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Recovery of Rare Earth Metals (REMs) from Nickel Metal Hydride Batteries of Electric Vehicles

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Abstract: Nickel metal hydride (NiMH) batteries are extensively used in the manufacturing of portable electronic devices as well as electric vehicles due to their specific properties including high energy density, precise volume, resistance to overcharge, etc. These NiMH batteries contain significant amounts of rare earth metals (REMs) along with Co and Ni which are discarded due to illegal dumping and improper recycling practices. In view of their strategic, economic, and industrial importance, and to mitigate the demand and supply gap of REMs and the limited availability of natural resources, it is necessary to explore secondary resources of REMs. Therefore, the present paper reports a feasible hydrometallurgical process flowsheet for the recovery of REMs and valuable metals from spent NiMH batteries. More than 90% dissolution of REMs (Nd, Ce and La) was achieved using 2 M H_2SO_4 at 75 °C in 60 min in the presence of 10% H_2O_2 (v/v). From the obtained leach liquor, the REMs, such as Nd and Ce, were recovered using 10% PC88A diluted in kerosene at eq. pH 1.5 and O/A ratio 1/1 in two stages of counter current extraction. La of 99% purity was selectively precipitated from the leach liquor in the pH range of 1.5 to 2.0, leaving Cu, Ni and Co in the filtrate. Further, Cu and Ni were extracted with LIX 84 at equilibrium pH 2.5 and 5, leaving Co in the raffinate. The developed process flow sheet is feasible and has potential for industrial exploitation after scale-up/pilot trails.

Keywords: NiMH batteries; rare earth metals; leaching; solvent extraction; precipitation

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1. Introduction

Rare earth metals (REMs) are called critical raw materials due to their economic, strategic and industrial importance [1]. The unavailability of the primary resources for these REMs and supply restrictions imposed by China, compelled the researchers to explore secondary resources for the extraction of REMs [2,3]. In this connection, e-waste is one of the fastest emerging areas for the generation of secondary resources containing REMs [4,5]; e-waste contains a several fold higher concentration of REMs in comparison to their availability in natural resources [6]. One of the most prominent secondary resources of e-waste is nickel metal hydride (NiMH) batteries, which are extensively used in portable electronic devices (e.g., notebook, computers, video recorders, mobile phones, etc.) as well as heavily in electric vehicles (EVs) due to their high energy density, specific volume, resistance to overcharge and no electrolyte consumption during charge and discharge [7,8]. Therefore, NiMH batteries are a potential secondary resource of rare earth elements as well as of cobalt and nickel. The proper recycling of NiMH batteries will not only add economic value but also protect the environment.

Several authors have reported various recycling methods for the recovery of REMs and other valuable non-ferrous metals from spent NiMH batteries. Three different paths (i.e., mechanical processing, pyrometallurgy and hydrometallurgy) have been employed for the recovery of metallic values from spent NiMH batteries [9]. Mechanical processing, usually consisting of grinding, sieving and bed elutriation, is carried out to recover valuable materials [10]. In pyrometallurgy, thermal separation of REMs, such as La, Ce, Nd and Pr in oxidized form, is carried out from spent NiMH batteries [11]. Among the three paths, hydrometallurgy is most suitable and appropriate for the refining and recovery of REMs from spent NiMH batteries [12–14].

Different hydrometallurgical processes, such as leaching [15–17], solvent extraction and precipitation [18–20], using various lixiviants, extractants and precipitants, respectively, have been reported for metal extraction from spent NiMH batteries. Yang et al. [20] reported the leaching of REMs using HCl from waste NiMH batteries. Other impurities were removed using ammonia solution and 99% rare earth oxides were recovered [20]. Bertuol et al. [18] reported the sulfuric acid leaching of REMs followed by their selective precipitation using NaOH at pH 1.2. Fernandes et al. [21] reported the chloride leaching of REMs from NiMH batteries followed by their selective extraction using PC88A. Innocenzi and Veglio [19] also studied the sulfuric acid leaching of REMs from spent NiMH batteries followed by their selective precipitation using NaOH at pH below 2. Zhang et al. [22] reported the chloride leaching of REMs followed by solvent extraction using D2EHPA. Xia et al. [23] reported recovery of REMs by a method of leaching using H_2SO_4, followed by solvent extraction using Cyanex 923. Finally, REM oxide was obtained using oxalic acid [23]. Two different extractants, D2EHPA diluted in kerosene [24] and Cyanex923 [25], have been used for the extraction of REMs from spent NiMH batteries. The method of chemical precipitation is most used for REM recovery owing to its ease of operation and low cost [20,26]. The use of anhydrous sodium sulfate [27] and sodium hydroxide [19] has been reported for the recovery of REMs from spent NiMH batteries. Pietrelli et al. [28] studied sulfuric acid leaching of REMs, Ni, Co, Cd and Fe from NiCd and NiMH batteries. Ahn et al. [29] reported the recovery of REMs in the form of their mixed oxides using a leaching and precipitation process. However, the selective extraction of Ce, La and Nd from the leach liquor of NiMH has not been discussed in this paper.

The literature review suggests that the precipitation process is not selective for the separation of REMs (e.g., La, Ce and Nd). A combined process, consisting of solvent extraction and precipitation, is needed to be developed to selectively extract the REMs. Therefore, the present study reports a novel hydrometallurgical process consisting of leaching, solvent extraction and precipitation for recovery of REMs, including valuable metals, such as Cu, Ni and Co, in a closed-loop system, considering the zero-waste discharge concept. REMs were first dissolved in sulfuric acid and thereafter Ce and Nd were extracted with PC88A. Subsequently, La was selectively precipitated in a range of pH 1.5 to 2. Further, Cu and Ni were selectively extracted using LIX 84 leaving Co in the raffinate. The developed process flow sheet is novel for the selective extraction of REMs.

2. Experimental

2.1. Materials

Discarded nickel metal hydride (NiMH) batteries of electric vehicles were collected locally to be used for the experimental purpose. Initially, discarded NiMH batteries were discharged using a saturated solution of sodium chloride. Then, NiMH batteries were pretreated by a crushing and flotation process to separate the black powder, metallic fraction and plastic materials, as shown in Figure 1. The black powder was dissolved in aqua regia to analyze the concentration of metals. The typical composition of black powder is shown in Table 1.

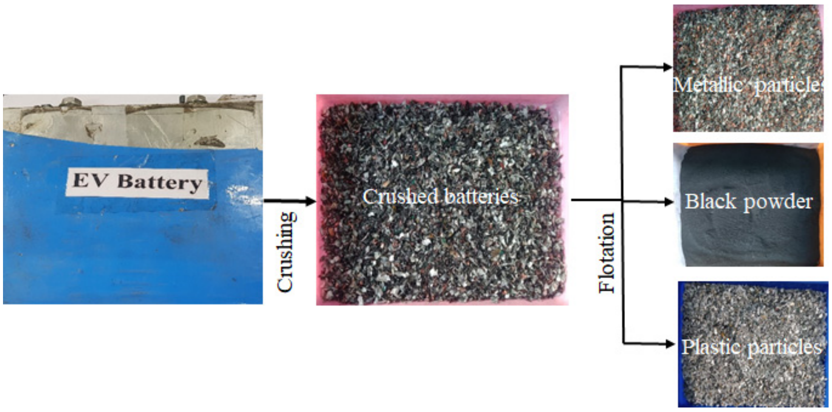

Figure 1. Pre-treatment of discarded electric vehicle batteries.

Table 1. Elemental composition by weight (wt. %) of the black power of an NiMH battery used in electric vehicles.

Elements	La	Ce	Nd	Cu	Ni	Co	Other
Contents	7.7	12	4.0	6.5	29	5.3	balance

2.2. Methodology

2.2.1. Leaching Procedure

Leaching experiments were carried out in a three-necked Pyrex glass reactor (1 L capacity) fitted with a condenser to condense back the gases evolved during the leaching studies (Figure 2). Hotplates with a temperature sensor and magnetic stirrer facilities were used during leaching experiments. The various concentration of sulfuric acid (0.5–2.0 M) was used as a leachant to dissolve the metals at different temperatures, varying from 25 to 75 °C. When the solution attained the desired temperature, a certain amount of black powder was poured into the solution to leach the metals. Further, the slurry was processed for filtration to obtain leach liquor and leached residue. The dissolved metals present in the leach liquor and remaining in the residue were analyzed using an atomic absorption spectrometer (AAS, AAnalyst 200, Perkin Elmer, Waltham, MA, USA) and inductively coupled plasma-optical emission spectroscopy (ICP-OES, VISTA-MPX, Varian Inc., Victoria, Australia).

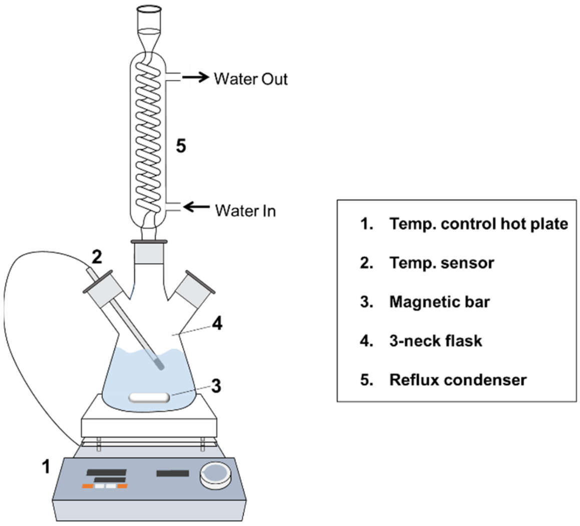

Figure 2. Experimental setup for leaching of REMs from discarded NiMH batteries.

2.2.2. Solvent Extraction Procedure

Liquid-liquid extraction studies were carried out in a beaker fitted with a magnetic stirrer using an equal volume of leach liquor and extractant (25 mL/25 mL). Two types of extractants were used, i.e., 2-ethylhexyl 2-ethylhexyphosphonic acid (PC88A, Figure 3a) for REMs and 2-hydroxy-5-nonylacetophenoneoxime (LIX 84, Figure 3b) for base metals. Kerosene and isodecanol were used as a diluent for extractant and a phase modifier, respectively. Ammonium hydroxide and dilute sulfuric acid were used to maintain the equilibrium pH (eq. pH) in the range of 1 to 5. When the solution attained equilibrium, the organic and aqueous phases were separated using a separating funnel and the concentrations of metal ions in the raffinate solution were analyzed using ICP-OES. The concentrations of metals in the organic phase were calculated.

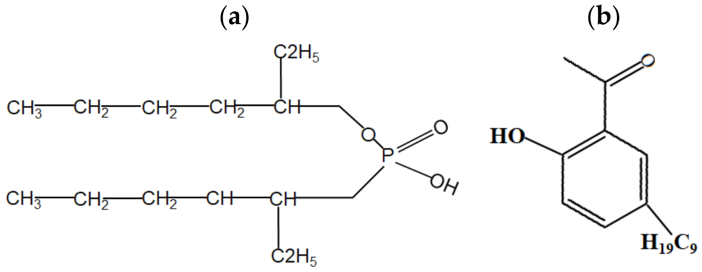

Figure 3. Structure of (**a**) PC88A and (**b**) LIX 84.

2.2.3. Precipitation Procedure

Precipitation studies were carried out in a beaker (capacity: 100 mL) under constant stirring speed at room temperature using ammonium hydroxide as a precipitant. During the experiments, solution pH was varied between 0.75 and 2.0 by adding ammonium hydroxide to precipitate La as lanthanum hydroxide. The solution was taken at different intervals of time to analyze the concentrations of metals in leach liquor after precipitation.

2.2.4. Characterization and Analysis of Samples

X-ray diffraction (XRD, D8, Bruker AXS Inc., Wisconsin, WI, USA) was used to analyze the phases of samples. Figure 4 shows that XRD patterns mainly contained the peak of oxides of rare earth metals, Cu and Ni in black powder of NiMH batteries. Further, AAS and ICP-OES were used to analyze the concentrations of metal ions in the leach liquor.

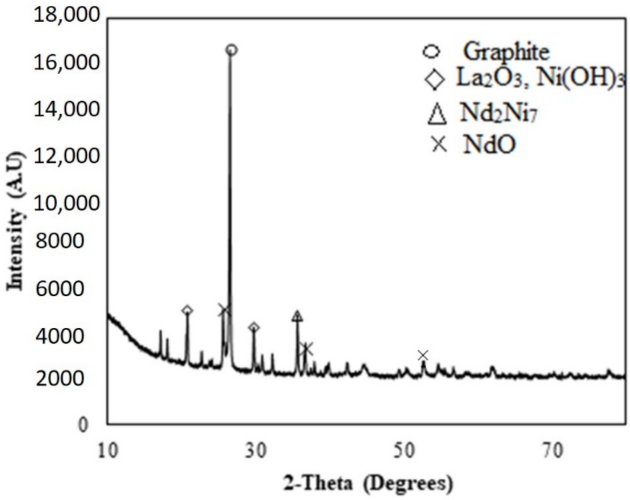

Figure 4. XRD pattern of black powder obtained from NiMH batteries.

3. Results and Discussion

Experiments were carried out using leaching followed by solvent extraction and precipitation to extract the REMs. The obtained results are discussed below:

3.1. Leaching Studies

To dissolve the metals and optimize the process parameters (i.e., acid concentration, time, temperature, pulp density, etc.), the leaching experiments were carried out varying the scale of the parameters.

3.1.1. Effect of Acid Concentration

To optimize the minimum acid requirement during the leaching experiments, studies were performed varying the sulfuric acid concentration in a range of 0.5 to 2.0 M H_2SO_4 at 75 °C for mixing time 75 min at pulp density 100 g/L in presence of 10% H_2O_2 (v/v).

The results (Figure 5a–f) showed that leaching of La, Ce, Nd, Cu, Ni and Co increased with increase in the concentration of acid, which happened due to the increase in acidic strength. It was found that 99.1% La, 96.1% Ce, 99.1% Nd, 99.9% Cu, 99.9% Ni and 99% Co leached in 2 M H_2SO_4 at 75 °C in 60 min. Further, the increase in acidic concentration had no significant effect on the enhancement of REM leaching efficiency. Therefore, 2 M H_2SO_4 was considered the optimum acid concentration for the dissolution of metals from black powder of spent electric vehicle batteries.

3.1.2. Effect of Temperature

To optimize the temperature for dissolution of REMs, experiments were carried out in a range of temperatures varying from 25 to 75 °C at a pulp density of 100 g/L in the presence of 10% H_2O_2 (v/v). Figure 6 shows that leaching of La, Ce, Nd, Cu, Ni and Co increased with increase in temperature due to the decrease in activation energy, resulting in the enhancement of metal leaching efficiency with rise in solution temperature [30]. It was found that 98.9% La, 98.5% Nd, 89.9% Ce, 99% Cu, 99% Ni and 95.9% Co leached at 75 °C. Thereafter, the increase in the solution temperature had no significant effect on the enhancement of metal leaching efficiency.

3.1.3. Effect of Pulp Density

Pulp density is an important parameter for the effective leaching of metals from solid. Therefore, to optimize the solid to liquid ratio (i.e., pulp density (S/L)), leaching experiments were carried out varying the pulp density from 50 to 200 g/L at temperature 75 °C, H_2O_2 10% (v/v) and mixing time 60 min for maximum dissolution of REMs. The results (Figure 7) showed that leaching of La, Ce, Nd, Cu, Ni and Co was found to decrease with increase in pulp density above 100 g/L. This may have been due to increase in the total number of moles of metallic constitutes (Nd, La Ce, Cu, Ni and Co) relative to available leachant molecules resulting in a decrease in the leaching efficiency of metals [31]. Therefore, 100 g/L pulp density has been chosen as the optimum condition for leaching of metals from black powder of NiMH batteries of electric vehicles.

3.2. Separation and Purification Studies

3.2.1. Solvent Extraction of Ce and Nd

Leach liquor containing 3.9 g/L Nd; 11.9 g/L Ce; 7.6 g/L La; 6.5 g/L Cu; 28.9 g/L Ni and 5.1 g/L Co was used for the solvent extraction studies using 10% PC88A to selectively separate the rare earth metals. It was found that extraction of Nd and Ce increased with increase in eq. pH, as shown in Figure 8. The maximum separation of Ce and Nd with respect to La, Cu, Ni and Co occurred at eq. pH~1.5. The separation factor (β) values of Nd and Ce were also calculated with respect to La and found to be 12.1 ($\beta_{Nd/La}$) and 56.33 ($\beta_{Ce/La}$) with respect to La. This indicates that selective extraction of Nd and Ce can be achieved by increasing the number of stages for extraction of Nd. Thus, a McCabe–Thiele plot was drawn to determine the number of stages required for the extraction of Nd from leach liquor. It was found that two counter-current stages were required to completely extract Nd from leach liquor, as shown in Figure 9.

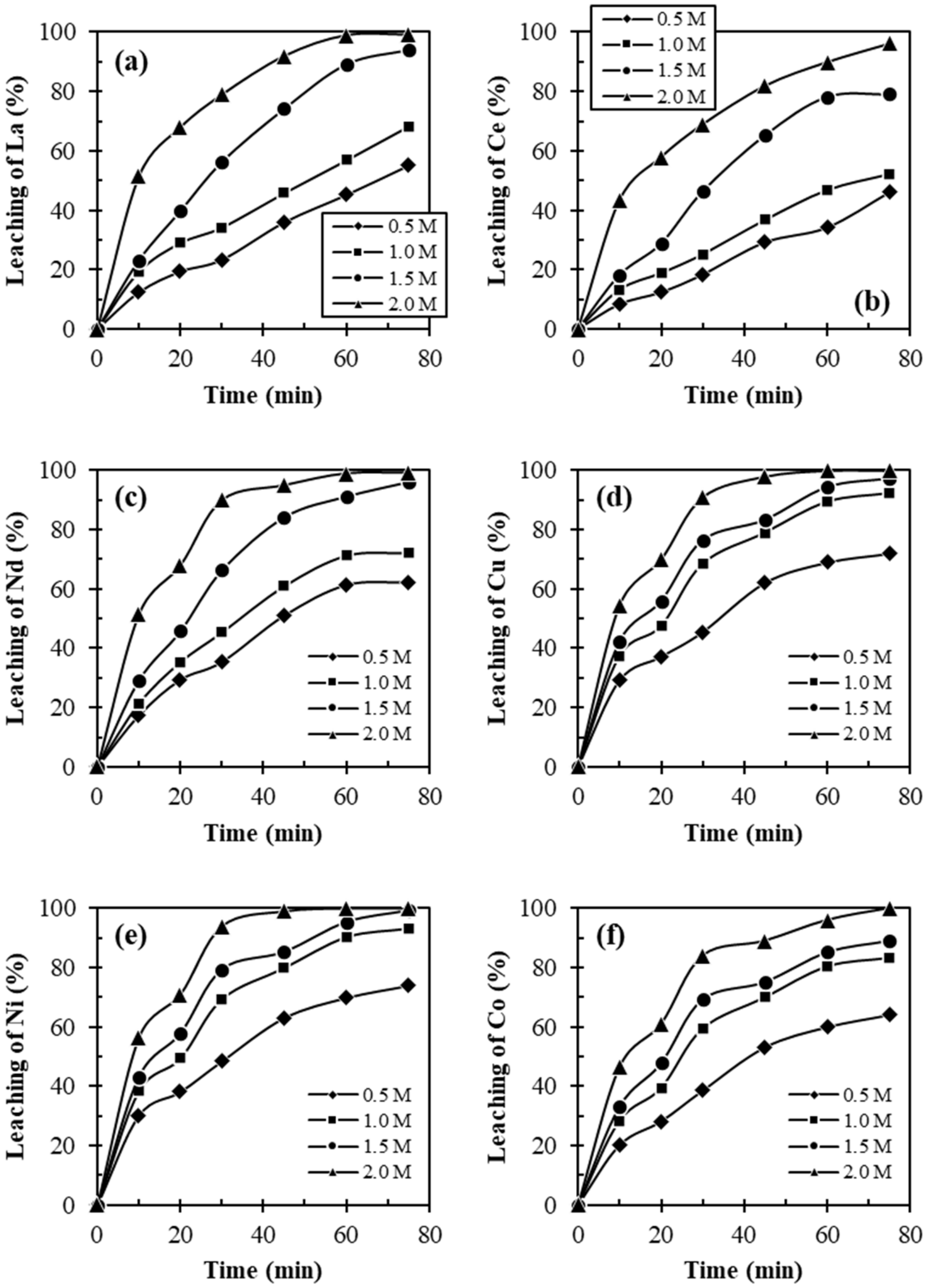

Figure 5. Effect of acid concentration on leaching of (**a**) La, (**b**) Ce, (**c**) Nd, (**d**) Cu, (**e**) Ni and (**f**) Co from black powder of spent NiMH battery of electric vehicle [Solid: black powder; liquid: 0.5–2.0 M H_2SO_4; additive: 10% H_2O_2 (v/v); temperature: 75 °C; pulp density 100 g/L; time: 75 min].

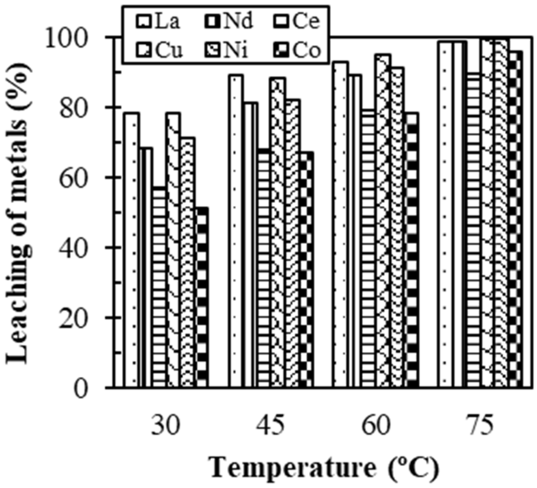

Figure 6. Effect of temperature on leaching of rare earth elements (La, Ce and Nd) and valuable metals (Cu, Ni and Co) [Solid: black powder; liquid: 2 M H_2SO_4; additive: 10% H_2O_2 (*v*/*v*); pulp density 100 g/L; time: 60 min].

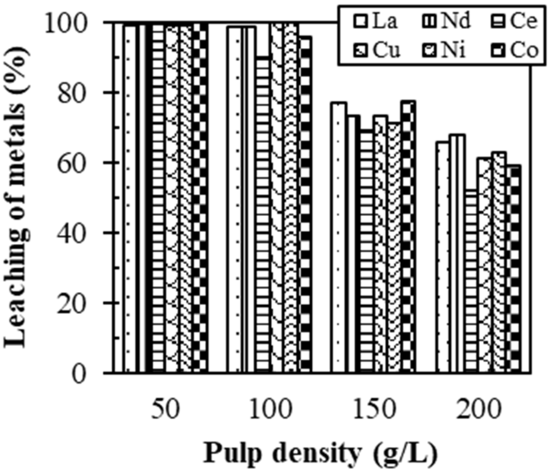

Figure 7. Effect of pulp density on the leaching of rare earth elements (La, Ce and Nd) and valuable metals (Cu, Ni and Co) [Solid: black powder; liquid: 2 M H_2SO_4; additive: 10% H_2O_2 (*v*/*v*); time: 60 min; temperature: 75 °C].

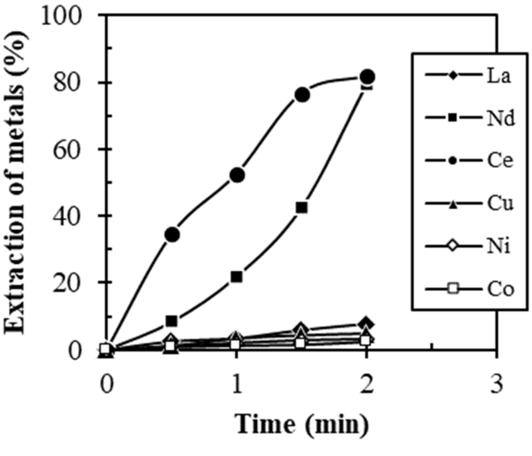

Figure 8. Solvent extraction of Ce and Nd from leach liquor [Metal concentration in leach liquor (g/L): Nd: 3.9; Ce: 11.9; La: 7.6; Cu: 6.5; Ni: 28.9; Co:5.1; extractant: 10% PC88A diluted in kerosene; time: 10 min; O/A ratio: 1/1].

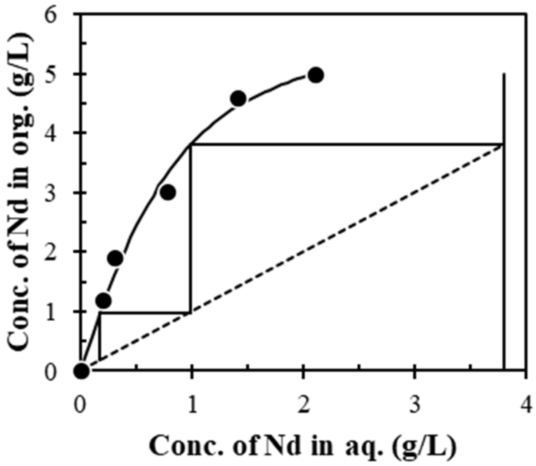

Figure 9. McCabe–Thiele plot for extraction of Nd from leach liquor [Leach liquor: 3.9 g/L Nd; extractant: 10% PC88A diluted in kerosene; time: 10 min; O/A ratio: 1/1].

3.2.2. Precipitation of La

The solution obtained after extraction of Nd and Ce that contained 7.6 g/L La; 6.5 g/L Cu; 28.9 g/L Ni and 5.1 g/L Co was used for recovery of La by precipitation process. It was found that precipitation of La increased with increase in pH as shown in Figure 10. The complete precipitation of La occurred at pH ~1.5 leaving Cu, Ni and Co remaining in the filtrate. Further, precipitation of La was validated with an Eh-pH diagram, which indicated that La precipitation occurred at or above pH 1.5 as $La(OH)_3$ [31], while Cu, Ni and Co form hydroxide above pH 5. Therefore, selective precipitation of La occurred between pH 1.5 and 2.0. Further, Cu, Ni and Co were separated by solvent extraction, as discussed below.

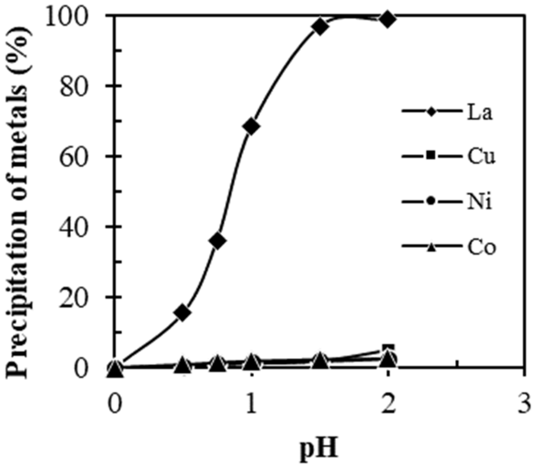

Figure 10. Precipitation of La from leach liquor [Metal concentration in leach liquor (g/L): La: 7.6; Cu: 6.5; Ni: 28.9; Co: 5.1; precipitant: ammonium hydroxide].

3.2.3. Extraction of Cu, Ni and Co

After the extraction of La, Ce and Nd, the obtained solution containing 6.5 g/L Cu, 28.9 g/L Ni and 5.1 g/L Co was used for selective extraction of Cu, Ni and Co using solvent extraction process. Solvent extraction experiments were carried out with LIX 84 varying eq. pH between 1.0 and 5.0 to extract the Cu, Ni and Co separately from leach liquor. The results (Figure 11) showed that 99.1% extraction of Cu occurred with 10% LIX 84 in a range of eq. pH 2.0 to 2.5 while Ni and Co remained in the raffinate. Further, 98.6% Ni was selectively extracted with 30% LIX 84 between eq. pH 4.5 and 5.0 at O/A ratio 1/1 in 10 min, leaving Co in the raffinate. This indicates that LIX 84 makes complexes with Cu and

Ni at different eq. pH resulting in the selective extraction of Cu and Ni. The metal-loaded organic was stripped using 10% H_2SO_4 to obtain a pure solution of Cu and Ni. Finally, a process flow sheet was developed for the extraction of La, Ce, Nd, Cu, Ni and Co, as shown in Figure 12. The metals/salts of Nd, Ce and La could be used in various applications, such as making magnets for hard disks, cathode ray tubes and phosphor powder, and pyrophoric alloys, respectively. In addition, the recovered REMs, Cu, Ni and Co can be re-used in the manufacture of rechargeable batteries. Based on the obtained results and pure metals/salts, the recovered materials of discarded NiMH can be used in different industries after scale-up/pilot trial.

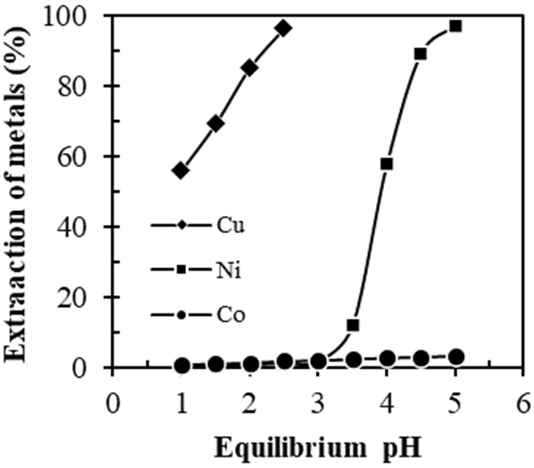

Figure 11. Solvent extraction of Cu, Ni and Co from leach liquor (Metals concentration in leach liquor (g/L): Cu: 6.5; Ni: 28.9; Co: 5.1; extractant: 10% & 30% LIX 84; time: 10 min; O/A ratio: 1/1).

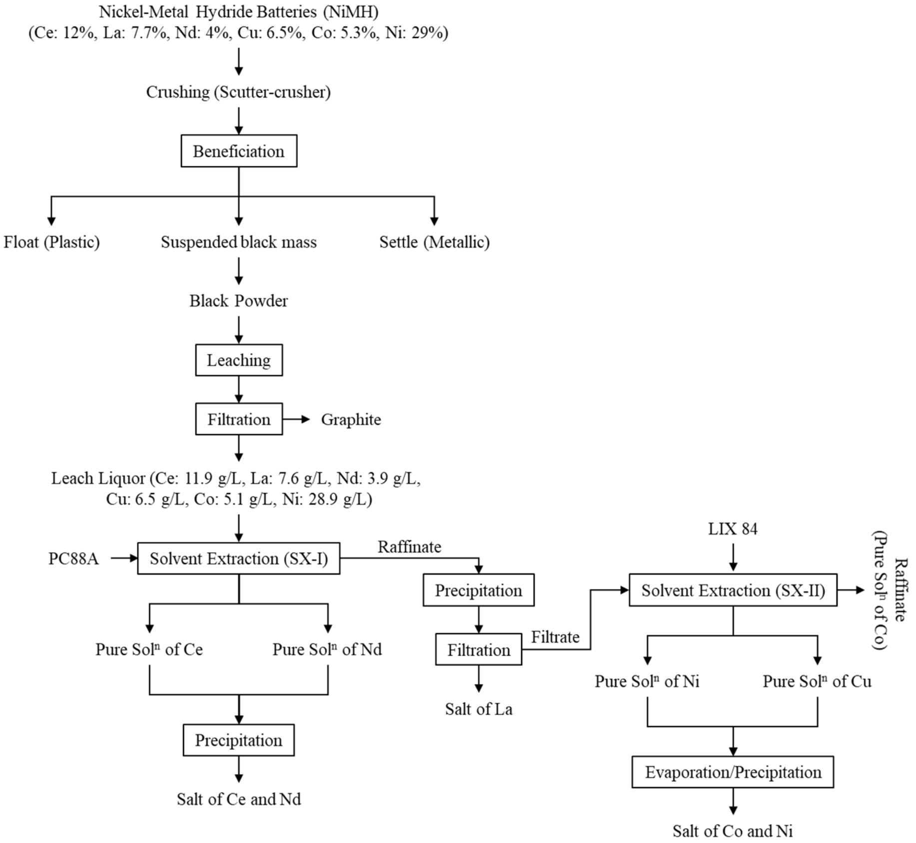

Figure 12. Developed process flowsheet for extraction of rare metals, Cu, Ni and Co from discarded NiMH batteries of electric vehicles.

4. Conclusions

Based on laboratory scale studies, the following conclusions have been drawn for the recovery of rare and valuable metals from NiMH batteries of electric vehicles:

(1) It was found that more than 98.9% La, 98.5% Nd, 89.9% Ce, 99% Cu, 99% Ni and 95.9% Co were leached in 2 M H_2SO_4 at 75 °C in 60 min in the presence of 10% H_2O_2 (v/v).

(2) Further, Nd and Ce were extracted using 10% PC 88A at equilibrium pH 1.5. A McCabe–Thiele plot showed that 99% Nd was extracted with PC 88A at eq. pH ~1.5 at O/A ratio 1/1 in two counter-current stages.

(3) After the extraction of Nd and Ce, 99% La was selectively precipitated with sodium hydroxide at room temperature between pH 1.5 and 2.0, leaving Cu, Ni and Co in the raffinate.

(4) Subsequently, Cu and Ni were selectively extracted using LIX 84 at eq. pH ~2.5 and ~5, respectively, leaving Co in the raffinate.

Author Contributions: Methodology, P.K.C. and O.S.D.; writing—original draft preparation, R.P. and P.K.C.; writing—review and editing, M.K.J., K.Y. and I.P.; supervision, M.K.J. and R.K.J.; project administration, M.K.J. and I.P.; funding acquisition, I.P. All authors have read and agreed to the published version of the manuscript.

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Data Availability Statement: The data presented in this study are available on request from the corresponding author.

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Conflicts of Interest: The authors declare no conflict of interest.

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