Sustainable Production, Life Cycle Engineering and Management
Series Editors: Christoph Herrmann, Sami Kara

Arno Kwade
Jan Diekmann *Editors*

Recycling of Lithium-Ion Batteries

The LithoRec Way

Springer

Sustainable Production, Life Cycle Engineering and Management

Series editors

Christoph Herrmann, Braunschweig, Germany
Sami Kara, Sydney, Australia

Modern production enables a high standard of living worldwide through products and services. Global responsibility requires a comprehensive integration of sustainable development fostered by new paradigms, innovative technologies, methods and tools as well as business models. Minimizing material and energy usage, adapting material and energy flows to better fit natural process capacities, and changing consumption behaviour are important aspects of future production. A life cycle perspective and an integrated economic, ecological and social evaluation are essential requirements in management and engineering. This series will focus on the issues and latest developments towards sustainability in production based on life cycle thinking.

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Series Editors' Foreword

Private transportation is going through a dramatic technological transformation. Increased uptake of electric mobility is expected to decrease the environmental impacts caused by the tailpipe emissions from internal combustion vehicles significantly. In addition, if powered with low carbon intensity energy sources, electric vehicles might contribute to reducing the amount of greenhouse gas emissions during their life cycle, therefore, diminishing their contribution to climate change. This technology, however, involves the development of new automotive components, which imply the emergence of new material supply chains. In this regard, the environmental impact of road transportation has started to transform. Its environmental hotspot increasingly shifts to the manufacturing phase of the vehicle and its new components. The traction battery required for the vehicle's operation is one of the key components. Current commercial traction batteries are expensive, heavy in weight, and linked with various potential environmental impacts. They contain high amounts of key engineering metals such as copper, aluminum, nickel, and cobalt. While the extraction processes of these materials cause numerous local environmental impacts linked to mine tailing, their refining processes usually demand large amounts of energy. Furthermore, some of the materials contained in current lithium-ion traction batteries might face future supply risks due to the geopolitical instability and potential market constraints caused by resources scarcity and the low technical feasibility of the extraction processes.

In this context, recycling is a very attractive solution, as it is promoted to return many of these materials back to their supply chains while preventing further environmental and social implications. However, recycling is not without environmental impact due to energy and resources use during the recovery processes. Therefore, recycling processes for traction batteries should be designed with the objective of compensating their life cycle environmental impacts. This means recovering as much valuable and high-quality material as possible while optimizing the consumption of energy, time, and resources during the recycling process.

The research within the LithoRec project aimed at developing recycling processes, which are able to recover much of the materials used in a commercial traction battery system while achieving a significant reduction of the energy consumption compared to the current commercial pyrometallurgical processes. This book is the result of more than 5 years of research. It gives a very detailed description of the processes developed within the LithoRec project. The authors provide the reader with highly valuable insights into understanding not merely the theoretical aspects of recycling processes for traction batteries, but also present analyses of all relevant technical and economic challenges emerging from its implementation. Therefore, this book makes a significant contribution to our understanding of many interactions among the technical, economic, and environmental dimensions surrounding the complex process of a battery recycling process.

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Preface

Lithium-ion batteries are increasingly applied in electrical vehicles, stationary energy storage systems, and other consumer products like power tools. The increasing usage of lithium-ion batteries requires a rise in their production capacity and a minimization of their ecological impact, e.g., carbon dioxide footprint. Moreover, the access to the raw materials has to be ensured and the material costs have to be kept down, although production rates will rise dramatically in the next years. In order to fulfill these tasks and goals, the spent lithium-ion battery systems, especially those of the electric vehicles, have to be recycled and fed back into the material cycle to close the loop. Today, the first recycling technologies have been developed and are used for the recycling of lithium-ion batteries. The most common way is the extraction of the most valuable components by disassembling followed by pyro-metallurgical processes in a smelting furnace. This process recovers components of high costs like cobalt, nickel, and copper, enabling synthesis of new battery materials. However, several other materials including lithium are transferred into slag and are therefore lost for further battery use.

In order to overcome these problems, a novel recycling process was developed in the two LithoRec projects financially supported by the German Ministry for Environment, Nature Conservation, Building, and Nuclear Safety. This book presents the results of the LithoRec II project. The LithoRec way incorporates the battery system transport and the establishment of safety strategies for further battery system handling, dismantling of the battery system, safe crushing of battery cells, and separation of the different battery components, including electrolyte and active materials. To gain new raw materials for the synthesis of active material, the individual compounds were extracted from the coating materials and further separated by hydro-metallurgical processes. Special attention was paid to a safe process design, the maximization of the recycling rate, and the ecological credits, as well as the minimization of recycling costs. The knowledge was finally consolidated in a pilot plant at the Technische Universität Braunschweig, where 1.4 tons of battery systems were recycled. The different book chapters show that a sophisticated technological strategy for the recycling of lithium-ion batteries exists already today, which will enable a closed loop for battery materials in the near future.

As the scientific speaker of the LithoRec projects, I would like to thank all involved partners and collaborators for the intensive and highly motivated work within the last years, and for the preparation of the different book chapters showing the state of the art in recycling of lithium-ion batteries. In place of the collaborators my special thanks go to Christian Hanisch, Jan Diekmann, and Martin Steinbild for their continuous commitment. Our special thank goes to the German Ministry for Environment, Nature Conservation, Building, and Nuclear Safety for the financial support and to the project executing organization VDI/VDE Innovation + Technik GmbH, especially to Dr. Randolf Schließer for the intensive supervision of the project.

Braunschweig, Germany

Arno Kwade
Scientific speaker of the LithoRec projects

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Abbreviations

µCT	Micro-computer tomography
3p4s	3parallel4series
A	Adsorptive
AC	Activated carbon
ACN	Acetonitrile
ADP	Abiotic depletion potential
AP	Acidification potential
ARS	Axial rotary shear
BET	Brunauer–Emmett–Teller theory
BMU	Battery management unit
BT	Breakthrough
CC	Constant current
CCD	Charge-coupled device
CE	Counter electrode
CED	Cumulative energy demand
CHB	Cyclo hexyl benzene
CID	Current interrupt device
CMC	Carboxymethyl cellulose
CP	Constant power
CR	Constant resistance
CV	Constant voltage
DC	Direct current
DEC	Diethyl carbonate
DEDOHC	Diethyl-2,5-dioxahexane dicarboxylate
DEFP	Diethyl fluorophosphate
DMC	Dimethyl carbonate
DMDOHC	Dimethyl-2,5-dioxahexane dicarboxylate
DSC	Differential scanning calorimetry
EC	Ethylene carbonate
EDX	Energy dispersive X-ray analysis
ELV	End-of-life vehicle

EMC	Ethyl methyl carbonate
EMDOHC	Ethylmethyl-2,5-dioxahexane dicarboxylate
EMFP	Ethylmethyl fluorophosphate
EOL	End-of-life
EP	Eutrophication potential
E_{pm}	Exergy primary material
E_{sm}	Exergy secondary material
EV	Electric vehicles
FTIR	Fourier transform infrared spectroscopy
GC	Gas chromatography
GC–MS	Gas chromatography–mass spectrometry
GHG	Greenhouse gas
GWP	Global warming potential
HEV	Hybrid electric vehicle
$HHPCO_2$	Helium head pressurized carbon dioxide
HSAL	High surface area lithium
HTP	Human toxicity potential
HV	High voltage
IAST	Ideal adsorbed solution theory
IC	Ionic chromatography
IC/ESI-MS	Ion chromatography–electrospray ionization-mass spectrometry
ICP-MS	Inductively coupled plasma mass spectrometry
ICP-OES	Inductively coupled plasma optical emission spectrometry
IDLH	Immediately dangerous to life and health
LCA	Life cycle assessment
LCO	Lithium cobalt oxide $LiCoO_2$
LEL	Lower explosion limit
LFP	Lithium iron phosphate
LIB	Lithium-ion battery
LOD	Limit of detection
LOQ	Limit of quantification
MMU	Module management unit
MOSFET	Metal oxide semiconductor field effect transistor
NA	Necessity to automate the corresponding disassembly operation
NCA	Lithium nickel aluminum oxide
NCM	Lithium nickel cobalt manganese oxide
NCM 111	Lithium nickel manganese oxide $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$
NCM 532	Lithium nickel manganese oxide $LiNi_{0.5}Co_{0.3}Mn_{0.2}O_2$
NCM 622	Lithium nickel manganese oxide $LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$
NMP	N methyl pyrrolidone
NPV	Net present value
OEM	Original equipment manufacturer
PA	Polyamide
PC	Propylene carbonate
PHEV	Plug-in hybrid electric vehicle

PLC	Programmable logic control
PMFP	Particle matter formation potential
POCP	Photochemical ozone creation potential
PTC	Positive temperature coefficient
PTFE	Polytetrafluoroethylene
PVDF	Polyvinylidene fluoride
RE	Reference electrode
rpm	Rounds per minute
RRS	Radial rotary shear
$scCO_2$	Supercritical carbon dioxide
SEI	Solid electrolyte interface
SEM	Scanning electron microscopy
SFE	Supercritical fluid extraction
SOC	State of charge
SOH	State of health
$subCO_2$	Subcritical carbon dioxide
TAA	Technical ability of a disassembly process to be automated
TAP	Terrestrial eutrophication potential
TGA	Thermogravimetric analysis
UEL	Upper explosion limit
WE	Working electrode
WEEE	Waste electrical and electronic equipment
XRD	X-ray powder diffraction

Symbols

V	Volume (mL)
X	Carbon loading (mmol/g)
m	Mass (g)
Y	Gas loading (g/L)
M	Molar mass (g/mol)
ρ	Density (kg/m^3)
b	Langmuir parameter (L/mmol)
b’	Langmuir parameter (–)
t	Tóth Parameter (–)
φ	Saturation (–)
π	Spreading pressure (–)
R	Ideal gas constant (J/molK)
T	Temperature (K)
y	Concentration is gas phase (mol/mol)
x	Concentration in adsorbed phase (mol/mol)
α	Seperation factor (–)
A	Area (m^2)
t	Time (h)

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