XVI International Mineral Processing
and Recycling Conference
28-30 May 2025, Belgrade, Serbia

DOI: 10.5937/IMPRC25298M

Original research article

THE RECYCLING OF PLASTICS AND METALS FROM END-OF-LIFE NMC-TYPE ELECTRIC VEHICLE LITHIUM-ION BATTERIES USING SELECTIVE FROTH FLOTATION

Fulya Mennik[1], 0009-0004-9883-7833,
Nazlım İlkyaz Dinç[2], 0000-0003-0720-667X,
Beril Tanç Kaya[3], 0000-0002-1757-0566,
Fırat Burat[1#], 0000-0001-7051-0063,
[1]Istanbul Technical University, Faculty of Mines, Mineral Processing Engineering Department, Istanbul, Türkiye
[2]Istanbul Technical University, Istanbul, Türkiye
[3]Istanbul Technical University, Faculty of Mines, Geological Engineering, Istanbul, Türkiye

ABSTRACT – The increase in end-of-life (EoL) Lithium-ion Batteries (LIBs) in recent times has become a major concern for the global community. Most of the research in the literature has focused on the recovery of cathode active metals from black mass. The separation of anode-cathode foils, plastics, and casing metals, which are essential parts of LIBs, remained limited. To reduce costs and maximize the recovery of valuable metals in the subsequent hydro or pyro metallurgical processes EoL LIBs need to be pre-treated accordingly. The main purpose of this study is to scrape off the black mass (or battery dust) adhering to the electrode foils as a result of gradual crushing and then to separate the plastic and copper from other metals by two-step selective flotation. The results showed that plastics with natural hydrophobicity could be removed with the help of a frother. After plastic flotation, the copper particles were floated in the presence of Aerophine 3418A, and a copper concentrate assaying 65.13% Cu was obtained with 96.4% Cu recovery at the end of the copper flotation circuit. In addition, the Al content in the material that could not float and remained in the cell was increased to approximately 77%.

Keywords: Lithium-ion Battery, Plastics, Copper, Aluminum, Recycling.

INTRODUCTION

Developments in energy storage technologies have caused significant transformations across various sectors and have become crucial for achieving a sustainable future. Moreover, the rising energy demand, the increasing use of renewable energy sources, and the extensive of electric vehicle (EV) technologies are contributing to the need for advanced energy storage systems. In particular, the high use rates of fossil fuels and the need to reduce environmental damage have led users to want to develop

---

[#] corresponding author: buratf@itu.edu.tr

renewable and sustainable energy sources such as high-quality batteries, wind turbines, fuel cells, and solar panels [1].

Lithium-ion batteries can be identified by their compact size, lightweight design, high energy density, and voltage, expanded lifespan, absence of memory effect, efficient operation across a wide temperature range, and eco-friendly characteristics [2]. The incorrect disposal of discarded LIBs in the environment results in significant issues. The pollution of soil and water can be caused by the leakage of heavy metals and compounds from batteries. In addition to its environmental and economic benefits, the exploitation of the resource potential of scraps is significant for the conservation of natural resources and the sustainable development of metals and related industries [3, 4]. Besides, EoL LIBs contain many components (such as Li, Co, Cu, Al, Ni, Fe, Mn, and graphite) with high economic value [5]. LIBs consist of several fundamental components, each serving distinct functions. Each component is essential in influencing the energy storage capacity, safety, and lifespan of the battery. Alongside the cathode and anode, LIBs comprise additional components including the electrolyte, separator, current collectors, and casing. The structure of a standard battery module is illustrated in Figure 1 [1].

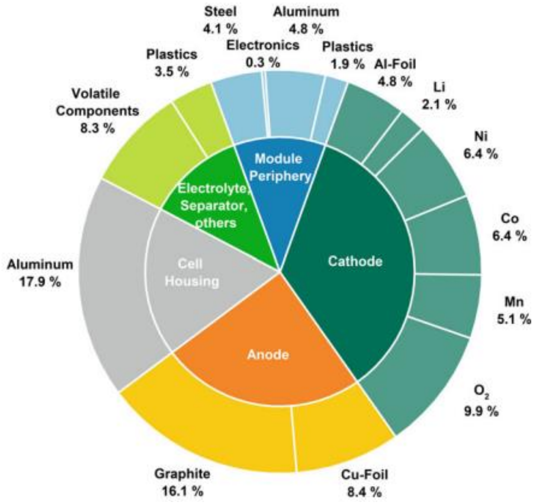

Figure 1 Components of LIBs [1]

Recycling LIBs enables the recovery of critical metals. The result of resource management reduces dependency on mining activities, protects natural resources, and supports sustainable production models. To recycle S-LIBs, different techniques are used, such as physical (dismantling, comminution, classification, gravity, magnetic, and electrostatic separation), physicochemical (flotation), hydrometallurgical (chemical precipitation, acidic leaching, electrochemical processes), and pyrometallurgical (roasting, thermal treatment). Nevertheless, the composition of LIBs is distinct from that of a natural ore due to their unique properties. Examining identical LIBs maintains the same physical properties, material amount, and type. On the other hand, different types of LIBs have different chemical structures, so the way they are recycled may be different depending on the type of battery [6, 7].

While flotation studies mostly focused on the separation of graphite from the black mass the selective recovery of coarse fractions including current collector metals (Cu and Al), case metals (Fe and Ni), plastics, and separators is very limited. Therefore, this study has aimed at the selective separation of plastics, Cu, Al, and case metals from NMC-type Lithium-ion batteries using different surface-active reagents. After separating the black mass from the metallic fraction through four stages of size reduction and sieving, in the first flotation stage, plastics with natural floatability were selectively removed with the help of a frother reagent (MIBC). In the second flotation stage, the effects of different collectors on copper flotation were investigated by collecting hydrophobic copper particles in float product and leaving the hydrophilic Al and case metals particles in the cell.

EXPERIMENTAL

In this study, approximately 60 kg of NMC-type EoL EV LIBs were supplied from Exitcom Co. located in Kocaeli, Türkiye. A multistage size reduction and classification system was applied and shown in Figure 2. The batteries were first fully discharged for 48 h with 5% NaCl solution, air dried, and then subjected to primary crushing using a dual shaft shredder in the recycling company to guarantee safety conditions. The crushed material was sent to the Mineral Processing Engineering Department Pilot Plant in Istanbul Technical University for subsequent comminution processes. The black mass 1 sample was separated using a 0.2 mm sieve and fed to a four-blade cutting mill (RAM200 model supplied from RANTEK Co.) for further scraping off the remaining black mass on collecting electrodes. The cutting mill was engineered to crush uniformly elastic, soft, medium-hard, fibrous, and heterogeneous materials. When the particles are reduced in size by shear forces, the material maintains its flat surface shape, which creates an advantage for the flotation process. The crushed material's size was adjusted by utilizing a replaceable sieve (6, 4, 2 mm, etc.,) located in the lower compartment of the mill. The black mass adhering to the surfaces of the current collectors was further removed using a 0.2 mm sieve and reserved for future studies. The coarse material (-2+0.2 mm), which constitutes about half of the feed, is mainly composed of copper foils, aluminum foils, battery casings, plastics, and separators. As seen in Figure 3, the d_{80} and the d_{50} sizes of the -2+0.3 mm fraction were determined as 1 mm and 0.65 mm, respectively. The coarse fraction accounts for about 32% of the feed, while the remaining amount belongs to the black mass.

Before flotation experiments, 50 g of the sample was weighed and added to 1000 mL of tap water, then mixed at 1500 rpm for 15 min to remove the remaining black mass. The dispersed black mass was removed through a 74 µm screen and the over-screen material was again fed to a 1.5 L self-aerated Denver-type flotation machine with an impeller speed of 1200 rpm. No pH adjuster was used to create a more environmentally friendly process. City tap water was used for all experiments and the natural pH of the pulp was around 8.3. After the reagents were sufficiently contacted with the particles, the air valve (3 L/min air flow rate) was opened and the floating particles were collected

(3 min flotation time). First-stage flotation experiment was carried out to separate the plastic fraction from metals. MIBC was preferred as the frother reagent in plastic flotation and no collector reagent was required. After the separation of plastics, the copper particles in the sink product were floated with the help of different collectors (KAX (Solvay Group), Aerophine-3418a, Aero-3739 (Syensqo), Aerofloat-242, and Aerofloat-211).

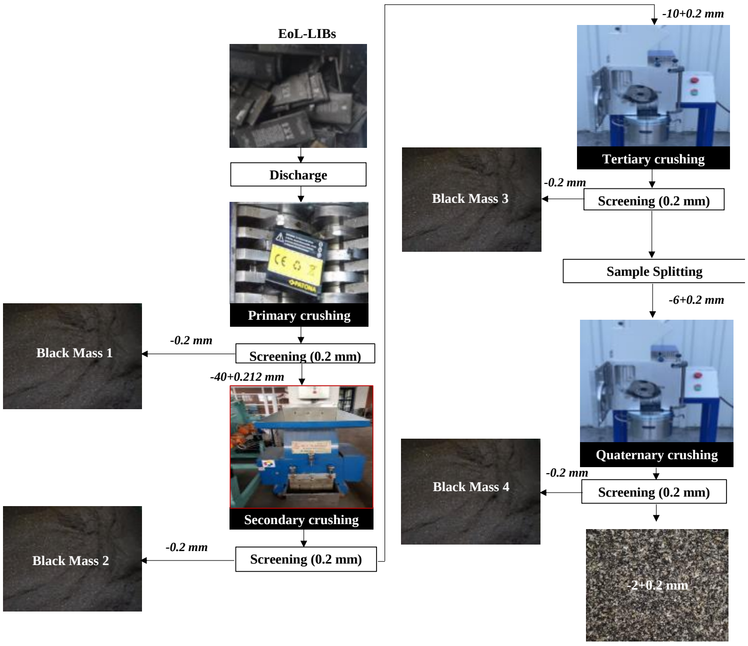

Figure 2 Flowchart of the multi-stage crushing process

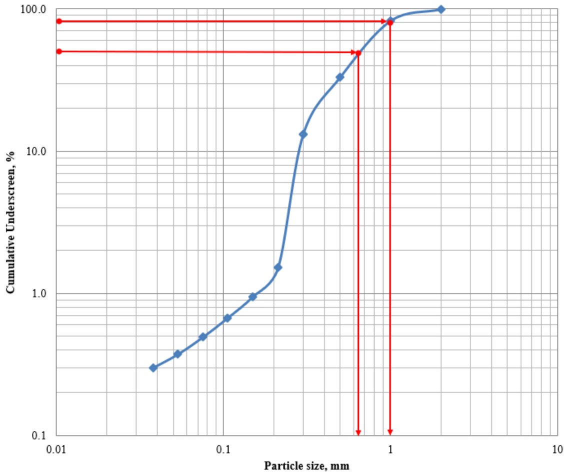

Figure 3 The particle size distribution of -2+0.2 mm fraction

The experimental data were interpreted based on the content and recovery of the products. The metal contents and distribution of metals were determined through chemical analysis. The XRF method was employed to determine the chemical properties of the LIBs sample in the geochemistry research laboratory at I.T.U. The analysis of the products was repeated at least two times and the weighted averages were calculated. The margin of error was around ±1.5%.

RESULTS AND DISCUSSION

The chemical modification of particle surfaces can regulate their hydrophobic or hydrophilic properties [8]. Plastics are known for their natural hydrophobic and their specific gravity is very low compared to metals. In froth flotation, a selective separation is made based on the hydrophobicity differences between the particles. Plastic flotation tests were carried out in 5 stages (no frother at the first stage and then 4x100 g/t MIBC). Without any frother, about 15% of the total plastics were floated and about 60% of the plastic particles were floated with the addition of 200 g/t of MIBC. The cumulative weight of the floated plastics systematically increased to 95% at 400 g/t. Although the surfaces of Al particles are more hydrophobic than Cu surfaces before the reagent addition, the wettability of Cu increases and exceeds that of Al with the addition of collectors, which were originally used in the flotation of natural copper ores [6]. The amount of collector concentration was fixed to 3x1000 g/t for the copper stage. Metal contents and recoveries of the flotation experiments are given in Table 1.

It is clear from Table 1 that approximately 16% of the material that was fed to the first flotation stage was removed as a plastic product. Approximately 5% by weight of the fine-sized Al particles, which have a greater floatability than Cu, were transferred into the floating product. This value is around 1.5% for Cu which has lower floatability over Al particles. A small portion of the cathode active materials still attached to the Al passed into the plastic product with this metal. With a cleaning flotation, the final plastic product can be obtained with lower metal contents. A copper concentrate assaying 65.13% Cu was successfully obtained with a 96.4% Cu recovery rate. Another commercial reagent which is mostly preferred in the flotation of natural copper ores, KAX, provided a successful flotation and a Cu concentrate assaying 62.47% Cu was produced with 95.8% Cu recovery. The image of floating copper particles is presented in Figure 4.

Although the other three collectors obtained concentrates with similar Cu contents, their recovery rates decreased due to the low amount of floating material. If the evaluation is made according to Al content, the best performance is again shown by the 3418A collector. A sink product with approximately 77% Al content was obtained with 71.6% Al recovery. The reason for the low Al recovery efficiency compared to Cu is the loss of small-sized Al particles mixed with the copper concentrate. To increase both Cu content and Al recovery, it can be suggested to reduce the floating ability of the Al particles using NaOH pre-treatment and feed to a cleaning circuit. Another solution can be to transform the particles into spherical shapes using a proper grinder and then to separate two metals using an electrostatic or Eddy current separator. After copper flotation, the sinking product with high Al content remaining in the cell cannot float and can be separated from the magnetic case metals accompanying this metal with the help of a magnetic separator. In this way, both the Al content is increased and a marketable product with high Fe and Ni content can be produced.

Table 1 Metal content and recoveries of flotation

Collector Type	**Products**	**Amount %**	**Cu**		**Al**		**Co**		**Ni**		**Mn**		**LOI**	
			C, %	**R, %**	**C, %**	**R, %**	**C, %**	**R, %**	**C, %**	**R, %**	**C, %**	**R, %**	**C, %**	**R, %**
KAX	Plastics	14.3	1.25	0.7	12.70	4.7	4.58	10.4	1.30	10.7	1.28	11.8	74.29	48.8
	Black Mass	8.5	1.24	0.4	6.30	1.4	20.18	27.3	5.66	27.6	5.14	28.1	50.96	19.9
	Copper	38.8	62.47	95.8	22.25	22.3	2.25	14.0	1.02	22.8	0.95	23.8	8.53	15.2
	Aluminium	38.5	2.02	3.1	71.84	71.6	7.84	48.3	1.75	38.9	1.46	36.3	9.03	16.0
	Total	100.0	25.28	100.0	38.63	100.0	6.25	100.0	1.73	100.0	1.55	100.0	21.69	100.0
3418A	Plastics	15.7	1.41	0.9	12.68	5.1	4.96	12.5	1.58	13.9	1.46	14.5	73.79	54.0
	Black Mass	10.0	1.33	0.5	6.28	1.6	19.96	32.0	5.51	30.7	5.41	34.2	51.81	24.1
	Copper	37.9	65.13	96.4	21.12	20.7	2.03	12.4	0.73	15.5	0.67	16.1	7.06	12.5
	Aluminium	36.5	1.52	2.2	76.98	72.6	7.33	43.1	1.95	39.9	1.52	35.2	5.55	9.4
	Total	100.0	25.58	100.0	38.70	100.0	6.21	100.0	1.78	100.0	1.58	100.0	21.43	100.0
AERO 3739	Plastics	15.5	1.52	0.9	12.47	4.9	5.03	12.7	1.56	14.1	1.40	14.5	74.03	55.3
	Black Mass	10.1	1.28	0.5	6.20	1.6	19.95	32.8	5.29	31.1	5.35	36.0	52.35	25.4
	Copper	5.9	63.27	15.0	21.97	3.2	1.83	1.8	0.74	2.5	0.63	2.5	8.69	2.5
	Aluminum	68.5	30.25	83.6	52.25	90.3	4.71	52.7	1.31	52.3	1.03	47.1	5.11	16.9
	Total	100.0	24.81	100.0	39.66	100.0	6.13	100.0	1.72	100.0	1.50	100.0	20.77	100.0
AERO 242	Plastics	15.1	1.65	1.0	12.84	4.9	4.70	11.5	1.47	13.4	1.36	13.9	74.00	53.6
	Black Mass	8.0	1.18	0.4	5.57	1.1	19.79	25.5	4.90	23.6	5.55	29.9	52.99	20.3
	Copper	21.4	65.85	56.6	15.93	8.5	1.56	5.4	0.94	12.2	0.78	11.3	12.33	12.7
	Aluminum	55.5	18.88	42.0	61.60	85.5	6.43	57.6	1.52	50.8	1.20	44.9	5.06	13.5
	Total	100.0	24.94	100.0	39.96	100.0	6.19	100.0	1.66	100.0	1.48	100.0	20.86	100.0
AERO 211	Plastics	15.7	1.58	1.0	12.32	4.9	4.83	12.1	1.48	13.2	1.44	14.5	74.12	53.8
	Black Mass	9.9	1.28	0.5	5.48	1.4	19.42	30.7	5.32	29.9	5.35	34.2	52.97	24.4
	Copper	28.6	69.22	80.1	16.21	11.8	1.82	8.3	0.68	11.0	0.60	11.1	8.82	11.7
	Aluminium	45.9	9.91	18.4	69.79	81.8	6.68	48.9	1.76	45.9	1.36	40.2	4.75	10.1
	Total	100.0	24.70	100.0	39.11	100.0	6.26	100.0	1.76	100.0	1.55	100.0	21.55	100.0

C: Content; R: Recovery

The LOI contents in Table 1 largely indicate a small amount of graphite that could not be removed as a result of the crushing and screening processes. Graphite is not used largely in the Al collector electrodes; however, graphite is mainly used as anode material on copper surfaces. This situation shows that some of the separator particles cannot float in the first flotation circuit (plastic) and the next copper circuit and remain in the sinking product.

Figure 4 The image of floating copper particles in the copper flotation stage

CONCLUSION

In the recycling process of EoL LIBs, chemical methods are preferred in obtaining battery powder, which has high economic value. In academic studies, the flotation method was used mainly to selectively separate anode and cathode active materials, but most of them are far from commercialization. The separation of other important economic values such as Cu and Al collector electrodes, case metals, and plastics remained in the background.

In this original study, the flotation method was used for the recovery of selective metal concentrates from EoL LIBs. As a result of the enrichment processes carried out in a rather large size compared to natural ores, plastics were separated with high content and recovery rates, while the copper particles in the remaining metallic fraction were concentrated with commercial copper reagents. The majority of plastic particles originate from separators.

A copper concentrate assaying 65.13% Cu was successfully obtained with a 96.4% Cu recovery rate using A3418. The drying process is required for the magnetic separation of case metals in sink product. The operational cost of drying will be low due to the coarse size and small amount of feed. A cleaning flotation stage can be added to produce a copper concentrate with much higher contents, or the Al and plastic particles can be recovered by electrostatic or Eddy current separation.

As a result of obtaining metallic concentrates with ore preparation and enrichment methods that reduce the complexity of the structure of LIBs, capacity can be increased in the refining process and toxic gas emissions can be prevented in the thermal process. Again, capacity can be increased and reagent savings can be achieved in the hydrometallurgical process. In conclusion, the findings in this study suggest an alternative process to efficiently recover metal foils, plastics, and case metals in LIBs.

ACKNOWLEDGEMENT

The present study is based on the results of the graduate thesis conducted at the Istanbul Technical University and supported by Istanbul Technical University, BAP (Scientific Research Project) Department with the project code MGA-2022-43640 and MYL-2023-44587. The authors sincerely thank the Mineral Processing Department for providing laboratory equipment and Exitcom Company for the supply of samples.

REFERENCES

1. Kaya, M. (2022) State-of-the-art lithium-ion battery recycling technologies, Circular Economy, 1 (2), 100015.
2. Mylarappa, M., Lakshmi, V. V., Mahesh, K. V., Nagaswarupa, H. P., Prashantha, S. C., Kumara, K. S., Raghavendra, N. (2017) Resource recovery and material characterization of metals from waste li-ion batteries by an eco-friendly leaching agent. Materials Today: Proceedings, 4 (11), 12215-12222.
3. Xu, J., Thomas, H.R., Francis, R.B., Lumb, K.R., Wanga, J., Liang, B. (2008) A review of processes and technologies for the recycling of lithium-ion secondary batteries, J. Power Sources 177, 512-527.
4. Zhang, T., He, Y., Wang, F., Li, H., Duan, C., Wu, C. (2014) Surface analysis of cobalt-enriched crushed products of spent lithium-ion batteries by X-ray photoelectron spectroscopy, Separ. Purif. Technol. 138 21-27.
5. Mennik, F., Burat, F. (2023) The effect of attrition scrubbing as a pre-treatment on the recovery of copper from end-of-life lithium-ion batteries in froth flotation. In: 28th International Mining Congress and Exhibition of Türkiye. Antalya, Türkiye, Proceedings, 1090-1101.
6. Mennik, F., İlkyaz, N. İ., Burat, F. (2023) Selective recovery of metals from spent mobile phone lithium-ion batteries through froth flotation followed by magnetic separation procedure, Results in Engineering, 17, 100868.
7. Çuhadar, İ. E., Mennik, F., Dinç, N.İ., Gül, A., Burat, F. (2023) Characterization and recycling of lithium nickel manganese cobalt oxide type spent mobile phone batteries based on mineral processing technology. Journal of Material Cycles and Waste Management, 25, 1746-1759.
8. Burat, F., Güney, A., Kangal, M.O. (2009) Selective separation of virgin and post-consumer polymers (PET and PVC) by flotation method. Waste Management, 29 (6), 1807-1813.