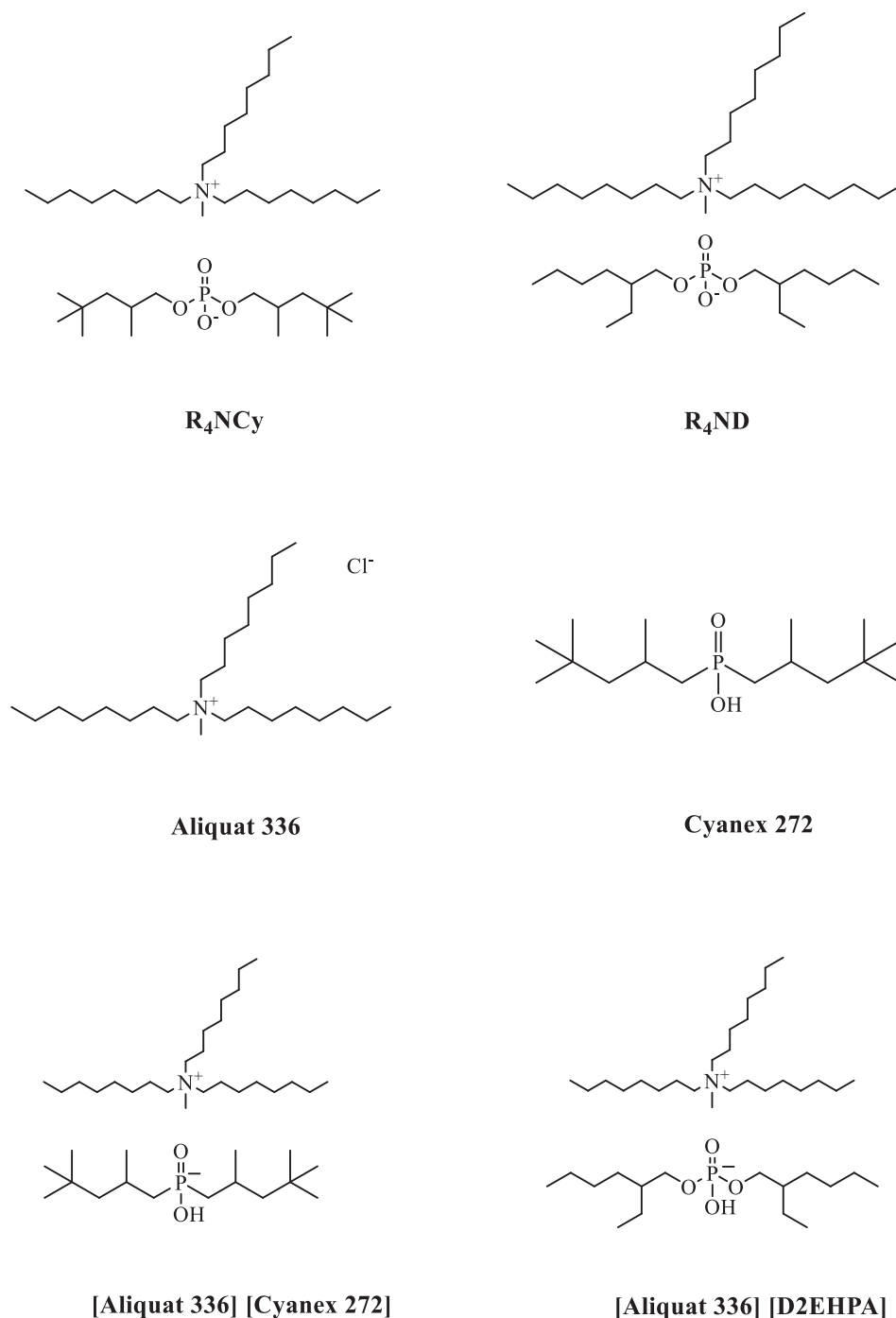
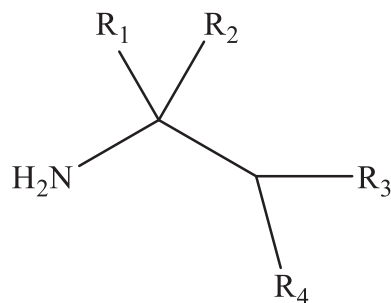


Fig. 9. Chemical structure of the ILs and extractants used in Ref. [99].

cobalt, copper, and iron from the leachate. The extraction efficiency for cobalt was enhanced by using a non-aqueous SX technique. In terms of leaching and extraction efficiency, an aqueous solution containing 37 wt % HCl was used as the lixiviant, and Sm was separated from Co/Cu/Fe. During stripping, 0.5 M HCl and 5 vol% NH_3 solution were used to extract cobalt, copper and iron from the solvent, respectively. Sm was successfully recovered from the leachate using 20 vol% Cyanex 272 in dodecane with pH adjustment by 2 M NaOH solution. Finally, Sm was extracted using 0.2 M oxalic acid precipitation stripping. The metals were precipitated with high purities, e.g., 99.4 wt% for Sm, 98.3 wt% for Co, and 100 wt% for Fe and Cu.

Similarly, Sm/Co/Cu were extracted from the leaching solution of SmCo magnets using a different IL as an extractant without any dilutant,

i.e., undiluted quaternary ammonium IL with chloride, thiocyanate and nitrate as anions [110]. This SX system with quaternary ammonium salts is ecologically beneficial due to the absence of any diluents and limited use of chemicals. As the concentration of anions in undiluted ILs is always more than that in traditional SX process, a concentrated SX method is essential. Nevertheless, it is always more convenient to work with high concentrations for industrial applications due to its better extraction efficiency. Two split-anion SX methods were also investigated. In general, different anions in aqueous and organic phases exemplified split-anion extraction [111]. Chloride anions were in the aqueous phase, while thiocyanate or nitrate anions were found in the water-insoluble organic phase (IL). As SCN^- and NO_3^- have higher affinity for the organic phase than chloride anions ($Cl^- < NO_3^- < SCN^-$), both were



Primene 81R

Fig. 10. Chemical structure of the extractant used in Ref. [105].

considered appropriate for usage in conjunction with chloride aqueous media, offering an excellent immiscibility of the phases [112]. This research demonstrated the indirect recycling of SmCo magnets using SX, which is useful for recovering specific high-purity metals rather than reprocessing magnets directly. A very high extraction efficiency was achieved after a single extraction process. The $\text{Cl}_{\text{aq}}/\text{SCN}_{\text{org}}$ method exhibited extremely high extraction efficiency for transition metals (e.g., > 99.9 % Co and > 99.7 % Cu) without any co-extraction of Sm at a finely regulated salt concentration. However, because of the unstable SCN^- anion, this system appeared to be unstable. Due to the low stability of the quaternary ammonium salts in alkaline circumstance and the impossibility of stripping with water, the extracted metals could not be removed successfully from $[\text{A336}][\text{SCN}]$. The $\text{Cl}_{\text{aq}}/\text{Cl}_{\text{org}}$ and $\text{Cl}_{\text{aq}}/\text{NO}_3$ organic systems, on the other hand, were found to be stable in concentrated chloride medium.

For the separation of Y and Eu from their respective metal oxides obtained from the processing of fluorescent lamp waste powder, trihexyl (tetradecyl)phosphonium thiocyanate, $[\text{C101}][\text{SCN}]$ and $[\text{A336}][\text{SCN}]$ were used as IL for the SX [113]. Under similar experimental conditions, $[\text{C101}][\text{SCN}]$ showed better extraction efficiency than $[\text{A336}][\text{SCN}]$, thereby proving the dominance of quaternary phosphonium based ILs over quaternary ammonium-based ILs. Before using the leachate from waste substrate, the experiments were performed using the synthetic chloride solution, and similar conditions were applied for the leaching solution. Nakamura et al. [114] achieved a purity higher than 98 % for Y_2O_3 but 10 % for Eu_2O_3 , whereas Tunsu et al. [104] demonstrated a better separation of Y/Eu with 99.82 % of Y_2O_3 and 91.6 % of Eu_2O_3 . For the SX stage, both procedures employed traditional organic solvents including PC-88A and Cyanex 572. Under comparable experimental settings, the regenerated IL $[\text{C101}][\text{SCN}]$ was evaluated for the extraction of Y(III) with only slight change in extraction efficiency. Ultimately, 98.2 % of Y_2O_3 and 98.7 % of Eu_2O_3 were obtained.

In one of these studies, an IL mixture of $[\text{P66614}][\text{NO}_3]$ and

DEHEHP, also called PND, was used as an extractant for the selective separation of REEs (Y, Ce, Eu, and Tb) from the leachate of waste fluorescent lamp phosphor [115]. After a four-stage extraction process using $[\text{P66614}][\text{NO}_3]$ -DEHEHP, all the REEs could be completely extracted into the IL phase, whereas non-REEs remained in the aqueous phase under the same extraction conditions. No obvious decrease in REEs extraction was found even after five continuous extraction-stripping cycles. Therefore, synergistic extraction based on quaternary phosphonium salts (e.g., $[\text{P66614}][\text{NO}_3]$ -DEHEHP) could be an ecologically favorable strategy to achieve excellent extraction of REEs from e-waste.

Recovery of yttrium (Y) and europium (Eu) from waste tube light phosphor powder leaching solution by SX and chemical reduction was recently studied [116]. D2EHPA, trioctylphosphine oxide (TOPO), and trihexyl(tetradecyl)phosphonium bis-2,4,4-(trimethylpentyl) phosphinate (Cyphos IL 104) were used to separate Y(III) and Eu(III) from the leachate (Fig. 11). SX with TOPO showed limited extractability of metals, while the highest separation factor of 60.6 was achieved with 0.1 M D2EHPA at an aqueous pH value of 2.56. At an O:A ratio of 1:1.5, yttrium was completely removed in two phases with 60 mg/L Eu co-extraction. Precipitate stripping with oxalic acid was further used to recover yttrium as yttrium oxalate $\text{Y}_2(\text{C}_2\text{O}_4)_3$ from the loaded organic phase. The chemical reduction of Eu(III) to Eu(II) was possible with Zn although there was 16 % Y (302.4 mg/L) precipitation with Eu(II) sulfate.

More recently, Deng et al. have studied recovery of Sm(III) from the spent SmCo magnets [117]. A novel IL, i.e., benzyltributylammonium decanedioate ($[\text{N444Bn}]_2[\text{SA}]$) (Fig. 12), was well-designed to extract REEs and isolate Sm(III) from wasted SmCo magnets in acetate solution. Extraction of Nd(III) and La(III) in three mediums were determined and compared in terms of equilibrium time, concentration of REEs, initial pH of aqueous solution, concentration of salting-out agent, extractant concentration, and extraction temperature. Acetate medium demonstrated superior extractability of La(III) than HCl solution (97.6 % versus 78.6 %) owing to the low-hydrated property of acetate ions. Back-extraction studies revealed that 1.5 mol/L HAC can remove 91.4 % Nd(III) from $[\text{N444Bn}]_2[\text{SA}]$, demonstrating its remarkable reusability for industrial

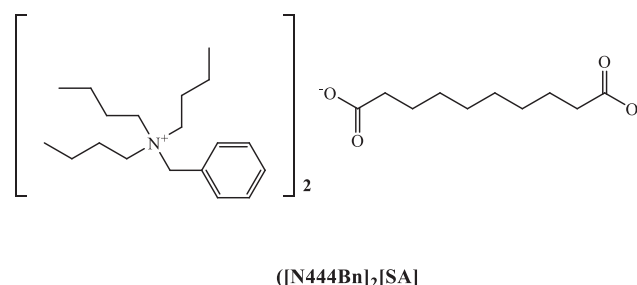
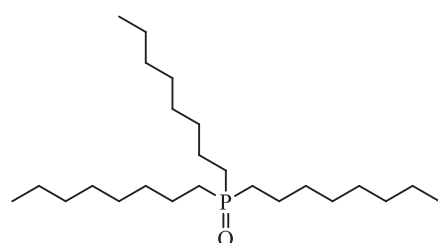
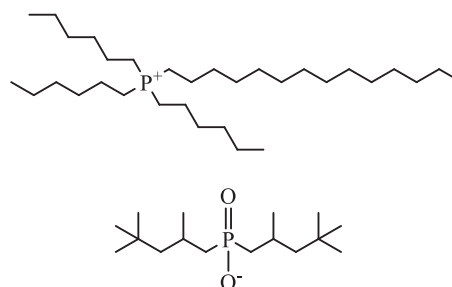


Fig. 12. Chemical structure of the IL and extractant used in Ref. [117].



TOPO



Cyphos IL 104

Fig. 11. Chemical structure of the IL and extractant used in Ref. [116].

separation-stripping cycles. In addition, an ion association mechanism was postulated to explain the extraction behavior. In simulated solution, the maximum separation factor of Sm(III)/Co(II) was 3078. To sum up, dicarboxylate-type IL can successfully recover Sm(III) from real SmCo magnets waste with a recovery rate of 96.8 % and a Sm(III)/Co(II) selectivity of 148.

Nevertheless, most of researchers have focused on simulated solutions as a mimic of waste-stream rather than real waste substrates. For example, synthesized hydrophobic 1-alkylcarboxylic acid-3-methylimidazolium bis(trifluoromethylsulfonyl) imide ILs, $[(CH_2)_nCOOH_{min}][Tf_2N]$ ($n = 3, 5, 7$), were used to separate Nd/Fe and Sm/Co from simulated aqueous solution impersonating NdFeB and SmCo permanent magnet leachate [118]. 1,3-dihexylimidazolium nitrate, $[HHIM][NO_3]$ IL was used in the SX process to separate the pairs of Sm/Co and La/Ni pairs, which could be considered important for the reutilization of SmCo magnets and NiMH batteries. *N*-butyl-*N*-ethyl-piperidinium bis(trifluoromethylsulfonyl)imide (EBPiP-NTf₂) and *N*-octyl-*N*-ethyl-piperidinium bis(trifluoromethylsulfonyl)imide (EOPiP-NTf₂) were used as extractants for the recovery of Ta from sulfuric acid solution. More relevant studies have been reviewed in detail elsewhere [40,42,44,61,65,106,107,119]. Although these ILs presented promising results, more efforts are desired in IL extraction applications for real waste materials with more complex composition in order to make this recycling process commercially viable in the field of e-waste management.

5. Challenges and perspectives

Nowadays, more waste materials are being recycled and reused towards CE [4]. Regardless of emerging REEs pollution resulting from inappropriate disposal of increased generation of e-waste, there is a significant potential to recycle REEs and other critical elements (e.g., Fe, Cu, Al, Au, Ag, Pd). So far, SX is the most extensively utilized method for extracting REEs from e-waste. Nevertheless, despite extensive commercial use of SX, it has several downsides, for instance, the practice of using large quantities of lethal and/or flammable solvents that might pose a key threat to human health and the environmental safety [27]. To overcome this issue, ILs are regarded as greener alternative to organic solvents owing to their low volatile and non-flammable properties [28]. However, some significant challenges remain: 1) limited scale-up ILs extraction data from real e-waste; 2) high cost of predominant fluorinated ILs; and 3) reusability and recycling of ILs after REEs extraction.

Therefore, more research associated with real e-waste (e.g., fluorescent powder, SmCo magnets, NdFeB magnets, etc.) is desirable to acquire data on extraction efficiency and selectivity of TSILs on an industrial scale. Acidic extractants are preferable to neutral extractants due to their enhanced extraction efficiency. When ILs are used as a dilutant, a soluble extractant (e.g., DODGAA) is desirable. Combination of different ILs could be used for the selective separation of REEs. Moreover, non-fluorinated hydrophobic ILs with long alkyl chains (quaternary ammonium or phosphonium based ILs) could minimize the cost of ILs. Apart from quaternary ammonium or phosphonium based ILs, primary amines also tend to give better results because of their highly basic nature. Appropriate combination of different cation and anions allows the flexibility to design an IL with certain selectivity towards a specific metal. In addition to extractability and selectivity, reusability of TSILs must be completely assessed through multiple loading/stripping studies before their commercial use [120]. More efforts must be devoted to separation of intra-REEs, which is crucial to achieve high selectivity if LREEs could be separated from HREEs [106]. For example, the usage of TSILs to extract and separate REEs present in permanent magnets (specifically neodymium and dysprosium) from an acidic aqueous medium has recently been patented by Queens University Belfast and Seren Technologies [121]. Seren Technologies, which is aggressively seeking commercialization, constructed a pre-commercialized magnet recycling factory in England in 2018,

delivering a highly selective process capable of handling multi-ton shipments of magnets [122].

6. Future prospects

Now that REEs continue to play an indispensable role in critical materials for modern technologies, their efficient recovery from e-waste is currently one of the most promising routes to cope with the challenge in supply chain. Although SX with ILs is more environmentally friendly than that with organic volatiles, in-depth research is desired in order to obtain better understanding on design of selective ILs for specific REEs, optimization of SX process, and extraction mechanisms towards the future industrialization. In particular, even minor changes in the structure of ILs could alter the extraction mechanism, no matter they function as dilutants or extractants. Therefore, it is significant to design an IL that is highly efficient in terms of extraction and reusability. Apart from SX process for the efficient extraction of REEs, ionic solvents could also be used for REEs recovery via electrodeposition of metals from non-aqueous electrolytes. As the electro-positivity of REEs is greater than that of hydrogen, electrodeposition of these metals cannot be performed in aqueous solution. However, ILs can provide suitable conditions for electrodeposition, so the electrochemical behavior of various IL systems and their role in the recovery of REEs could be further investigated in the future.

7. Concluding remarks

IL is a promising alternative to organic/volatile solvents for selective recovery of REEs in terms of green separation and mitigated risks of toxicity and explosion. This review has systematically demonstrated the fundamental design of TSILs and their applications for selective extraction of REEs from e-waste. Specifically, Bif-ILs (e.g., [A336][P204] and [A336][P507]) are more advantageous than their precursors and neutral extractants owing to synergic effect. Incorporation of dilutant/extractant plays an important role in efficient extraction and separation. More TSILs (e.g., [A336][NO₃], [P66614][NO₃], [P66614][SCN], etc.) should be designed for cost-effective and selective extraction of REEs from e-waste. Economical and environmentally benign non-fluorinated ILs can be designed using hydrophobic, long-alkyl-chain quaternary ammonium or phosphonium cations as a substitute for hydrophilic 1-alkyl-3-methylimidazolium cations in fluorinated ILs. Synergistic extraction based on quaternary phosphonium salts (e.g., [P66614][NO₃]-DEHEHP) can achieve excellent extraction of REEs into IL phase with non-REEs in the aqueous phase for easier separation. In addition to extractability and selectivity, reusability of TSILs must be completely assessed through multiple loading/stripping studies before their commercial use. Thus, regeneration of spent ILs and subsequent separation of extracted REEs from ILs require more efforts in order to recover REEs from e-waste. Furthermore, it is essential to acquire more fundamental data on extraction efficiency and recovery rate of REEs from real e-waste using novel TSILs towards industrialization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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