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Abstract

The Latest Trends in Recycling Spent Lithium-Ion Batteries †

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As we know, the use of electric vehicles is for reducing carbon emissions as well as reducing environmental pollution. On the other hand, the production of the batteries causes environmental pollution, because the heavy metals used in the batteries are toxic and have a negative impact on the environment [1]. The production of batteries, which contain expensive and rare elements, influences the high price of electric cars. Lithium-ion batteries (LIBs) are one of the main elements of electric vehicles (EVs) that have a significant impact on the price of the car. The recycling of LIBs is very important from an economic viewpoint, since cathodes in lithium-ion batteries contain valuable metals such as lithium, cobalt, and nickel.

In accordance with current regulations, 100% of LIBs—both automotive and industrial—must be recycled. Each battery factory is obliged to do it itself or commission it to an authorized recycler with whom it must conclude a contract before selling the product. Unfortunately, there is no company in Poland that recycles LIBs on an industrial scale. It is in our interest to recycle them in an environmentally friendly way using the latest technologies [2].

The recovery of metals from used batteries can be performed using various methods (pyrometallurgical, hydrometallurgical, and biometallurgical). The recovery of metals from LIBs in a hydrometallurgical process is a multistep procedure that contains the following stages: (I) pretreatment, (II) metal extraction, and (III) final product application [3,4].

The liquid–liquid extraction process is a classical method of separation and concentration of metal ions from aqueous solutions, used in many industries. Currently, a lot of information can be found in the literature about the use of ionic liquids in extractive processes of separation and the separation of metal ions from aqueous solutions. Imidazolium, pyridinium, phosphonium, sulfonium, and quaternary ammonium salts play a special role here [5]. Among them, imidazolium ionic liquids are solvents whose physicochemical properties are well known. For example, 1-butyl-3-methylimidazolium hexafluorophosphate [C_4mim][PF_6] was used in the process of extractive release of Cd(II), Pb(II), Cu(II), Hg(II), and Ag(I) ions [5]. It was found that the tested ionic liquid can replace the previously used chloroform as an organic solvent. The research results confirm that ionic liquids can act as solvents for other extractants, such as crown ethers, calixarenes, and organophosphorus compounds, during the extraction of alkali metal ions (Li, Na, K, Rb, Cs), heavy metal ions (Cu, Pb, Zn, Cd, Hg), and rare earth elements (Nd, La, Er, Ce, Sm, Eu) [6–9]. The presence of low-temperature ionic liquids in the extraction system next to classic extractants when separating metal ions from aqueous solutions may cause the so-called synergistic effect, i.e., increasing or decreasing extraction efficiency. Research on the impact of low-temperature ionic liquids on the efficiency of metal extraction using traditional extractants revealed that ionic liquids act as synergists. They improve the extraction efficiency of many metal ions, including Zn(II), Cd(II), Cu(II), and Hg(II) [10,11]; Ce(III), Eu(III), and Y(III) [12,13]; and Cs(II) and Sr(II) [14].

Ionic liquids are used not only as solvents and synergists, but also as extractants. In the process of extracting silver(I), gold(III), and palladium(II) ions [4], ionic liquids containing pyridinium, pyrrolidinium, piperidinium cations, and bis(trifluoromethylsulfonyl) imide anion were used. Ionic liquids containing nitrile or disulfide groups have very good extraction properties towards Ag(I) and Pd(II) ions [15]. Au(III) ions were efficiently extracted using functional ionic liquids containing an alkenyl group, a disulfide group, and a nitrile group [15]. Table 1 shows examples of ILs used as extractants in solvent extraction processes of metal ions. In turn, commercial ionic liquids, i.e., Cyphos IL 101 and Cyphos IL 109, were used as extractants of Zn(II) and Fe(III) ions from aqueous chloride solutions. It turned out that Cyphos IL101 in toluene allows the extraction of almost 100% zinc(II) both from a solution containing 1.8% hydrochloric acid and from a solution containing no acid. It was found that the extraction efficiency of the studied metal ions using Cyphos IL 101 increased in the Fe(II) < Zn(II) < Fe(III) series. The efficiency of metal extraction using Cyphos IL 109 was low and amounted to less than 20% [16]. In the work of Regel-Rosocka et al. [17], it was found that the extraction of Zn(II) and Fe(III) using Cyphos IL101 takes place according to the anion exchange mechanism, and the reaction order in relation to the metal and the extractant is equal to 1. Imidazolium and ammonium ionic liquids were tested as extractants of Zn(II), Cd(II), Cu(II), and Fe(III) ions from an aqueous solution of hydrochloric acid. The used trioctylmethylammonium chloride [MTOA+][Cl^-] allows the almost complete separation of Zn(II), Cd(II), and Fe(III) ions from an aqueous chloride solution, but the efficiency of their extraction process decreased with the increase in the concentration of metal ions. The extraction efficiency increased with an increase in the concentration of hydrochloric acid in the aqueous phase in the case of the extraction of Zn(II), Cd(II), and Fe(III) ions with 1-methyl-3-octylimidazolium tetrafluoroborate [18]. Cyphos IL 101 and Cyphos IL 104 are commercial ionic liquids that can be used as selective extractants of Pd(II) ions from 0.1 M HCl solution containing Ni(II), Cu(II), Pb(II), Fe(III), Rh(III), and Ru(III). The selectivity of Pd(II) extraction towards Pb(II), Fe(III), and Pt(IV) ions depended on the acidity of the aqueous phase and decreased with increasing HCl concentration [19]. Cyphos IL 101 and Cyphos IL 104 were also used to separate Ni(II) and Co(II) ions from aqueous chloride solutions [20]. The best separation effect of Co(II) and Ni(II) ions was achieved in the case of extraction from an aqueous solution containing no hydrochloric acid, from which over 95% of Co(II) was extracted using Cyphos IL 104 as an extractant.

Table 1. Examples of ILs used as extractants in solvent extraction processes of metal ions.

Ionic Liquid	Metal Ion	Ref.
Cyphos IL 101	Ni(II), Co(II), Zn(II), Fe(II), Fe(III), Pd(II)	[16–20]
Cyphos IL 104	Ni(II), Co(II)	[20]

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