

UNIVERSITI TEKNOLOGI MARA

**CO-PRECIPIATION SYNTHESIS
OF $\text{LiNi}_{0.59+x}\text{Co}_{0.3-x}\text{Ti}_{0.1}\text{Al}_{0.01}\text{O}_2$ (NCTA)
AS NI-RICH CATHODE
MATERIALS AND THEIR
ELECTROCHEMICAL
PERFORMANCE**

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ABSTRACT

The increasing adoption of electric vehicles (EVs) has driven the demand for efficient, sustainable, and cost-effective cathode materials. Nickel-rich cathode materials (Ni content more than 50%) are highly promising due to their high energy density. However, Ni-rich cathode materials suffer from high capacity fading due to their structural instability and particles microcracking after repeatedly undergo charge/discharge process. Herein, a single-crystal $\text{LiNi}_{0.59+x}\text{Co}_{0.3-x}\text{Ti}_{0.1}\text{Al}_{0.01}\text{O}_2$ (NCTA) was synthesized using a co-precipitation method followed by high annealing temperature with excess lithium. Three synthesis parameters such as washing durations, annealing temperatures and nickel-to-cobalt ratios were optimized and the effects on the structural, morphological, and electrochemical properties also studied systematically. In the first part, co-precipitation synthesis of NCTA_0.59 ($\text{LiNi}_{0.59}\text{Co}_{0.3}\text{Ti}_{0.1}\text{Al}_{0.01}\text{O}_2$) that annealed at 920 °C was washed with deionized water at different duration. Rietveld refinements revealed that the washed sample exhibited lower cation mixing compared to the unwashed sample showed that the unwashed samples had particle agglomeration and existing of Li residue on the surface of the cathode materials. Interestingly, FESEM revealed that after the washing treatment, the NCTA appeared as single crystals particles with clean, smooth surfaces. Charge/discharge studies demonstrated that the washed sample showed a significant increase in initial discharge capacity compared to the unwashed sample. The optimal washing duration was found to be 15 minutes (W15), resulting in the best initial discharge specific capacity of 161.86 mAh/g at 0.1 C with a capacity retention of 81.9% after 100 cycles at room temperature. In the second part, NCTA single-crystal cathode materials, synthesized via co-precipitation were subjected to different annealing temperatures (900°C, 920°C, and 950°C). The results revealed that 920°C annealing temperature is an optimal crystallinity, reduced cation mixing, and enhanced electrochemical stability, with capacity retention of 81.90% after 100 cycles. Lastly, the effects of lowering Co content by compensating with Ni content in $\text{LiNi}_{0.59+x}\text{Co}_{0.3-x}\text{Ti}_{0.1}\text{Al}_{0.01}\text{O}_2$ ($x = 0, 0.06, 0.1$), to form NCTA_0.59, NCTA_0.65 and NCTA_0.69 cathode materials. XRD results showed that all samples were pure and single phase with the RIR value more than 1.2 indicating a well-ordered layered structure. It was revealed that the initial discharge capacity was increases as the Ni content increased. The NCTA_0.69 achieved the highest initial discharge capacity of 202 mAh/g with capacity retention of 73% after 50th cycles. This study confirmed that the co-precipitation synthesis method at optimum annealing temperature and washing duration is possible to form the Ni-rich NCTA cathode materials. The formation of NCTA with highest Ni-to-Co ratios can enhance the battery capacity but the capacity retention can be improved further. Overall, the study is providing valuable insights into the design of single crystal nickel-rich cathode materials for high-performance lithium-ion batteries.

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TABLE OF CONTENTS

	Page
CONFIRMATION BY PANEL OF EXAMINERS	ii
AUTHOR'S DECLARATION	iii
ABSTRACT	iv
ACKNOWLEDGEMENT	v
TABLE OF CONTENTS	vi
LIST OF TABLES	ix
LIST OF FIGURES	xi
LIST OF SYMBOLS	xv
LIST OF ABBREVIATIONS	xvi
LIST OF NOMENCLATURE	xviii
CHAPTER 1 INTRODUCTION	1
1.1 Background of Study	1
1.2 Problem Statement	2
1.3 Significance of Study	4
1.4 Objectives of Study	4
1.5 Scope and Limitation of Study	5
1.6 Research Novelty	5
CHAPTER 2 LITERATURE REVIEW	6
2.1 Growth and Development of Lithium Batteries	6
2.2 Components in Li-ion Battery Technology	7
2.2.1 Anode (Negative Electrode)	9
2.2.2 Electrolyte	11
2.2.3 Separator	13
2.2.4 Cathode (Positive Electrode)	14
2.3 Mechanism of Li-ion Battery	16
2.4 Types of Cathode Materials	17
2.5 Modified Lithium Nickel Oxide	20

CHAPTER 1

INTRODUCTION

1.1 Background of Study

The growing demand for electric vehicles (EVs) has driven a surge in the advancement of high energy density, cost-effective, nickel-rich (Ni-rich) cathode materials are gaining attention as a promising option because of their high energy density and lower dependence on cobalt (Co), a costly and critical element in cathodes [1], [2], [3]. By minimizing Co content, the overall cost of EV batteries can be significantly lowered, making EVs more affordable and accessible [4], [5]. Ni-rich cathode materials, including NCA (nickel-cobalt-aluminum) and NCM (nickel-cobalt-manganese), have recently gained significant interest due to high capacity, making them crucial for high-performance applications [6], [7]. Although these materials offer significant benefits, they encounter issues related to stability and sustained performance over extended periods [7], [8]. To tackle these challenges, elements such as titanium (Ti) have been used to dope NCA materials [9]. Ti-doped shows potential stabilizing the cathode structure, improving cycling stability, and mitigating common issues such as capacity fading. This modification effectively maintains the integrity of Ni-rich materials during prolonged usage, thereby extending the battery lifespan [10], [11].

A critical aspect of cathode material performance lies in the crystal structure, with single-crystal cathode materials demonstrating superior electrochemical stability compared to their polycrystalline counterparts [12]. Single-crystal materials, lacking grain boundaries, show slower degradation, less impedance growth, and better resistance to cracks during charge-discharge cycles, resulting in better capacity retention and longer cycle life [13], [14]. In contrast, polycrystalline materials, composed of multiple crystallites, often suffer from grain boundary-related issues, such as increased surface area for side reactions and mechanical stress concentration, resulting in faster capacity fade and decreased overall battery performance [15]. The inherent structural advantages of single-crystal materials make them a compelling choice for high-performance and durable applications [14], [16]. The synthesis of single-crystal cathode materials is an advanced process that requires precise control over growth conditions to achieve optimal crystallinity and performance [17], [18].