



**MICROWAVE-ASSISTED HYDROTHERMAL SYNTHESIS OF NMC955 CATH-  
ODE MATERIAL PRECURSOR FOR LITHIUM-ION BATTERIES**

Lappeenranta–Lahden teknillinen yliopisto LUT

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## TIIVISTELMÄ

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Chemical Engineering

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### **Microaaltotuettu hydroterminen synteesi NMC955-katodimateriaalin esiasteen litiumioni akuille**

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Nykymaailmassa laitteiden määrä, jotka vaativat sähkön käyttöä, kasvaa jatkuvasti, ja samalla kasvaa myös kehittyneempien energian varastointijärjestelmien kysyntä. Innovaatioiden tarve tällä alalla on kasvanut entisestään uusiutuvan energian integroinnin, sähköajoneuvojen ja kannettavan elektroniikan lisääntyvän tarpeen vuoksi. Tämä tutkimus keskittyy akkuihin käytettyihin materiaaleihin, jotka ovat keskeisiä näiden kasvavien vaatimusten täyttämiseksi. Ihmiskunta pyrkii optimoida järjestelmiä, eikä energian varastointi ole poikkeus – tavoitteena on maksimoida tehokkuus, vähentää kustannuksia ja minimoida haitallisten materiaalien käyttö. Tässä tutkimuksessa keskitytään kahteen pääaspektiin: koboltin käytön vähentämiseen lisäämällä nikkelin määrää aktiivisessa materiaalissa sekä synteesiajan lyhentämiseen hyödyntämällä suhteellisen vähän tutkittua synteessimenetelmää. Tämä opinnäytetyö alkaa yleiskatsauksella akkujen toimintaperiaatteisiin, synteessimenetelmiin ja aiempaan tutkimukseen perustuviin aktiivisiin materiaaleihin. Opinnäytetyön toisessa osassa käsitellään tutkimuksen aikana tehtyjä kokeita, joissa käytettiin mikroaaltotuettua hydrotermistä synteesiä, ja tutkitaan erilaisten muuttujien, kuten pH:n, lämpötilan, mikroaaltotehon ja paineen vaikutuksia. Tämä uusi synteessimenetelmä pyrkii nopeuttamaan suorituskykyisten materiaalien kehitystä ja alentamaan tuotantokustannuksia. XRD-mittausten ja galvano-staattisten elektrokemiallisten analyysien avulla arvioitiin syntyneiden akkujen suorituskykyä tavoitteena parantaa niiden tehokkuutta ja käyttöikä. Tulokset edistävät kestävämpien ja tehokkaampien energian varastointiratkaisujen kehitystä, mikä voi vaikuttaa sekä tulevaan akateemiseen tutkimukseen että käytännön sovelluksiin teollisuudessa, kuten sähköajoneuvoissa, uusiutuvan energian varastoinnissa ja kulutuselektroniikassa.

## ABSTRACT

Lappeenranta–Lahti University of Technology LUT

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Chemical Engineering

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### **Microwave-assisted hydrothermal synthesis of NMC955 cathode material precursor for lithium-ion batteries**

Bachelor's thesis

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47 pages, 9 figures, 1 table

Examiners: Associate Professor Ekaterina Laakso

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In today's world, the number of devices requiring constant electricity usage is continuously increasing, along with the demand for more advanced energy storage systems. The demand for innovation in this field has become even more pressing due to the growing need for renewable energy integration, electric vehicles, and portable electronics. This research focuses on the materials used in batteries, which are crucial to meeting these growing demands. Humanity is constantly striving to optimize systems, and energy storage is no exception, aiming to maximize efficiency, reduce costs, and minimize the use of harmful materials. This research focuses on two main aspects: reducing the use of cobalt by increasing the amount of nickel in the active material and reducing the synthesis time by utilizing a relatively unexplored synthesis route. This thesis starts with an overview of the general operating principles of batteries, synthesis methods, and types of active materials based on previous research. The second part of the thesis focuses on the experiments done during this research using microwave-assisted hydrothermal synthesis, exploring the influence of various variables such as pH, temperature, microwave power, and pressure. The novel synthesis approach aims to accelerate the development of high-performance materials while lowering production costs. XRD data and galvanostatic electrochemical analysis of the resultant batteries were used to estimate their performance, with the goal of enhancing battery efficiency and longevity. The results contribute to the ongoing search for more sustainable and efficient energy storage solutions, potentially influencing both future academic research and practical applications in industries like EVs, renewable energy storage, and consumer electronics.

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## SYMBOLS AND ABBREVIATIONS

### Symbols

mAh	Milliampere hour
kV	Kilovolt
V	Volt
wt %	Weight percentage

### Abbreviations

C-rate	Charging Rate
EVs	Electric Vehicles
LIBs	Li <sup>+</sup> ion Batteries
NMC(OH) <sub>2</sub>	Nickel manganese cobalt hydroxide
LiCoO <sub>2</sub>	lithium nickel manganese cobalt oxide
LNCAO <sub>2</sub>	lithium nickel cobalt aluminum oxide
LNMCO <sub>2</sub>	lithium nickel manganese cobalt oxide
NH <sub>4</sub> OH	Ammonium hydroxide
NMC	Lithiated oxide LiNi <sub>x</sub> Mn <sub>y</sub> Co <sub>2</sub> O <sub>2</sub>
SEM	Scanning Electron Microscope
XRD	X-Ray Diffraction

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# 1 Introduction

Throughout human history, people have sought increasingly efficient and advanced energy sources. From the use of wood and coal to the development of oil and natural gas, the advancement of energy technologies is inextricably linked to societal progress. The discovery of electricity was one of the most significant achievements, marking the beginning of the electric energy era, which today forms the foundation of modern infrastructure and technology.

In recent decades, particular attention has been given to the development of alternative energy sources and the creation of highly efficient energy storage systems. In the context of the global energy transition, lithium-ion batteries are incredibly important, powering electric vehicles, renewable energy sources, and portable electronics. A key factor in development of lithium-ion batteries is the advancement of cathode materials that offer high capacity, stability, and durability, as these materials play a crucial role in determining the overall performance of the battery cell.

This study examines the microwave-assisted hydrothermal synthesis of a cathode precursor material mixed nickel, manganese, and cobalt hydroxide-(NMC(OH)<sub>2</sub>) This method represents a promising approach to creating materials with enhanced properties, enabling control over the morphology, composition, and crystalline structure of the obtained product. The research focuses on optimizing synthesis conditions, analysing the structural and electrochemical properties of the obtained materials, and evaluating their applicability as cathode components for lithium-ion batteries.

## 2 Literature review

This chapter provides general information based on a literature review on the topic of lithium-ion batteries, cathode materials and types of synthesis that is essential for understanding and performing further experiment.

### 2.1 Operating Principle

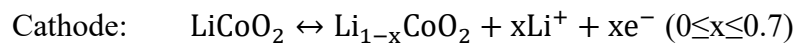
A lithium-ion (LiB) battery is an electrochemical device that converts chemical energy into electrical energy through reversible reactions between its electrodes. Its functioning relies on the transfer of lithium ions through an electrolyte, moving between the anode and cathode.

During battery charging, lithium ions move from the cathode through the electrolyte to the anode, embedding in the layers of graphite. During discharge, lithium ions are released from the anode and move back to the cathode, which is accompanied by the movement of electrons through the external circuit, generating electric current to power the device. Figure 1 illustrates the processes of intercalation–de-intercalation reaction mechanism in a rechargeable lithium-ion battery containing solid electrodes and a liquid electrolyte (Roy & Srivastava, 2015).

According to (Roy & Srivastava), a lithium-ion battery consists of main components that enable it to store and release energy efficiently. The main elements of a lithium-ion battery include four major components such as: anode, cathode, separator and an electrolyte.

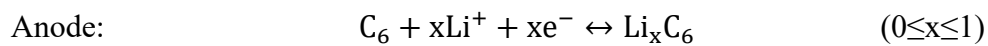
### 2.1.1 Cathode (Positive Electrode)

The cathode is the source of Li ions in the battery. It typically consists of a complex metal oxide, such as lithium cobalt oxide ( $\text{LiCoO}_2$ ), lithium nickel manganese cobalt oxide ( $\text{LNMC O}_2$ ), or lithium nickel cobalt aluminum oxide ( $\text{LNCAO}_2$ ). The cathode determines the energy density of the battery and partially influences its cost, as it may contain expensive materials like Co and Ni. Reaction describing the processes occurring at the cathode (Roy & Srivastava, 2015):



### 2.1.2 Anode (Negative Electrode)

The anode in modern lithium-ion batteries is made of graphite. Its layered structure allows to store Li ions during charging and release them during discharging. The movement of Li ions between the anode and cathode creates a flow of electrons, generating electric current. Electrons flow through the external electrical circuit to balance the excess positive charge  $\text{Li}^+$  in the electrode. During the discharging process, the reverse mechanism takes place. Reaction describing the processes occurring at the anode (Roy & Srivastava, 2015):



### 2.1.3 Electrolyte

The electrolyte serves as the medium for lithium ions to move between the cathode and anode. It can be a liquid, gel, or solid substance containing Li salts (such as  $\text{LiPF}_6$ ) dissolved in a solvent. The electrolyte has high ionic conductivity but acts as an electronic insulator to prevent short circuits.

### 2.1.4 Separator

The separator is a critical component in lithium-ion batteries, designed to physically isolate the anode and cathode to prevent direct contact and short circuits. It is made of a thin, porous polymer membrane, typically composed of materials like polyethylene or polypropylene. The separator's microporous structure allows Li ions to move freely between the electrodes while blocking the flow of electrons. This ensures proper ion exchange during charging and discharging cycles. Advanced separators may include multilayer designs or ceramic coatings to enhance thermal stability and safety under high temperatures or extreme operating conditions.

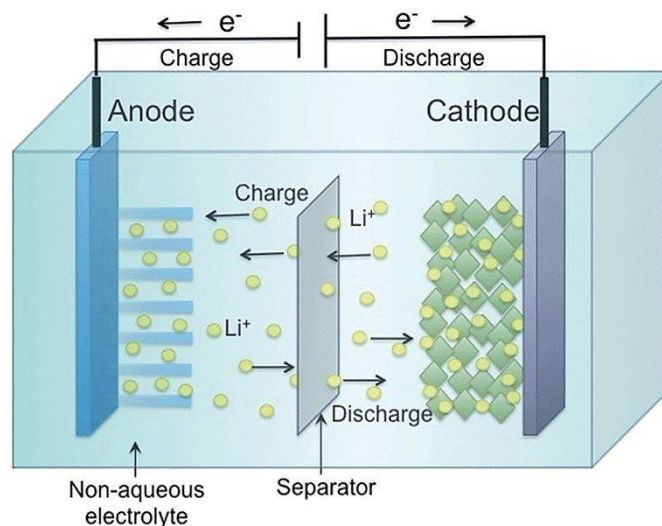


Figure 1. Schematic diagram of the lithium-ion battery operation (Roy & Srivastava, 2015)

### 2.1.5 Casing and Control Electronics

The internal components of the battery are enclosed in a sealed casing that protects them from moisture and mechanical damage. Most commercial lithium-ion batteries also include control electronics (Battery Management System, BMS), which monitor the battery's condition, preventing overheating, overcharging, and deep discharging to ensure safety and extend the battery's lifespan (Aalto, 2012).

## 2.2 Cathode materials for LIB

Approaching the main topic of the study, more specifically nickel-rich cathode precursor material, it is necessary to familiarize ourselves with the types of cathode materials. According to (Ma, 2021) it is possible to distinguish main types of cathode materials.

### 2.2.1 LMO (Lithium Manganese Oxide)

$\text{LiMn}_2\text{O}_4$  (LMO) is a commonly used cathode material in lithium-ion batteries, valued for its affordability, safety, and high-power output. Its spinel crystal structure provides robust thermal stability and allows for fast lithium-ion diffusion, enabling quick charge and discharge cycles (Zhang, Y. et al., 2021). While LMO offers moderate specific capacity (100-120  $\text{mAhg}^{-1}$ ) (Mishra et al., 2023) and operates at an average voltage of 4.3 V (Park & Kim, 2024), it is prone to capacity fade, particularly under high temperatures or prolonged cycling. Manganese's abundance and low cost make LMO an economical choice, and its excellent thermal stability enhances safety, reducing the risk of overheating. These characteristics make it ideal for applications like power tools, medical devices, and energy storage systems, (Blomgren, 2017) as well as electric vehicles, where it is often blended with other materials like NMC to improve performance. Despite its limitations in energy density and longevity, advancements such as surface coatings, elemental doping, and blended cathodes aim to enhance LMO's efficiency and durability, securing its role in cost-sensitive and high-power battery applications. (Blomgren, 2017)

### 2.2.2 NMC (Lithium Nickel Manganese Cobalt Oxide)

$\text{LiNi}_x\text{Mn}_y\text{Co}_z\text{O}_2$  (NMC,  $x + y + z = 1$ ) is one of the most prominent cathode materials in lithium-ion batteries, prized for its high energy density, long cycle life, and versatility across various applications. Its layered crystal structure enables efficient lithium-ion diffusion and high capacity, delivering energy densities in the range of  $>200 \text{ mAhg}^{-1}$  depending on the composition ratio (Bin Abu Sofian et al., 2024). By adjusting the nickel, manganese, and cobalt proportions—commonly referred to as NMC ratios such as 532, 622, or 811—manufacturers can optimize performance for specific requirements, balancing energy density, thermal stability, and cost (Bin Abu Sofian et al., 2024). NMC's average operating voltage is around  $\sim 3.8 \text{ V}$ , and its excellent electrochemical properties make it suitable for electric vehicles, portable electronics, and grid storage systems (Bin Abu Sofian et al., 2024). However, challenges include thermal instability at high nickel content and the high cost and scarcity of cobalt, prompting extensive research into reducing cobalt dependency (Panella & De Putter, 2022) or finding alternative stabilizers like aluminium or doping elements (Kim et al., 2023). Despite these challenges, innovations in manufacturing, such as gradient-layering, single-crystal NMC, and advanced coating techniques, continue to enhance its safety, cycle life, and energy density (Wang, W. et al., 2020). These advances ensure that NMC remains a cornerstone in lithium-ion battery technology, especially in industries requiring a balance of energy density, longevity, and performance.

### 2.2.3 LFP (Lithium Iron Phosphate)

$\text{LiFePO}_4$  (LFP) is a popular cathode material in lithium-ion batteries, known for its high safety, long cycle life, and thermal stability. LFP offers a lower specific capacity compared to other cathode materials (typically around  $170 \text{ mAhg}^{-1}$ ) (Cheruvally, 2008) but excels in power output and stability. It operates at a nominal voltage of about  $3.4 \text{ V}$  (Cheruvally, 2008). The abundance of iron and phosphate makes LFP an economical choice, and its resistance to thermal runaway ensures safer operation, making it ideal for applications in electric vehicles, energy storage, and power tools.

#### 2.2.4 LCO (Lithium Cobalt Oxide)

$\text{LiCoO}_2$  is one of the earliest and most widely used cathode materials in lithium-ion batteries (Monconduit et al., 2015). Known for its high energy density,  $150\text{--}200 \text{ mAhg}^{-1}$  (Mishra et al., 2023) it operates at a voltage of 4.2–4.5 V (Monconduit et al., 2015) making it ideal for small electronics like smartphones and laptops. However, LCO has significant drawbacks, including high cost, toxicity, and poor thermal stability, which limit its use in larger-scale applications such as electric vehicles. Efforts to improve LCO's performance often focus on enhancing its stability and addressing environmental concerns.

#### 2.2.5 NCA (Lithium Nickel Cobalt Aluminum Oxide)

$\text{LiNi}_x\text{Co}_y\text{Al}_z\text{O}_2$  (NCA,  $x + y + z = 1$ ) is a high-performance cathode material used primarily in electric vehicles. NCA offers a high energy density (around  $200\text{--}250 \text{ mAhg}^{-1}$ ) (Bin Abu Sofian et al., 2024) and a voltage range of 3.6–3.7 V (Bin Abu Sofian et al., 2024), making it suitable for applications where long-range and power output are critical. NCA's key advantages include its high capacity and thermal stability, although it is more expensive than some alternatives due to the use of cobalt. The incorporation of cobalt into the material enhances the stability of nickel's crystallographic structure by minimizing cation mixing (Delmas et al., 1999). Aluminum helps maintaining the layered structure of the cathode by reducing phase transitions, which improves long-term cycling stability. The material's high performance and stability make it ideal for large-scale applications despite cost concerns.

### 2.3 Nickel-rich cathode materials: a pathway to superior lithium-ion batteries

As the demand for high-energy lithium-ion batteries (LIBs) continues to rise, particularly in sectors such as electric vehicles, portable electronics, and grid storage, choosing the right materials for battery components has become critical. Nickel-rich materials in LIB cathodes have emerged as a popular choice due to their ability to significantly enhance battery performance. By increasing energy density, reducing costs, and improving sustainability, nickel-rich cathode materials address many of the limitations found in traditional lithium-ion battery chemistries.

This chapter explores why nickel-rich cathode materials are an advantageous choice for LIBs. We will examine the benefits of nickel in cathode composition, including higher capacity and energy density, reduced reliance on cobalt, and potential cost savings. Additionally, we will discuss the technical challenges and innovations associated with using high-nickel materials, from stability issues to advanced battery management requirements. Understanding the advantages and considerations of nickel-rich precursors provides insight into the evolving landscape of lithium-ion technology and its future applications.

### 2.3.1 Comparison of NMC with other basic cathode active materials

Nickel-Manganese-Cobalt Oxide (NMC) stands out as one of the most versatile and widely used cathode materials in lithium-ion batteries. Its ability to balance energy density, longevity, and cost makes it a go-to choice for various applications, including electric vehicles, portable electronics, and stationary energy storage. However, like any material, NMC has its strengths and weaknesses, especially when compared to alternatives like Lithium Manganese Oxide (LMO), Lithium Iron Phosphate (LFP), Lithium Cobalt Oxide (LCO), and Nickel-Cobalt-Aluminium Oxide (NCA).

One of NMC's greatest advantages is its high energy density, which can reach up to  $220 \text{ mAhg}^{-1}$  (Bin Abu Sofian et al., 2024) depending on the composition. This makes it far superior to LMO and LFP in applications like electric vehicles, where maximizing driving range is critical (Blomgren, 2017). Furthermore, NMC offers unparalleled flexibility thanks to its customizable Ni, Mn, and Co ratios. Compositions like NMC 532, 622, and 811 allow manufacturers to fine-tune its properties for specific needs, such as enhancing energy density for long-range EVs or improving cycle life for grid-scale energy storage (Tesla, 2021). This level of adaptability gives NMC a competitive edge in industries that require a tailored balance of performance and cost.

NMC also strikes a balance between affordability and efficiency. While it is more expensive than simpler materials like LFP or LMO, it remains a cost-effective alternative to LCO and NCA, thanks to efforts to reduce its cobalt content and increase the proportion of more abundant nickel (Wang, B. et al., 2021). This affordability makes it particularly advantageous in the mass-market electric vehicle segment, where cost and performance must align to meet consumer demands. Additionally, NMC's stability under cycling conditions ensures it outlasts LMO and LCO, providing a reliable solution for long-term applications like home energy storage systems and commercial power grids (Bin Abu Sofian et al., 2024).

In terms of safety, NMC offers reasonable thermal stability, outperforming LCO and NCA, though it does not match the exceptional safety profile of LFP. Despite this, NMC is widely used in mid-tier and premium electric vehicles (Baczyńska et al., 2018), where its energy density and cycling performance outweigh the need for LFP's higher thermal stability. For portable electronics, where space constraints and energy density are critical, NMC also

excels, offering better longevity and cost efficiency compared to LCO, which has historically dominated the sector.

However, use of NMC meets many challenges. Its reliance on cobalt, despite being reduced in newer formulations, presents ethical and supply chain concerns (Panella & De Putter, 2022). Materials like LFP and LMO have a clear advantage in this area, as they avoid such issues entirely. Additionally, while its energy density surpasses that of LFP, it falls short of NCA, which remains the leader in high-performance applications such as premium electric vehicles and aerospace technologies (Blomgren, 2017). NMC's complex multi-component structure also makes recycling more challenging compared to simpler chemistries like LFP, further complicating its long-term sustainability (Fridrich et al., 2024).

When viewed alongside its competitors, NMC emerges as the quintessential jack-of-all-trades. It outperforms LMO in energy density and longevity, outshines LFP in capacity and voltage, and offers better stability and affordability than LCO. While it cannot match NCA's energy density, it compensates with greater stability and adaptability, particularly in applications where safety and cost must be balanced with performance.

In conclusion, NMC represents a careful balance of energy density, cost-effectiveness, and safety, making it a cornerstone of modern lithium-ion battery technology. It dominates the electric vehicle market, especially in mid-tier and semi-premium segments, in portable electronics with its balance of cost and performance and still a good choice in stationary energy storage solutions. While it is not the ideal solution for every scenario, its versatility ensures it remains a top choice in a rapidly evolving industry where no single material can address every need.

### 2.3.2 Challenges of nickel-rich cathode materials

Ni-rich cathode materials, particularly high-Ni NMC formulations, are regarded as next-generation solutions for lithium-ion batteries due to their high energy density, operating voltage, and reduced reliance on Co. These properties make them especially attractive for applications such as EVs and portable electronic devices. However, despite their promising characteristics, these materials encounter various challenges that impede their commercialization and long-term reliability.

One of the primary challenges of Ni-rich cathodes is their structural instability (Jamil et al., 2022). Over cycling, these materials experience phase transformations from a layered to spinel and eventually to a rock-salt structure, which leads to capacity decay and voltage drops. Additionally, mechanical stress generates microcracks, further degrading their structural integrity and electrochemical performance (Zhang, S. S., 2020). These issues are compounded by thermal instability, as Ni-rich cathodes are prone to overheating and thermal runaway, which raises significant safety concerns in high-voltage and high-stress applications (Jamil et al., 2022).

Chemical reactivity poses another significant hurdle. Ni-rich materials react with moisture and carbon dioxide during storage, forming impurities that reduce battery efficiency and complicate manufacturing processes (Jamil et al., 2022). Cation mixing, where Ni and Li ions interchange within the crystal lattice, disrupts the material's electrochemical performance. Furthermore, lattice oxygen loss destabilizes the cathode and increases the risk of thermal runaway.

The cathode-electrolyte interface is also a point of vulnerability. Erosive interactions at this boundary lead to the degradation of ion transport pathways, reducing the efficiency and lifespan of the battery (Jamil et al., 2022). These challenges are particularly pronounced in advanced formulations, such as single-crystal or ultra-high-Ni cathodes, which require innovative structural designs and precise synthesis techniques to maintain stability.

Efforts to address these limitations include strategies such as doping with stabilizing elements (e.g., Mg and Zr) to enhance structural integrity and prevent cation mixing (Bai et al., 2020). Protective surface coatings, often applied using techniques like atomic layer deposition, shield the cathodes from chemical degradation while improving their compatibility with

electrolytes. Grain boundary engineering and tailored microstructures are also being explored to mitigate cracking and improve durability. For single-crystal cathodes, optimized synthesis methods and advanced structural designs are essential to enhance their performance under demanding conditions.

Despite these advancements, Ni-rich cathodes present additional challenges related to environmental and economic factors. Recycling Ni-rich materials is complex, and the environmental impact of nickel mining raises sustainability concerns (Bin Abu Sofian et al., 2024). Moreover, reducing Co content, while lowering costs and addressing ethical sourcing issues, compromises the material's stability, necessitating further innovation in material composition and processing (Panella & De Putter, 2022).

In conclusion, Ni-rich cathode materials hold immense potential for high-performance batteries but require significant research and engineering to overcome their limitations. Advanced characterization techniques, coupled with innovative material design and synthesis methods, are critical to ensuring their reliability and safety. Addressing these challenges will pave the way for the widespread adoption of nickel-rich cathodes in electric vehicles and other applications demanding high energy density and long-term durability.

### 2.3.3 The Importance of Morphology and Size Distribution

Lithium-ion batteries are one of the most used energy sources thanks to their high energy density, long lifespan, and efficiency. A crucial factor affecting battery performance is the morphology of the cathode material, particularly its particle shape and size. Studies show that cathode particles with a spherical shape and a size of approximately 10  $\mu\text{m}$  provide optimal battery performance (Liu et al., 2016). This chapter examines the reasons for selecting these parameters and their influence on the electrochemical properties of Li-ion batteries.

The particle size of the cathode material plays a crucial role in lithium-ion transport kinetics. Nanoparticles (around 10 nm) reduce the diffusion path for lithium ions, accelerating intercalation and deintercalation processes during battery charge and discharge. Moreover, minimizing particle size lowers the internal resistance of the cathode material, improving the battery's charge and discharge rates.

During cyclic charging and discharging, lithium-ion batteries experience mechanical deformations due to volume changes in the active material. Large particles are prone to crack formation, leading to gradual degradation. In contrast, nanoparticles (~10 nm) are less susceptible to mechanical damage, improving battery longevity (Liu et al., 2016). The spherical shape of particles further reduces the likelihood of structural defects and ensures uniform stress distribution within the cathode material.

Smaller particle sizes lead to an increase in the specific surface area, improving contact with the electrolyte and enhancing electrochemical reaction rates. This is especially crucial for achieving high power output, as increased contact between the cathode material and the electrolyte enables faster lithium-ion penetration into the material's structure.

Optimizing particle shape also affects the packing density of the cathode material. Spherical particles enable more uniform distribution within the electrode, reducing void spaces and increasing the battery's energy density. This is especially crucial for developing high-energy-density batteries.

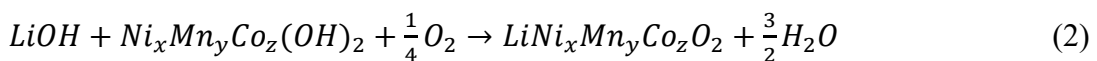
## 2.4 Synthesis methods of NMC cathode material

This chapter explores common synthesis techniques for NMC cathode materials, examining their principles, advantages, and limitations. Synthesis methods for NMC materials typically aim to control particle size, morphology, and phase composition to address inherent challenges such as structural instability, cation mixing, and interfacial degradation. These methods can range from conventional solid-state reactions to advanced techniques like co-precipitation, hydrothermal synthesis, and spray pyrolysis. Each method offers unique advantages in terms of scalability, material quality, and process complexity. Understanding these methods is essential to advancing the performance and commercial viability of NMC-based lithium-ion batteries.

To understand the methods of synthesis of materials for cathode, it is necessary to consider that the process of lithiation (described in more detail in Chapter 3.1.2) is possible in two ways.

The first one is the direct synthesis method, where a source of lithium, usually lithium carbonate or lithium hydroxide is mixed directly with the precursor material, before the synthesis process. This method is highly controversial and is considered less stable than the traditional method, which will be discussed below. It may cause complications such as unstable phase stability, issues with dissolving of lithium sources and increasing of the whole process complexity.

The second method is mixing the lithium source with the already prepared cathode precursor and further calcination in a tube furnace, in a constant oxygen flow. This method is the most efficient and is used in all major synthesis methods. The reaction in a tube furnace is described below:



According to Malik et al., it is possible to distinguish 3 main types of synthesis such as: Co-precipitation, sol-gel and hydro-thermal.

Chapters 2.4.1, 2.4.2, and 2.4.3 will describe ways to synthesise precursors for active cathode materials.

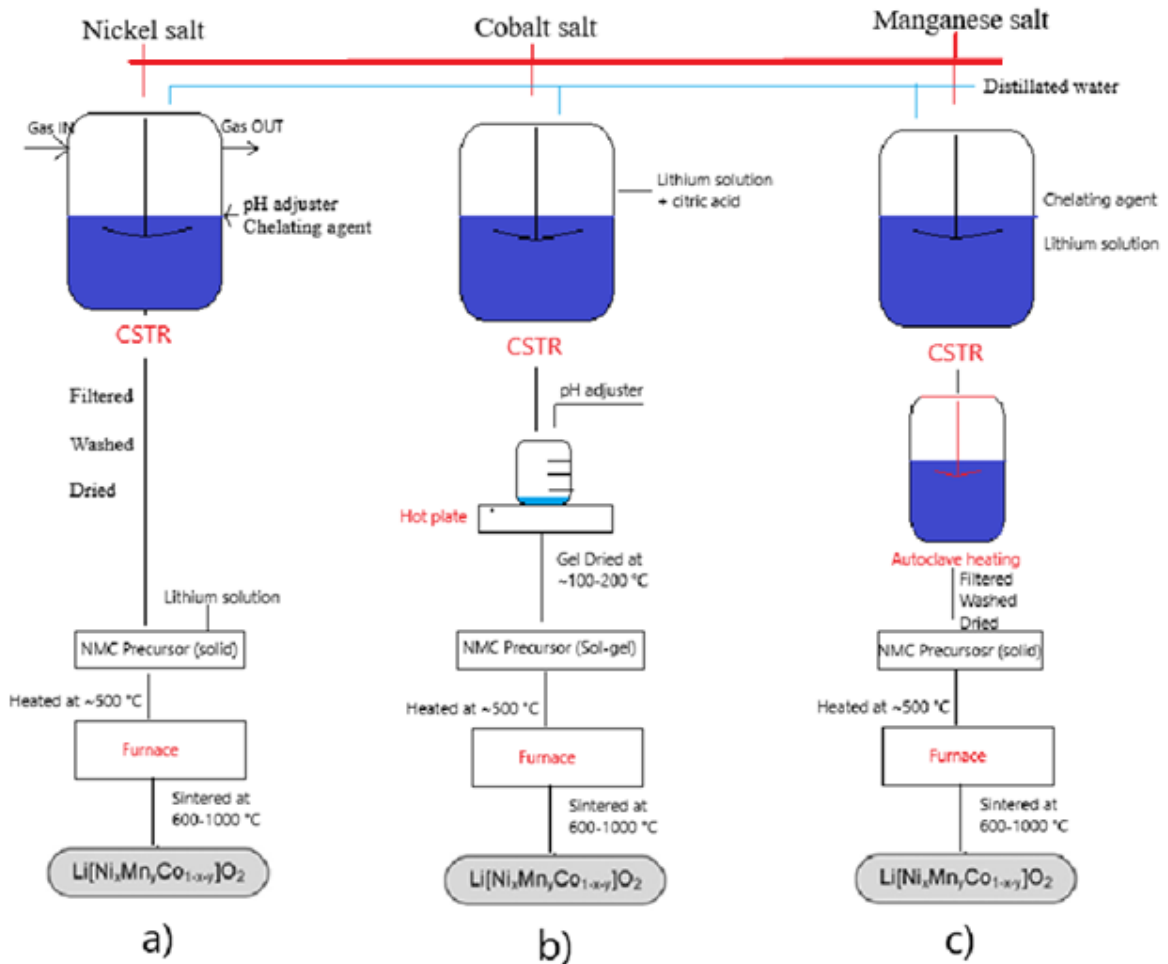
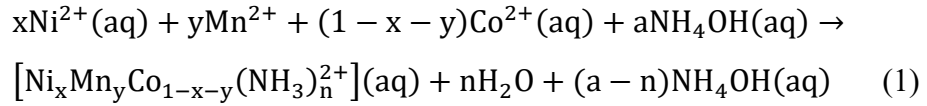


Figure 2. The figure describes three most popular types of synthesis (a) Co-precipitation (b) sol-gel (c) hydrothermal.

### 2.4.1 Co-precipitation method

The co-precipitation method is a key technique for producing NMC cathode material precursors, widely used in lithium-ion batteries due to its ability to ensure homogeneous mixing of transition metals, precise control over particle morphology, and industrial scalability. This method consists of two main stages: precursor precipitation and sintering. The schematic of the process is provided in a) of figure 2. Transition metal salts are dissolved in stoichiometric

ratios to form an ionic solution, and precipitation is achieved by adding an alkaline and chelating agent. A chelating agent, typically ammonia, regulates particle morphology and prevents phase separation (Malik et al., 2022).



Three primary types of co-precipitation are widely studied. Hydroxide co-precipitation, the most common, produces dense spherical particles with high tap density, although it requires an inert atmosphere to prevent manganese oxidation. Carbonate co-precipitation, which avoids oxidation and is environmentally friendly, often results in higher porosity that may compromise particle strength. Oxalate co-precipitation provides high-purity precursors, but its low solubility limits production rates and requires careful control to avoid phase separation (Saaid et al., 2024).

Process parameters significantly affect the properties of NMC materials. Optimal precipitation time (18–24 hours) improves crystallinity and reduces cation mixing. Temperature is critical; for example, 60°C ensures dense spherical particles in carbonate co-precipitation while minimizing impurities. The pH range of 10.5–11.5 is optimal for controlling nucleation and growth, yielding uniform and dense particles (Malik et al., 2022). Chelating agents like ammonia, in controlled molar ratios to metal ions, enhance particle uniformity and tap density. Stirring rates around 1000 rpm ensure homogeneous mixing and reduce agglomeration (CHEN et al., 2005).

The co-precipitation method offers advantages such as uniform atomic-scale mixing, precise control over particle morphology, and adaptability for large-scale production, making it essential for industrial applications. However, challenges such as impurity formation due to transition metal oxidation and the need for precise process optimization persist. Despite these, the method remains a cornerstone of high-performance lithium-ion battery production. Further advancements in parameter optimization and scalability will enhance its industrial viability and environmental sustainability.

## 2.4.2 Sol-gel method

The sol-gel method is an advanced synthesis path used for producing NMC cathode materials, offering accurate control over particle morphology, size distribution, and crystalline structure. Originating as a method for synthesizing silica gels in the mid-19<sup>th</sup>-century, its application in producing lithium-ion battery materials is relatively recent. This method is particularly favored for its ability to achieve homogeneous mixing of transition metals (Malik et al., 2022) at the molecular level and for producing materials with superior electrochemical properties, including high specific capacity and cycling stability. The schematic of the process is provided on figure 3b.

The process begins with dissolving transition metals (Ni, Mn, Co) in their nitrate or acetate forms in distilled water. The chelating agent, typically citric acid added to control pH of the solution. The pH of the solution is adjusted to around 8 using  $\text{NH}_4\text{OH}$ . The mixture is then heated at approximately 80°C to form a viscous gel, which is subsequently dried at 120°C to remove residual solvents. The dried precursor is calcined at 450°C, followed by sintering at 800–900°C to achieve the desired crystallinity and phase composition (Malik et al., 2022).

Key parameters influencing the sol-gel process include the choice and concentration of the chelating agent, pH, drying temperature, and sintering conditions. Citric acid is the most used chelating agent due to its low cost and environmental safety. Adjusting the citric acid-to-metal ion ratio ( $R'$ ) affects particle size, with higher ratios promoting smaller, uniform particles. pH adjustments during gel formation regulate the nucleation and growth processes, influencing the morphology and density of the final particles (Çetin et al., 2020).

The sol-gel method has distinct advantages, such as low processing temperatures, uniform particle distribution, and excellent control over stoichiometry, which contribute to improved structural stability and electrochemical performance of the cathode materials. However, the method also has limitations, including complex operational steps, longer preparation times, and higher costs compared to simpler techniques like co-precipitation. These factors currently restrict its widespread industrial adoption, confining its use mainly to laboratory-scale research.

Despite its challenges, the sol-gel method remains a valuable synthesis technique for developing high-performance NMC cathode materials. Its ability to produce materials with high

crystallinity, tailored particle size, and uniform morphology positions it as an essential tool for advancing lithium-ion battery technology. Future research should focus on simplifying the operational steps and reducing costs to enable broader industrial application.

#### 2.4.3 Hydrothermal method

The hydrothermal method is a versatile synthesis technique for producing NMC cathode materials, known for its ability to yield highly crystalline particles with uniform morphology and excellent electrochemical performance. This method leverages aqueous-phase reactions under high temperature and pressure, offering advantages such as enhanced crystallinity, controlled particle size, and energy efficiency. The schematic of the process is provided in figure 2. While primarily used in laboratory-scale research, its potential for scalability makes it a promising method for industrial applications.

The hydrothermal process begins by dissolving stoichiometric amounts of transition metal salts (e.g., nitrates or acetates of Ni, Mn, Co) in distilled water, often with a chelating agent such as ammonium hydroxide or oxalic acid to stabilize the solution. The pH is adjusted to around 10.5-11.5 to optimize nucleation and growth conditions. The resulting solution or slurry is transferred into a Teflon-lined stainless-steel autoclave, filling up to 80–85% of its capacity. The sealed autoclave is heated to 150–250°C under autogenous pressure for 5–24 hours, depending on the desired properties. After cooling, the solid product is filtered, washed to remove impurities, and dried. The dried powder is typically mixed with excess lithium hydroxide and sintered at 800–900°C to achieve the final cathode material (Pimenta et al., 2017).

Key parameters in the hydrothermal method include reaction temperature, duration, and choice of precipitating agent. Higher temperatures (e.g., 200°C) enhance crystallinity and reduce cation mixing, while excessive temperatures can lead to agglomeration or impurity formation. Reaction times exceeding 18 hours allow sufficient energy for regular atomic arrangement, improving electrochemical performance. Precipitating agents such as urea or oxalic acid control the morphology and chemical stability of the precursor, with each offering unique advantages. For example, urea decomposes gradually, releasing carbonate ions that facilitate the precipitation of transition metal cations in desired stoichiometries.

The hydrothermal method provides superior control over particle morphology and crystallinity, resulting in cathode materials with improved specific capacity, cycling stability, and rate capability. Additionally, its low energy consumption compared to traditional solid-state methods makes it an environmentally friendly alternative. However, limitations such as long reaction times, complex setups, and challenges in scaling up for industrial production persist. Despite these challenges, the hydrothermal method remains a valuable tool for synthesizing high-performance NMC cathode materials. Its adaptability and ability to produce advanced materials with tailored properties position it as a cornerstone technique in the development of next-generation lithium-ion batteries. Further research into optimizing process parameters and reducing operational complexity could pave the way for broader industrial adoption.

#### 2.4.4 Microwave-assisted hydrothermal method

Also, since the topic of this study is microwave-assisted hydrothermal method, it is important to mention this type of synthesis and talk more about it.

The microwave-assisted hydrothermal method is an advanced and efficient approach for synthesizing NMC cathode materials, particularly Ni-rich variants such as NMC811, widely used in lithium-ion batteries. This method integrates the principles of hydrothermal synthesis with the advantages of microwave heating, enabling the production of materials with superior crystallinity, uniform morphology, and controlled composition while significantly reducing energy consumption and processing time (Skvortsova et al., 2022). Unlike conventional hydrothermal methods, which rely on surface heat transfer, microwave radiation directly interacts with polar molecules and ions in the reaction mixture, causing rapid oscillation and uniform volumetric heating. This mechanism accelerates reaction rates, minimizes temperature gradients, and ensures homogeneity in the resulting materials.

The process begins with the preparation of an aqueous solution containing stoichiometric amounts of transition metal salts, such as Ni, Mn, and Co sulphates. A chelating agent like glycine or ammonium is added to stabilize the solution by forming metal-ion complexes, preventing premature precipitation and promoting uniform mixing. Sodium hydroxide is then introduced as a precipitating agent, initiating the formation of a slurry-like precursor. This mixture is transferred into a Teflon-lined stainless-steel reactor designed for microwave

heating and subjected to controlled microwave-assisted hydrothermal treatment at temperatures ranging from 160 to 200°C and pressures of approximately 6 bar for durations between 15 and 60 minutes. After the reaction, the resulting hydroxide precursors are filtered, washed, dried, and subsequently mixed with lithium hydroxide before undergoing calcination at 750–900°C to form the final NMC cathode material. The use of microwave radiation significantly enhances the efficiency of the synthesis process, reducing reaction times from hours to minutes while eliminating the need for constant monitoring of parameters such as pH, stirring rates, and reagent flow (Skvortsova et al., 2022). The rapid and uniform heating provided by microwaves also minimizes the formation of undesirable phases and impurities, resulting in highly crystalline materials with low cation disorder and a homogeneous distribution of transition metals. These characteristics are critical for achieving high electrochemical performance in lithium-ion batteries. Moreover, the method simplifies the synthesis process and reduces production costs by eliminating the need for complex equipment and intensive mechanical agitation.

Parameters such as microwave power, reaction time, and reagent ratios play a critical role in determining the properties of the final material. Higher microwave power and longer reaction times generally enhance crystallinity and particle size, while the precise molar ratios of chelating agents and precipitating agents influence the stoichiometry and compositional uniformity. NMC811 materials synthesized via this method have demonstrated excellent electrochemical performance, with tap densities reaching up to 2.9 gcm<sup>-3</sup> and volumetric energy densities exceeding 2100 mWhcm<sup>-3</sup> when optimized and blended with conventionally synthesized fractions. Such materials also exhibit high reversible discharge capacities and stable cycling performance, making the microwave-assisted hydrothermal method a promising approach for developing high-energy-density cathode materials.

## 2.5 An Overview of Materials for Lithium-Ion Batteries

Precursors for lithium-ion batteries play a key role in forming active materials for cathodes and anodes. Their synthesis determines the morphology, chemical composition, and electrochemical properties of the final materials, ultimately affecting battery performance and lifespan. The choice of synthesis method depends on the requirements for the final material, its crystalline structure, particle size, and electrochemical characteristics. In recent years,

emphasis has been placed on both the development of new approaches and the improvement of classical ones, which allow better control over structures. Ultimately, further research is focused on increasing synthesis efficiency, reducing energy consumption, and minimizing environmental impact, all of which are crucial for advancing lithium-ion battery technology.

### 3 Materials and methods

This chapter describes the process of NMC 955 cathode material synthesis using a microwave reactor with following sintering. The subchapters describe: the preparation process of the precursor material, its microwave-assisted hydrothermal synthesis and further optimisation by changing different variables such as pH, time of synthesis and amount of power. Changing these variables directly affects the morphology and electrochemical properties of the future material. Then the process of lithiation, calcination and the subsequent analysis of the electrochemical properties of the obtained cathode material by different methods.

For convenience, all work will be divided into 3 main stages. Stage 1 is synthesis of NMC precursor, it is included in chapter 3.1.1. Stage 2 is calcination and lithiation, described in chapter 3.1.2. Stage 3 is the assembly of lithium-ion batteries, described in chapter 3.1.3. Figure 3 describes the complete synthesis pathway from NMC precursor synthesis to lithium-ion battery assembly.

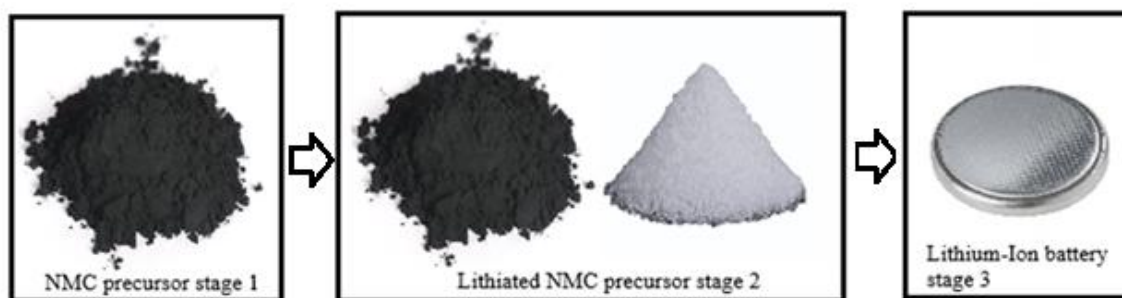


Figure 3. Complete synthesis pathway

During the experiments, the optimum parameters for the synthesis of the precursor were experimentally achieved. Therefore, this subchapter will describe the synthesis of the most successful product and in chapter 4 will describe the effects of different parameters on the final synthesis product.

### 3.1 Material preparation

#### 3.1.1 Synthesizing NMC precursor

For the microwave assisted hydrothermal synthesis of cathode material precursor, a sample of NMC with molar ratio of  $\text{Ni}_{0.9}\text{Mn}_{0.05}\text{Co}_{0.05}(\text{OH})_2$  was prepared. Masses of 2.505 g of nickel (II) sulphate hexahydrate ( $\text{NiSO}_4 \cdot 6\text{H}_2\text{O}$ , 98+ %, Thermo scientific), 0.0913 g of manganese (II) sulphate heptahydrate ( $\text{MnSO}_4 \cdot \text{H}_2\text{O}$ ,  $\geq 98$  %, MERCK) and 0.1519 g of cobalt (II) sulphate heptahydrate ( $\text{CoSO}_4 \cdot 7\text{H}_2\text{O}$ ,  $\geq 99$  %, MERCK) were used as a source of the transition metals. The sulphates were dissolved in 20 ml of distilled water and mixed for 30 min. After that 25% ammonium hydroxide solution ( $\text{NH}_4\text{OH}$ , 25 % extra pure, Thermo scientific) was added as a chelating agent to control a pH of the solution. During the addition of the chelating agent, the colour of the solution changes from dark green to dark grey. After obtaining the desired pH of 11, the sample was transferred to the microwave reactor.

In the microwave reactor (CEM Discover 2.0), the parameters were set at 180 °C, 35 min, 250 W and dynamic programme. After completion of synthesis, the solution is filtered using a vacuum filter and kept in an ultrasonic bath for 5 minutes to separate possibly sticking particles. After this procedure the obtained solid precipitate is dried in a vacuum oven (Thermo Scientific) at 60 °C for 12 hours

Figure 4 shows a simplified stage 1 synthesis scheme.

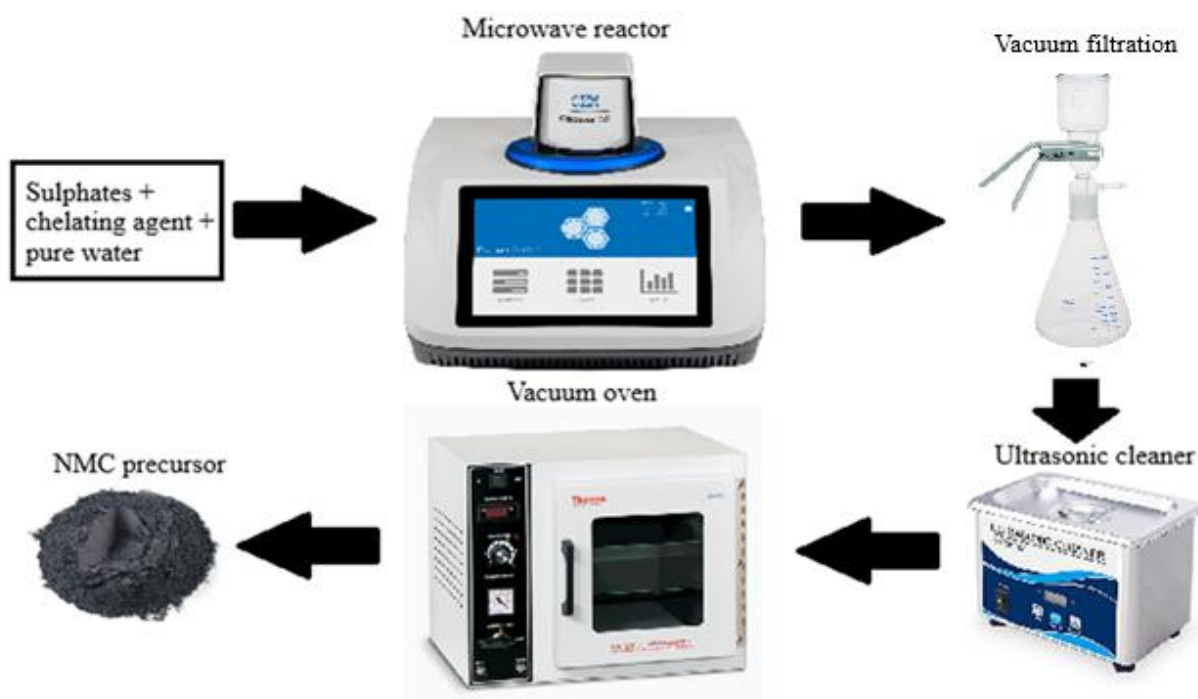


Figure 4. Schematic picture of NMC precursor synthesis using microwave reactor.

### 3.1.2 Lithiation of NMC precursor

The use of a mortar and pestle is required to distribute the material evenly while mixing the precursor and lithium hydroxide. Stirring should be done as gently as possible without excessive force so as not to spoil the shape of the precursor particles. The required mass of lithium hydroxide can be calculated using equation 2 and equation 3. It is important to note that the required mass of lithium hydroxide should be added in excess of 5% than the mass calculated by using the equations, as the lithium may volatilise while in the tube furnace.

After mixing, the sample is placed in a tube furnace under a constant oxygen flow (100 ml/min) for 6 hours at temperature 450 °C. Then, after the furnace has cooled down, it is necessary to take it out and mix it. After that, it is placed in the oven for another 16 hours at temperature 820 °C. The resulting black powder is a LNMC 955. Figure 5 shows a simplified scheme of stage 2.

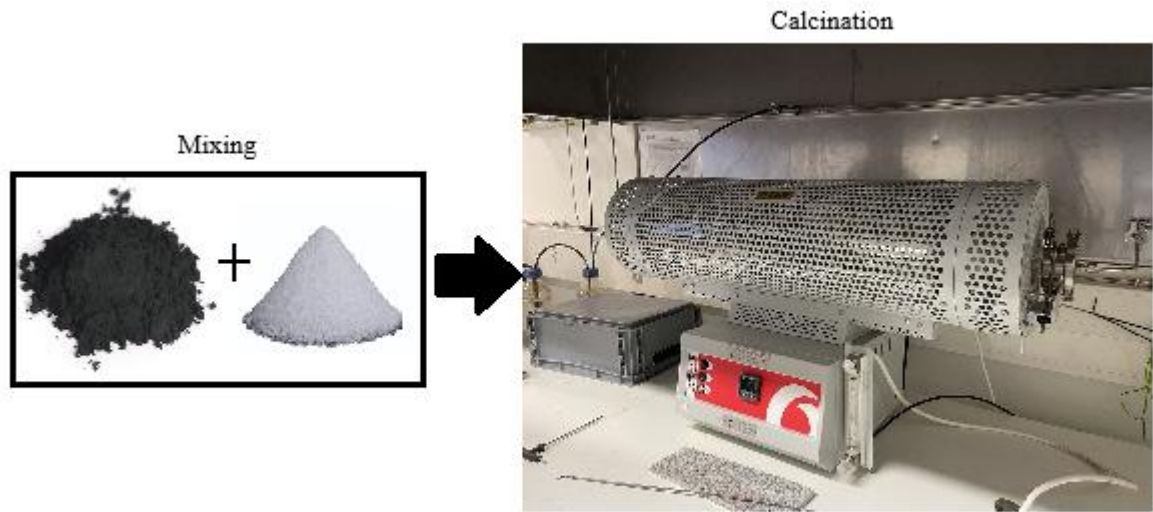


Figure 5. Schematic picture of synthesis of LNMC 955

### 3.1.3 Preparation of cathode material for lithium-ion battery

After obtaining the material from the tube furnace, it is necessary to prepare slurry for the cathode of the future battery. The slurry was prepared using 93 wt % CAM (LNMA955), 3 wt % conductive CB powder (Timcal Super C65), and 4 wt % PVDF (Solvay, Solef 5130) suspended in NMP (Thermo Scientific, 99.0 +%) as the solvent.

The obtained cathode slurry was spread on aluminum foil (MTI, 20  $\mu\text{m}$ ) using automated film coater (Neware). The aluminium foil with the slurry on it was then put to a vacuum oven (Thermo Scientific) for 60  $^{\circ}\text{C}$  for 12 hours.

### 3.1.4 Assembly of lithium-ion batteries for further electrochemical analysis

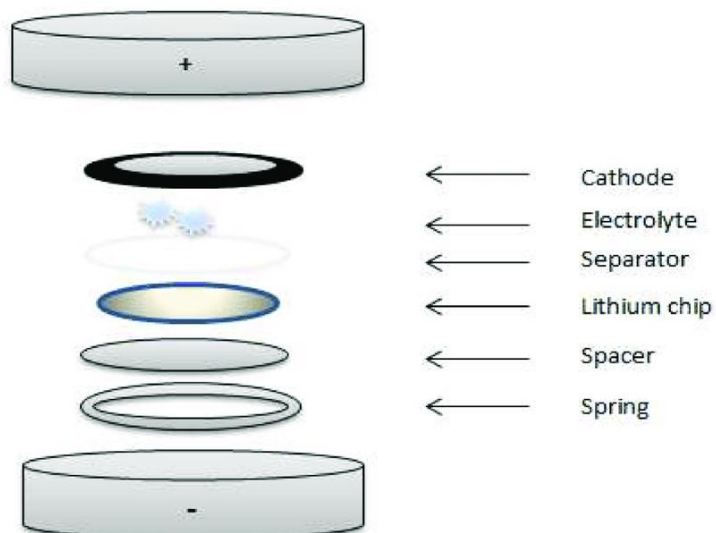


Figure 6. The schematic picture of the half-cell assembly of coin cells in glovebox

Once the material has been taken from the vacuum oven, it is possible to start assembling the lithium-ion battery. The process is shown in the figure 6. The assembly is carried out in an Argon-filled glovebox (Jacomex). To assemble the batteries were used: Lithium chips (16 mm) as an anode, PP film (25  $\mu\text{m}$ ) as a separator and a  $\text{LPF}_6$  in EC/DMC/DEC (1.0 M) as electrolyte.

## 4 Results and discussion

This chapter summarizes the results of synthesizing NMC955 precursors using microwave-assisted hydrothermal synthesis. The primary objective of the research was to produce particles with a spherical morphology, size distribution about 10  $\mu\text{m}$  and satisfying electrochemical properties. To begin with, it would be worth noting that all the changes described in chapter 4.1 affect directly the morphology and particle size, which is the main reference point in synthesis. In synthesis, there are a lot of easily changeable parameters that can greatly affect the synthesis product, such as pH, pressure, temperature.

Table 1. Synthesis conditions for each run.

Run #	Temperature (°C)	pH	Time (min)	Power (W)
1	180	10	30	250
2	180	11.15	30	250
3	180	11.45	30	250
4	180	11.15	30	200
5	180	11.15	30	250
6	180	11.15	30	300
7	180	11.15	30	250
8	180	11.15	40	250
9	180	11.15	60	250

## 4.1 Effects of Synthesis Condition on Morphology and Size Distribution

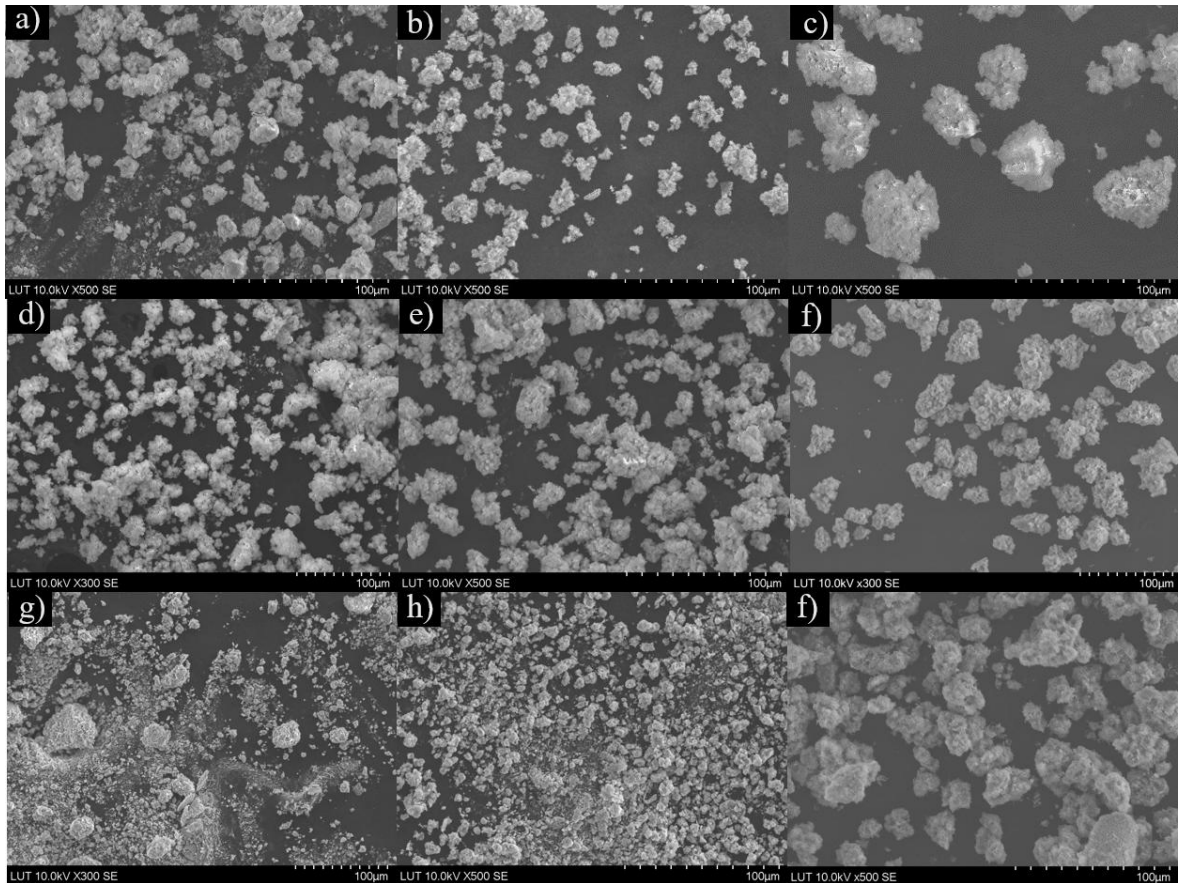


Figure 7: SEM images of how different synthesis parameters affect morphology and size distribution described in table 1. a) run 1 b) run 2 c) run 3 d) run 4 e) run 5 f) run 6 g) run 7 h) run 8 f) run 9

### 4.1.1 Effect of $\text{NH}_4\text{OH}$

During the test syntheses and based on the literature, it was found that the pH of the solution after the addition of  $\text{NH}_4\text{OH}$ , directly affects the morphology and particle size. A series of tests were carried out to determine the optimum pH to obtain the desired properties (at an optimal power of 250W, 30 minutes, 180 degrees). Figure 7 (a,b,c) shows that the optimal pH is 11.15, because the average size distribution is about 10  $\mu\text{m}$  and the morphology is close to spherical (Malik, Chan and Azimi, 2022). From these visible changes, it can be concluded that: as the pH increased, the particles became sharper and lost their sphericity,

and as the pH decreased, they clumped together into larger particles. According to Barai et al. it is possible to make the same conclusion, which confirms the results of the experiment.

#### 4.1.2 Effect of Power Adjustments

One of the main parameters is the change in a power of microwave reactor. Once the optimal pH parameter was chosen, it was decided to try changing the power parameters to obtain a morphology closer to that obtained in the literature (Malik et al., 2022; Skvortsova et al., 2022) - a rounder and less “ripped” texture. During the synthesis, three different settings were tried, where the power was set to 200, 250 and 300 W (at an optimal pH of 11.15, 30 minutes, 180 degrees). As shown in figure 7 (d,e,f), the result was that as the power was increased and decreased, the particles clumped together. No particularly strong difference was detected.

#### 4.1.3 Effect of Synthesis Time

According to (Skvortsova et al., 2022), no significant difference in morphology was observed when the temperature parameters were changed in the microwave reactor, but it was still decided to test this adjustment as well, since the synthesis time is very short.

Three test syntheses were performed, one for 30 minutes, second for 40 minutes and the third for 60 min (at an optimal pH of 11.15, 250 W, 180 degrees). As the time increases, it is clearly visible in figure 7 (g,h,f) that the particles significantly increase in size. No significant changes in morphology were observed. The second synthesis appeared to be the most successful, so it was decided to use it in the future.

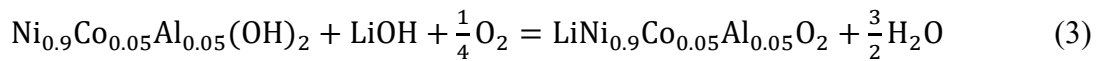
#### 4.1.4 Optimal Parameters for Further Synthesis

By summarising all the data from previous experiments, the optimal parameters for future synthesis were chosen. The main parameter that affects morphology and size, compared to the others, was pH. With a small change in pH, the difference in the obtained result could be significant. This confirms the statements in the literature (Barai et al., 2019).

The optimal parameters for continuing the synthesis were selected based on the SEM analysis. 40 min, pH - 11.15, 250W, 180 °C. Two lithiations were performed, but the initial sample was the same.

## 4.2 Lithiated NMC Oxide

Two experiments were conducted. The first experiment was conducted under the conditions of 4h 400 °C, 12h 800°C. After the XRD (chapter 4.2.2.) and electrochemical analysis results (chapter 4.2.3.), it was decided to perform a second experiment with increasing temperature and time. According to (Kong et al., 2024) increasing the calcination temperature should improve the electrochemical properties of the lithiated material. The second experiment was conducted under the conditions of 4 h 450°C, 16 h 820°C. The lithiation process of NMC 955 went according to the chemical equation (3):



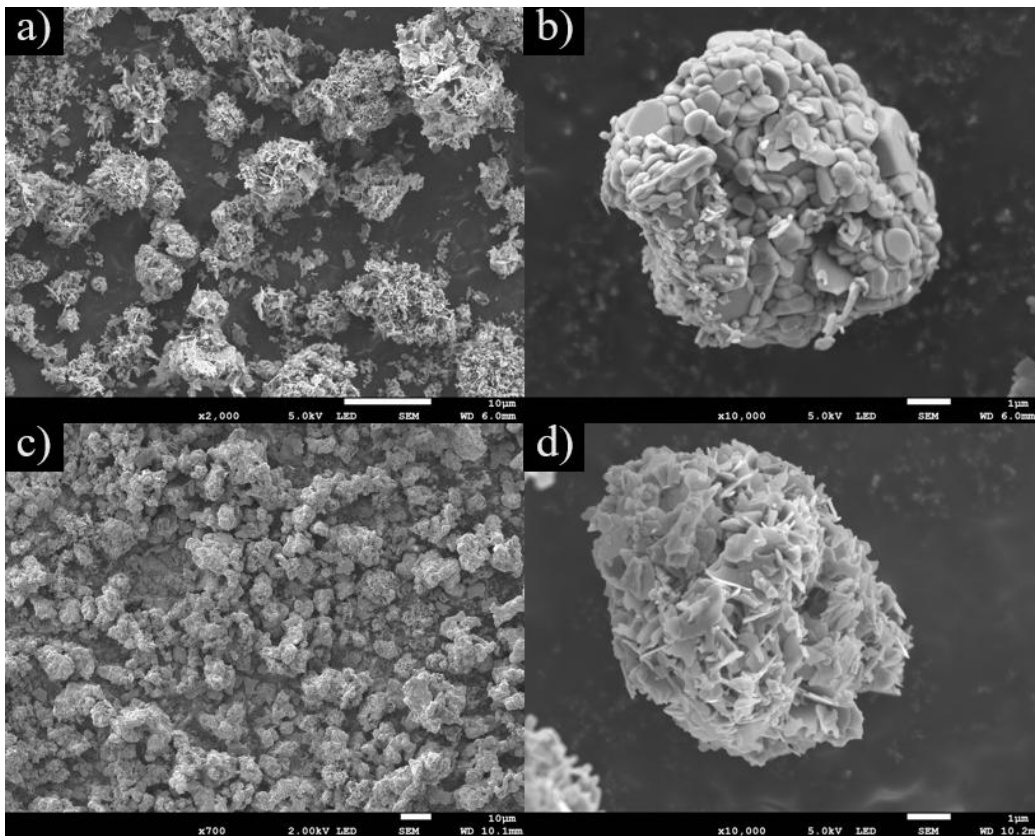


Figure 8: SEM images of LNMC. (a,b) LNMC 4h 400 °C, 12h 800°C, (c,d) LNMC 4h 450°C, 16 h 820°C

#### 4.2.1 Morphology of Lithiated Material

As shown in figure 8 (a,b) the material of the first experiment was not completely lithiated. This can be indicated by the porous texture of the particles and the “flakiness” of single particles. By visible analysis of the material from the second experiment as provided in figure 8 (c,d), it is possible to notice changes in the homogeneity of the particle texture. The texture became less “shattered”. The morphology of single particles did not change much and the average size of the single particle of the 10 μm.

#### 4.2.2 Results of XRD analysis

For convenience, the “LNMC1” will be given for the first experiment and “LNMC2” for the second. They will be referred to as follows later in the text.

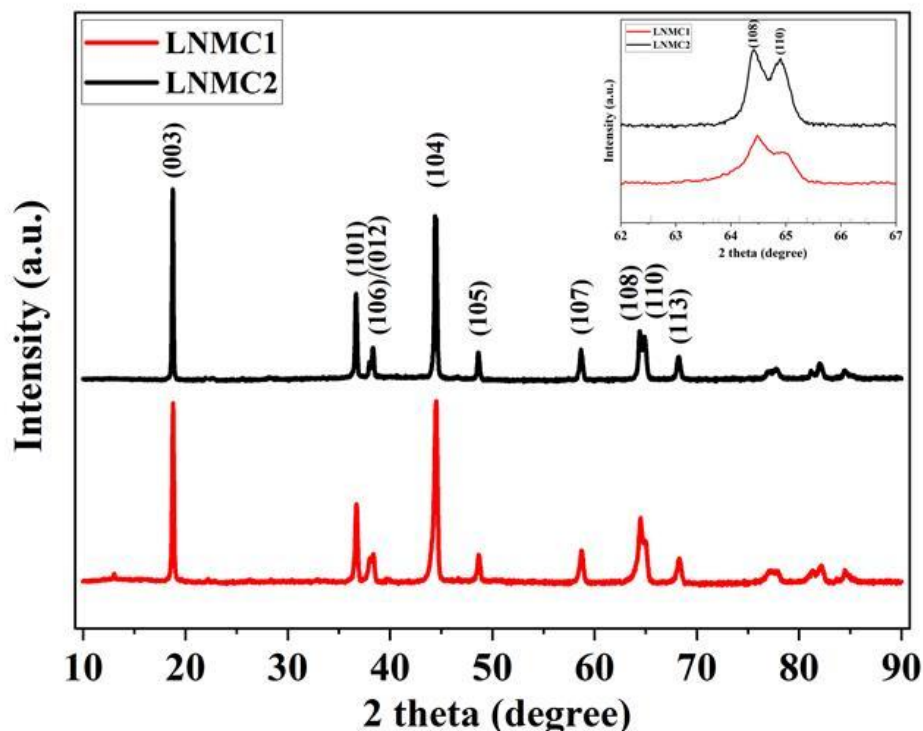


Figure 9. XRD patterns that show two LNMC samples

The diffraction peaks are indexed with Miller indices (hkl), which correspond to the hexagonal  $\alpha$ - $\text{NaFeO}_2$ -type layered structure (space group:  $R\bar{3}M$ ), commonly observed in NMC materials. The peaks at specific  $2\theta$  values (e.g., (003), (104), (101), (107), (108), (110), (113)) confirm the formation of the layered structure. The absence of additional peaks suggests that both samples are phase-pure, meaning there are no significant impurity phases like spinel ( $\text{LiMn}_2\text{O}_4$ ) or rock-salt (NiO) phases.

The splitting of peaks (108) and (110) serves as an indicator of the formation of a layered structure, which leads to a detailed analysis of the amplification of these peaks. A pronounced peak splitting is observed at a calcination temperature of 820 °C for 16 h, resulting in the formation of a well-defined layered structure. On the other hand, at 800 °C for 12 h,

the diffraction peak splitting is minimal, indicating that higher calcination temperature plays a crucial role in affecting the layered structure of the material.

An intensity ratio of the (003) to (104) diffraction peaks greater than 1.2 indicates the degree of Li/Ni minimal cation mixing, which contributes to a more stable structure and improved electrochemical performance (Kalyani, Kalaiselvi 2005).

In this study, LNMC1 exhibits a (003)/(104) ratio of 1.25, exceeding the stability threshold. This suggests a well-ordered arrangement of lithium and nickel ions within the layered structure, leading to enhanced structural integrity and superior electrochemical properties. In contrast, LNMC2 has a (003)/(104) ratio of 1.01, which is below the critical value of 1.2. This lower ratio indicates increased Li/Ni cation mixing, resulting in greater structural disorder and potentially hindering lithium-ion diffusion. Consequently, the electrochemical performance of LNMC2 may suffer, with effects such as higher impedance and capacity fade over time.

Overall, the observed differences in the (003)/(104) intensity ratio suggest that LNMC2 is more likely to exhibit stable electrochemical behavior, including better capacity retention and cycle stability, whereas LNMC1 may suffer from performance degradation due to structural instability.

### 4.2.3 Electro-chemical performance of the assembled batteries

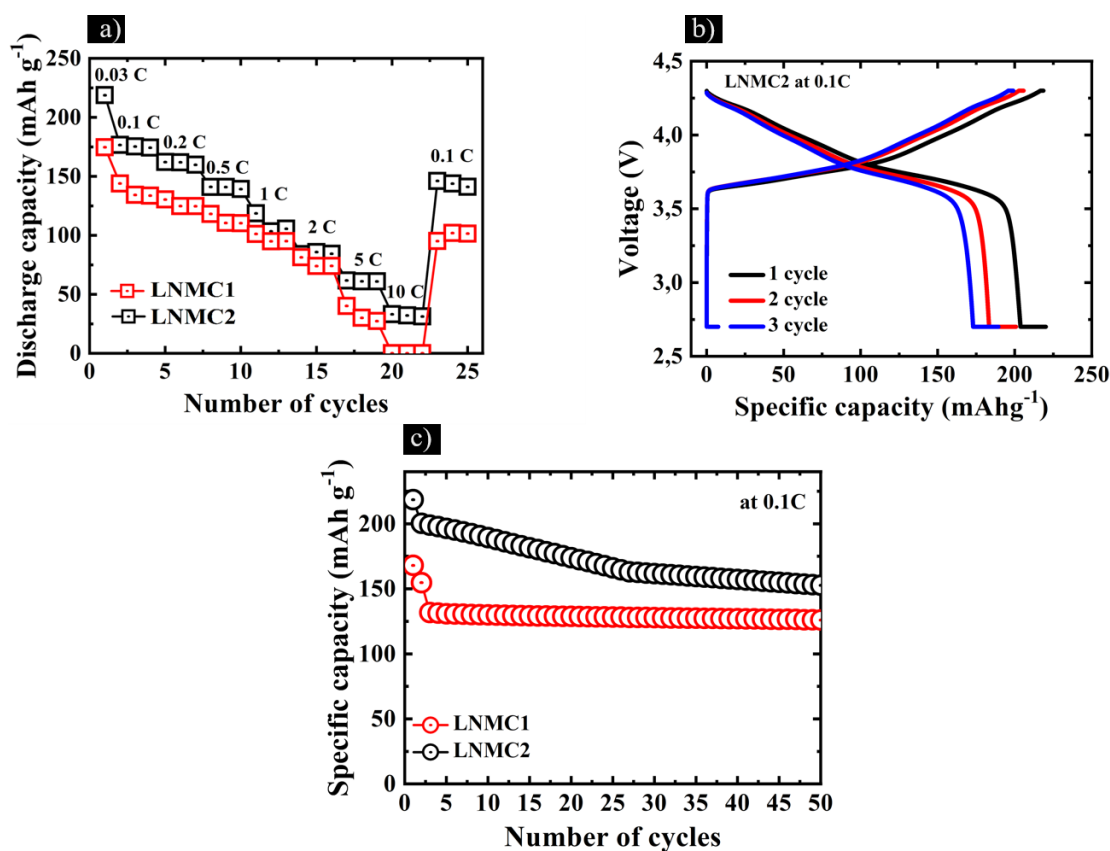


Figure 10. a) Rate capability of the LNMCM1 and LNMCM2 at C-rates 0.03C-10C. b) The initial charge-discharge curves of the LNMCM2 at 0.1 C c) Cycle stability of the LNMCM1 and LNMCM2.

The material was electrochemically analysed using the charge-discharge experiment. In figure 10a is shown that the maximum discharge capacity of LNMCM1 is much higher, than the maximum discharge capacity of LNMCM2. For the LNMCM2 discharge capacities are equal to 220, 175, 160, 132, 100, 75, 52, 30 mAhg<sup>-1</sup> at the normalized current rate of 0.03, 0.1, 0.2, 0.5, 1, 2, 5, and 10 C. For the LNMCM1 discharge capacities are equal to 175, 130, 125, 105, 95, 75, 25, and 1 mAhg<sup>-1</sup> at the normalized current rate of 0.03, 0.1, 0.2, 0.5, 1, 2, 5, and 10 C. At higher C-rates, the discharge capacity decreases due to increased internal resistance, slower ion diffusion, and incomplete active material utilization (Yerkinbekova et al., 2025).

With the increase of the lithiation temperature and time, the discharge specific capacity of material became better and more stable. The reason for this is not fully formed LNMCM

particles, at a lower temperature, which is also confirmed by the results of XRD analysis in the chapter 4.2.2.

The initial charge-discharge profiles of the LNMC2 at the voltage range of 2.7 to 4.3 V are shown in figure 10b. The initial discharge capacities are: 205 mAhg<sup>-1</sup> for the first cycle, 180 mAhg<sup>-1</sup> for the second cycle and 170 mAhg<sup>-1</sup> for the third cycle. According to (Yerkinbekova et al., 2025) average specific capacity for LNMC955 is approximately 210 mAhg<sup>-1</sup>.

As can be seen from figure 10c at a cycle count of 50, LNMC2 shows better capacity compared to LNMC1. There is also a smoother decrease in capacity with the number of cycles, while LNMC1 performs much worse. The resulting LNMC 2 has the desired electrochemical properties, thereby confirming that the synthesis was carried out under suitably selected conditions.

## 5 Conclusions

Summarizing all the results obtained in this study, we can conclude that the hydrothermal synthesis of the precursor using microwave irradiation was successfully conducted. It was demonstrated that this synthesis is effective, and SEM analysis confirmed that the resulting precursor exhibits relatively good morphology and particle size. It is important to mention that while the shape and size of the particles were not ideal, they were close to values reported in previous experiments and literature. It is possible that by modifying the synthesis conditions, even more optimal particle shape and size can be achieved, as well as a significant reduction in synthesis time. The key variables affecting the synthesis process were found to be pH and reaction time. XRD analysis also confirms that the lithiation of the material should be performed at a higher temperature, because at lower temperatures, hydroxide is probably not lithiated or nickel and lithium cations tend to mix, increasing disorder in the material's structure. Additionally, to properly analyse the obtained precursor, lithiation of the material was carried out for electrochemical testing, which was also successfully completed. Electrochemical tests revealed that the material obtained at an increased calcination temperature performed significantly better than the previous one. Its electrochemical properties are nearly comparable to those found in literature and previous experiments. This suggests that hydrothermal synthesis using microwave irradiation is a promising area for future research, given the ongoing trend of reducing cobalt and increasing nickel concentration in precursor materials for cathodes.

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