Recovery of Metals from Spent Lithium-ion Battery by Supercritical Carbon Dioxide with Chelating Ligand

Xiaoqian Zhao, Miyu Ozaki, Rodolfo Morales Ibarra, Wahyudiono, Hideki Kanda, Motonobu Goto

Department of Materials Process Engineering, Nagoya University, Nagoya 464-8603, Japan

1. Introduction

With the increasing demand of Lithium-ion batteries (LIBs), the supply disruption risk of metal resources and environmental performance will become a serious challenge. Hence, a clean process for recycling spent LIBs is vital for both of metal sources and environmental pollution. Solvent extraction is considered as one of the most widely used method as conventional recycling process of LIBs. However, it has many disadvantages such as the utilization of large amount of organic solvent. Compared with conventional recycling process, supercritical fluid extraction of metals has considered as a cleaner process because of reducing the amount of organic solvents. Among which, the attention is particularly focused on super -critical carbon dioxide (scCO₂). The formation of complexes of metal ions with organic matter reduces their polarity, allowing them to be easily dissolved in scCO₂ for efficient extraction.

Here, separation of valuable heavy metal such as Co from LIBs using scCO₂ with chelating ligand was investigated.

2. Experimental Methods

Here, the feasibility of a semi-batch system designed to extract Co using chelating ligand is investigated. Fig. 1 shows the schematic diagram of experimental extraction process. The pressurized scCO₂ was delivered to the extractor I containing chelating ligands, leaching solution of metals was loaded in extractor II. The saturated scCO₂ with ligand was delivered to extractor II and the extract flows in the vial finally. The effects of ligand category, pressure and temperature were observed. The



experiments were performed at pressure, temperature and flow rate of 10-25 MPa, 60 °C and 3.5

mol/min, respectively. The extracted product is collect

Fig. 1 Schematic diagram of extraction process

-ed in a vial every 30 minutes and analyzed by Inductively coupled plasma atomic emission spectroscopy (ICP-AES). As for ligands, 2,2,6,6-Tetramethyl 3,5-heptane -dione (THD), acetylacetone (ACAC), hexafluoro-

acetylacetone (F-ACAC), 2,2,7-trimethyloctane -3,5-dione (TMOD) and caffeine were used.

3. Results and Discussion

From the results of extraction efficiency of Li, Co and Ni under different ligands, at different ligand concentration and pressure. The relatively higher extraction efficiency and selectivity on extraction of Co are obtained at 0.01 M ligand concentration, 20 MPa of pressure and at temperature of 60 °C.The extraction efficiency was the highest for F-ACAC, whereas the selectivity was the highest for THD.

4. Conclusion

Metal extraction has been performed successfully by scCO₂ with chelating ligands. The extraction experiments using THD perform effective and are of higher selectivity than the other ligands.

The optimal experimental condition is performed at 60 °C of temperature, 20 MPa of pressure, 0.01 M of concentration of ligand solution.

Acknowledgement

This work was supported by JST-Mirai Program, Japan.