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Investigating the leaching performance of ferric chloride solution for metallic alloys resulting from reduction smelting of spent lithium-ion batteries

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ABSTRACT

Smelting reduction of spent lithium-ion batteries (LIBs) produces metallic alloys containing Co, Ni, Cu, Mn, and Fe. Finding suitable reagents in terms of efficiency, economics, and friendly environment for the dissolution of these metals from the alloys is very important for the recovery process of the metals. In this work, the employment of ferric chloride solution for the dissolution of the metals from the alloys was studied. The effect of parameters like $FeCl_3$ concentration, temperature, time, and pulp density on the leaching efficiency of metals was investigated. Our results indicate that ferric ions in the leaching solutions act as oxidizing agents for the dissolution of the metals, while chloride anions as ligands for the formation of the complexes of the dissolved metal ions. The best conditions for the dissolution of full metals were 0.7 mol/L $FeCl_3$, 12.5 g/L pulp density, 22°C, and 30 min. In comparison with HCl or H_2SO_4 leaching agents, ferric chloride shows some advantages like a decrease in the dosage of acids and oxidizing agents, fast reaction kinetics, and low energy consumption. With its advantages, ferric chloride solution is considered a potential leaching agent in the recovery process of valuable metals from spent LIBs.

1. INTRODUCTION

Currently, rechargeable lithium-ion batteries (LIBs) are used widely in electronic devices (e.g., mobile phones, laptops, personal care devices, etc.) for energy storage because of their excellent electrochemical performance (e.g. high energy density, lightweight, and quick charging) (Dutta et al., 2018; Harper et al., 2019; Chen et al., 2021). The rising application of LIBs in electric vehicles (EVs) and stationary energy storage systems have further boosted the production of new LIBs in recent years and this trend is expected to continue (Or et al., 2020; Yang et al., 2020; Santos et al., 2021). However, such an increase could result in some future challenges: (1) the shortage of strategic metals like cobalt, nickel, and lithium due to the depletion of high-grade ores; (2) potential environmental risks after the end-of-life, resulting from hazardous heavy metals (e.g. cobalt, copper, iron, and manganese), electolytes, plastics, etc. (Gao et al., 2017; Meshram et al., 2019; Prasetyo et al., 2022). It is reported that only about 5% of spent LIBs are recycled (Zhao et al., 2020). Therefore, the recycling of spent LIBs is considered an effective solution to the sustainable production of LIBs and reduction in the environmental damage.

Great efforts have been undertaken for the recycling of spent LIBs and some processes have been commercialized as the Umicore process, the Sony-Sumitomo process, the Toxco process, and the Recupyl process (Xu et al., 2008; Zeng et al., 2014; Gratz et al., 2014; Thompson et al., 2020). Generally, there are three main technologies employed for metal recycling from spent LIBs consisting of pyrometallurgy, hydrometallurgy, and biometallurgy (Asadi et al., 2020; Zhang et al. 2020). Yet, the combination of pyro- and hydro-metallurgy technologies shows some advantages in the recovery of valuable metals like Co, Ni, and Li (Zheng et al., 2018; Lin et al., 2019; Tran et al., 2022a). These combined processes include two main steps: (1) the treatment of spent LIBs at various temperatures for removing organic matters without pretreatments such as classification of the spent LIBs size/type or dismantling of shells/plastics, and smelting reduction of metal oxides (e.g. Co, Ni, Cu, Fe, and Mn) to metallic alloys, while Li and Al remain in slags or dust (Zhou et al., 2020); (2) hydrometallurgical processes such as leaching, solvent extraction, ion exchange, and precipitation methods were employed for the purification of metals (Roman et al., 2018; Djoudi et al., 2019; Silva et al., 2021; Tran et al., 2022b). The separation performance of metal significantly depends on the composition and concentration of elements in the alloys/slags/dust and leaching agents. Therefore, it is necessary to seek suitable leaching agents with a low cost, high efficiency, and which are less harmful to the environment.

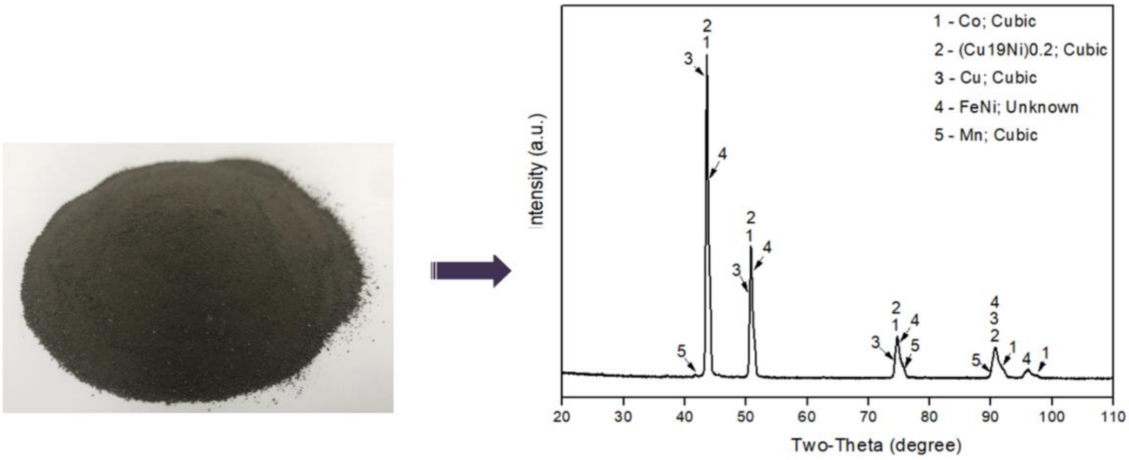
In our work, metallic alloys containing Co, Ni, Cu, Mn, Fe, and Si were produced from the smelting reduction of spent LIBs. To dissolve the metals, acidic solutions like H_2SO_4 and HCl were employed with the addition of hydrogen peroxide (H_2O_2) for enhancing the dissolution efficiency of the metals, especially Cu because of its high reduction standard ($E^o_{Cu^{2+}/Cu} = 0.34\ V$) (Tran et al., 2021; Tran et al., 2022c). In our study, the mixture of 2.0 mol/L acids and 10% (v/v) H_2O_2 solutions were required for the complete dissolution of the metals. However, the use of acidic solutions and oxidizing agents at high concentrations may result in problems in the maintenance of equipment, as well as process economics. Since iron exists in metallic alloys, ferric ions with their strong oxidizing property ($E^o_{Fe^{3+}/Fe^{2+}} = 0.77\ V$) can be considered as a potential oxidizing agent for the leaching of metals, which can avoid the addition of strange metals after the leaching. According to the reported literature, ferric chloride shows fast kinetics as compared with ferric sulfate as a leaching agent (Cordoba et al., 2008; Kaplun et al., 2011). Therefore, in this work, ferric chloride aqueous solution as a lixiviant was employed for the dissolution of metals from the metallic alloys. The effect of parameters such as ferric ion concentration, time, temperature, and pulp density was surveyed.

2. MATERIALS AND METHODS

2.1. Materials and reagents

Material powders were obtained from a Korean company with less than 100 μm particle size and black-gray color (Figure 1a). The chemical structure of the powders as metallic alloys of Co, Ni, Cu, Mn, and Fe was confirmed by X-ray diffractometer measurement (XRD, X'Pert-PRO, the Netherlands) (Figure 1b). To determine the content of the metals, a certain amount of the powders were completely dissolved in aqua regia solution and then measured by Inductively coupled plasma-optical emission spectrometry (ICP-OES, Spectro Arcos, Cleve, Germany). Chemical composition of material powders is presented in Table 1.

Ferric solutions were prepared by dissolving iron (III) chloride hexahydrate ($FeCl_3 \cdot 6H_2O$, Showa Chemicals INC., Japan, ≥97.0%) in doubly distilled water to the desired concentrations.



(a) (b)

Figure 1. The photo (a) and XRD pattern (b) of material powders

Table 1. Chemical composition of material powders

Element	Co	Ni	Cu	Fe	Mn	SiO_2
Weight percentage, wt.%	12.90	61.99	22.33	0.50	1.58	0.70

2.2. Leaching procedure

Experiments were carried out by adding a certain amount of the alloy powders into a 250 cm^3 three-neck round bottom flask containing $FeCl_3$ leaching solutions. Pulp density (the weight ratio of the powder sample to the volume of the $FeCl_3$ solution) was kept at 20 g/L, except for experiments on the effect of pulp density. The reaction mixtures were stirred at 500 rpm using a magnetic stirrer (WiseStir MSH-20D, Daihan Scienctific Co., Korea) at the desired reaction temperature and time. After the required period of time, the concentration of metals in the leaching solution were analyzed by ICP-OES. The leaching percentage (dissolution percentage) of the metals was calculated as follows:

$$Leaching\ percentage\ (\%) = \frac{Mass\ of\ a\ metal\ in\ the\ leaching\ solution}{Mass\ of\ a\ metal\ in\ the\ powder\ sample} \times 100\% \quad (1)$$

The acidity of the aqueous solutions before and after the leaching was measured by the titration method or Orion Star thermal scientific pH meter (model A221, USA). Experiments were duplicated with errors within ±5%.

3. RESULTS AND DISCUSSION

3.1. Effect of $FeCl_3$ concentration

To consider the effect of $FeCl_3$ concentration on the dissolution of metals from the alloys, leaching experiments were examined by varying $FeCl_3$ concentration from 0.3 to 1.2 mol/L with 20 g/L pulp density at 60°C for 180 min. In Figure 2, the leaching percentage of metals increased as concentration of $FeCl_3$ in the leaching solution increased. Namely, Ni, Mn, and Fe were completely leached with 0.5 mol/L $FeCl_3$, whereas the leaching percentage of Co, Cu, and Si increased from 44.6%, 46.1%, and 27.0% to 90.9%, 90.7%, and 50.0%, respectively. The dissolution efficiency of metals was ascribed to the strong oxidizing property of ferric ions (see Table 2). The coordination of dissolved metal ions with chloride ions resulting in the formation of chloro-complex ions can also contribute to a rise in the dissolution of metals (see Table 3). As is presented in Table 4, the acidity of FeCl3 solutions was enhanced with rising $FeCl_3$ concentration in the leaching solution, which could be explained by the strong hydrolysis tendency of ferric ion in aqueous solutions due to its high charge density (see Eqs. (2)-(3)) (Millero et al., 1995; Persson et al., 2018). The hydrolysis of $FeCl_3$ in the aqueous solution is represented as:

$$FeCl_{3(s)} \rightarrow Fe^{3+}_{(aq)} + 3Cl^{-}_{(aq)} \quad (2)$$

$$Fe^{3+}_{(aq)} + xH_2O_{(l)} \rightarrow [Fe(OH)_x]^{3-x}_{(aq)} + xH^{+}_{(aq)} \quad (3)$$

The slight increase in pH values (below 0.5 units) after the leaching indicated that hydrogen ions took part in the leaching of metals, except for Cu due to its high reduction potential. Therefore, leaching reactions of metals by $FeCl_3$ solutions can be expressed as:

$$M_{(s)} + 2Fe^{3+}_{(aq)} \rightarrow M^{2+}_{(aq)} + 2Fe^{2+}_{(aq)} \quad (4)$$

$$M^{2+}_{(aq)} + nCl^{-}_{(aq)} \rightarrow MCl_n^{2-n}{}_{(aq)} \quad (5)$$

Where M is a metal like Co, Ni, Mn, Cu, and Fe.

From the obtained results, 0.7 mol/L $FeCl_3$ was selected to be the optimum concentration for the leaching of metals. To completely leach metals from the alloys, the effect of other leaching parameters were studied further.

Table 2. Standard reduction potentials (E^o) of some chemical species at 25ºC (Petrucci et al., 2011).

Reduction haft-reaction	E^o, Volt
$Mn^{2+}_{(aq)} + 2e \rightarrow Mn_{(s)}$	-1.18
$Fe^{2+}_{(aq)} + 2e \rightarrow Fe_{(s)}$	-0.40
$Co^{2+}_{(aq)} + 2e \rightarrow Co_{(s)}$	-0.28
$Ni^{2+}_{(aq)} + 2e \rightarrow Ni_{(s)}$	-0.26
$Cu^{2+}_{(aq)} + 2e \rightarrow Cu_{(s)}$	+0.34
$Fe^{3+}_{(aq)} + 1e \rightarrow Fe^{2+}_{(aq)}$	+0.77

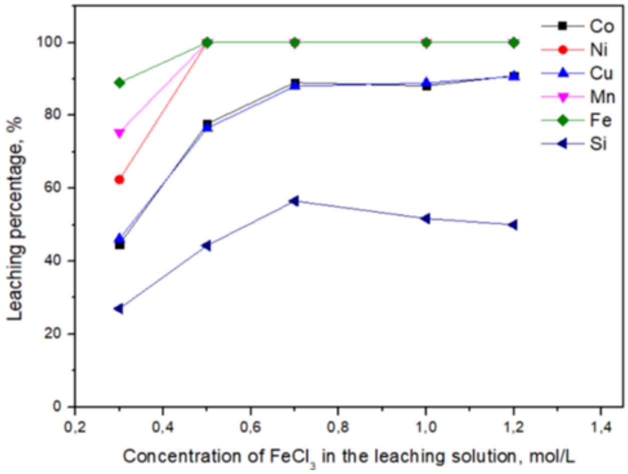


Figure 2. Effect of $FeCl_3$ concentration on the leaching efficiency of metals from the alloys. Leaching conditions: $[FeCl_3]$ = 0.3-1.2 mol/L, temperature = 60ºC, reaction time = 180 min, pulp density = 20 g/L.

Table 3. Stability constants of metal complexes with chloride ions at 25ºC (Hogfeldt, 1982; Zhao and Pan, 2011)

Reaction	logK	Reaction	logK
$Co^{2+} + Cl^- \rightarrow CoCl^+$	0.22	$Fe^{2+} + Cl^- \rightarrow FeCl^+$	-0.37
$Co^{2+} + 2Cl^- \rightarrow CoCl_2$	-3.95	$Fe^{2+} + 2Cl^- \rightarrow FeCl_2$	-1.74
$Co^{2+} + 3Cl^- \rightarrow CoCl_3^-$	-3.02	$Fe^{3+} + Cl^- \rightarrow FeCl^{2+}$	1.48
$Co^{2+} + 4Cl^- \rightarrow CoCl_4^{2-}$	-9.06	$Fe^{3+} + 2Cl^- \rightarrow FeCl_2^+$	2.13
$Ni^{2+} + Cl^- \rightarrow NiCl^+$	-1.29	$Fe^{3+} + 3Cl^- \rightarrow FeCl_3$	1.99
$Cu^{2+} + Cl^- \rightarrow CuCl^+$	0.09	$Fe^{3+} + 4Cl^- \rightarrow FeCl_4^{2-}$	0.01
$Cu^{2+} + 2Cl^- \rightarrow CuCl_2$	-0.68	$Mn^{2+} + Cl^- \rightarrow MnCl^+$	0.60
$Cu^{2+} + 3Cl^- \rightarrow CuCl_3^-$	-1.50	$Mn^{2+} + 2Cl^- \rightarrow MnCl_2$	+0.20
$Cu^{2+} + 4Cl^- \rightarrow CuCl_4^{2-}$	-2.69		

Table 4. Change in the initial and final pH of the solution with $FeCl_3$ concentration.

Concentration of $FeCl_3$, mol/L	0.3	0.5	0.7	1.0	1.2
Initial pH	1.29	1.15	0.94	0.76	0.66
Final pH	1.68	1.55	1.42	1.03	0.87
$\Delta[H^+]$, mol/L	0.03	0.04	0.08	0.08	0.08

**Note: $\Delta[H^+]$ is concentration of hydrogen ions that participated in the leaching reactions.*

3.2. Effect of reaction temperature

The effect of reaction temperature on the dissolution of metals from the alloys was undertaken in the range from 22–80°C by fixing 0.7 mol/L $FeCl_3$ concentration, 20 g/L pulp density, and 180 min reaction time. In Figure 3, Ni, Mn, and Fe from the alloys were quantitatively leached in the range of studied temperature, whereas the leaching percentage of Co, Cu, and Si increased from 80%, 85%, and 47.1% to 94%, 98%, and 56.5%, respectively. These results displayed that temperature insignificantly affected the dissolution efficiency of metals. Thus, to decrease the energy consumption, as well as the operation cost, the reaction temperature of 22°C was considered to be the best condition for the dissolution of metals.

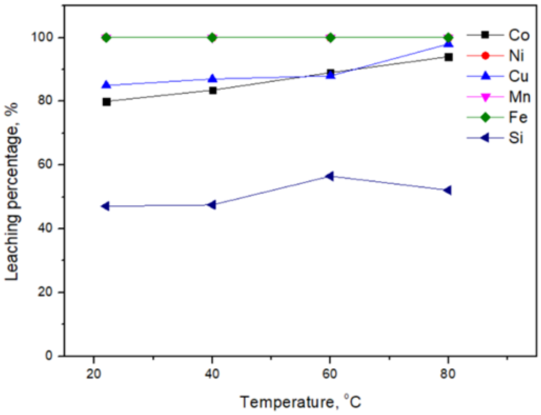


Figure 3. Effect of reaction temperature on the dissolution efficiency of metals from the alloys. Conditions: [$FeCl_3$] = 0.7 mol/L, temperature = 22–80°C, reaction time = 180 min, pulp density = 20 g/L.

3.3. Effect of reaction time

The effect of reaction time on the dissolution efficiency of metals from the alloys was conducted from 30 to 360 min. Experiments were fixed at 0.7 mol/L $FeCl_3$, 20 g/L pulp density at 22°C. In Figure 4, most Ni, Mn, and Fe were leached within 30 min, while the leaching percentage of other metals increased from 80.1% to 82.4% for Co, 84.1% to 87.2% for Cu, and from 35.2% to 49.9% for Si with increasing the reaction time to 360 min. Thus, the leaching reaction of metals by $FeCl_3$ solution occurred fast. Thus, the optimal reaction time for the dissolution was selected to be 30 min.

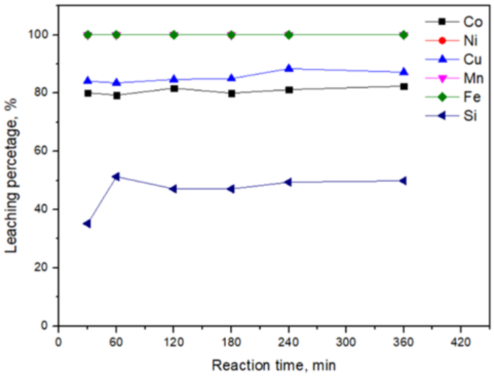


Figure 4. Effect of reaction time on the dissolution efficiency of metals from the alloys. Conditions: [$FeCl_3$] = 0.7 mol/L, temperature = 22°C, reaction time = 30–360 min, pulp density = 20 g/L.

3.4. Effect of pulp density

To completely disolve the metals from the alloys by $FeCl_3$ solution, pulp density was decreased from 20 g/L. All experiments were done at 0.7 mol/L $FeCl_3$ and 22°C for 30 min. As it is presented in Figure 5, the dissolution efficiency of metals significantly rose as the pulp density decreased. The metals were completely leached at 12.5 g/L pulp density, whereas the leaching percentage of Si was 40.6%.

Thus, the best conditions for the complete dissolution of metals from the alloys by $FeCl_3$ solution were 0.7 mol/L $FeCl_3$, 12.5 g/L pulp density, 30 min reaction time, and 22°C. The concentration of the dissolved metal ions and Si(IV) in the solution under the best conditions are displayed in Table 5. The final pH (pH_f) of the solution was determined to be 1.42.

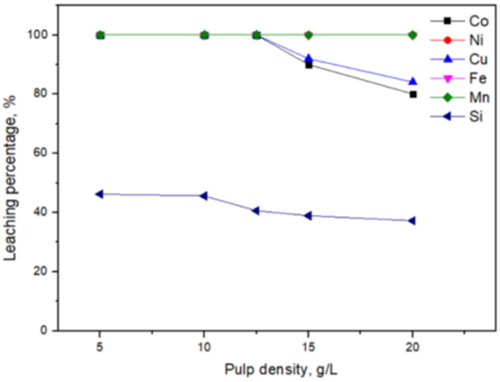


Figure 5. Effect of pulp density on the dissolution efficiency of metals from the alloys. Leaching conditions: [$FeCl_3$] = 0.7 mol/L, temperature = 22°C, reaction time = 30 min, pulp density = 5-20 g/L.

Table 5. Concentration of metals in the $FeCl_3$ leaching solution under the best leaching conditions (0.7 mol/L Fe(III), 12.5 g/L pulp density, 30 min at 22ºC)

Concentration of metal ions	Co(II)	Ni(II)	Cu(II)	Total iron	Mn(II)	Si(IV)
$FeCl_3$, mg/L	1643.0	7908.2	2820.1	40286.4	205.4	25.0

In our previous studies, the best conditions for the dissolution of metals from the alloys by HCl or H_2SO_4 solutions were 2.0 mol/L acid, 10% (v/v) H_2O_2 with 25 or 30 g/L pulp density, and 150 or 240 min reaction time at 60ºC (Tuan et al., 2021; Tuan et al., 2022c). The use of $FeCl_3$ solution for the dissolution of metals showed some advantages as compared with that of acidic solutions such as fast reaction kinetics (30 min), low energy consumption (22ºC), and low consumption of acids and oxidative agents. With its advantages, the application of $FeCl_3$ as a lixiviant for the leaching of metals could improve the process economics in the recovery of metals from the spent LIBs. Furthermore, the separation of Co(II), Ni(II), Cu(II), Mn(II), Fe(II), Fe(III), and Si(IV) from the leachate can be obtained by using some extractants such as D2EHPA for the extraction of iron, Cyanex 301 or ALi-Cy301 for Cu(II), ALi-SCN for Co(II), oxidative precipitation for Mn(II) to MnO_2 by NaClO solution, and Ni(II) selectively precipitated over Si(IV) by NaC_2O_4 (Tuan et al., 2021c, Moon et al., 2021).

4. CONCLUSIONS

The dissolution ability of ferric chloride solution for Co, Ni, Cu, Mn, Fe, and Si present in the alloys produced from spent LIBs smelted reduction was investigated. In the leaching reactions, ferric ions acted as a strong oxidizing agent for the dissolution of metals, whereas chloride ions as ligands coordinated with the dissolved metal ions to form complexes. Moreover, the hydrogen ions generated from the hydrolysis of ferric ions also contributed to the dissolution of metals. The best conditions for the complete dissolution of metals from the alloys were 0.7 mol/L $FeCl_3$, 12.5 g/L pulp density, 22ºC reaction temperature, and 30 min. Compared with either H_2SO_4 or HCl leaching systems, the use of ferric chloride as a lixiviant for the leaching of metals showed low consumption of acid and oxidizing agents, fast reaction rates, and low energy consumption.

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