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Functional Materials for a Sustainable Replacement of the Lithium-ion Battery

Bachelor's thesis in BKJ
Supervisor: Mari-Ann Einarsrud
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Abstract

In recent years, there has been a growing concern about the impact of fossil fuels on the environment and the need for sustainable energy sources. As a result of this there has been an increased need for renewable energy sources such as solar, wind and hydropower. However, the intermittent nature of these sources has made it challenging to meet the energy demands of modern society.

Batteries have the potential to play a crucial role in the transition to a more sustainable future. They can store excess energy from renewable sources and provide it when needed. Moreover, with the rapid advancements in battery technology, they are becoming more efficient, durable and cost-effective. The most rapid used battery today is the lithium-ion battery (LIBs), but there has been several safety -and sustainability concerns about them. There has been incidents where the batteries have caught fire, or even exploded, due to thermal runaway or overheating. The production and disposal of the batteries are not developed to a sustainable level yet, and there are also severe problems when it comes to the mining of the raw materials used in the LIBs.

This scientific report will take a look into more sustainable materials that can be used as alternatives at the cathode, anode and as electrolyte in the LIB. New battery chemistries which might replace the LIB are also presented before the recycling process of the different parts of the battery is discussed.

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1 Introduction

The world is currently experiencing severe climate changes due to the long-term excessive use of non-renewable fossil fuels¹. Fossil fuels have been our primary energy source for a long time, and an increasing concentration of carbon dioxide (CO₂) and other greenhouse gasses (GHGs) in the atmosphere have led to a warmer climate.³³. The consequences of global warming are many, such as polar ice melting, reduced biodiversity and more extreme weather²⁶.

The demand for sustainable energy sources based on renewable resources is growing rapidly and it has led to an increasing amount of research on battery energy storage systems (BESS)¹¹. Within this field of research, the lithium-ion battery (LIB) is regarded as the most promising option for portable devices that need lightweight and small batteries that can store a lot of energy, such as smartphones, laptops and electric vehicles¹⁵. Although, it has some improvement potential when it comes to sustainability and safety⁴².

The lithium ion battery is considered a "green" battery due to their high energy density and long lifespan⁴⁵. However, taking a look inside of the battery and the production of it, gives reasons to believe that it is a way to go before we can actually call it green. Several parts of the battery consists of flammable and toxic materials which are geographically distributed and the mining of these materials is far from ethical nor sustainable⁴⁵.

For batteries to be a sustainable alternative to fossil fuels they need to be optimised to be effective, have a long life span and not have a negative impact on the environment and humans when the materials for them are mined¹¹. Today, the manufacturers of the LIB use non-abundant materials such as lithium, cobalt and natural graphite. The cobalt mining is highly disputed due to child labour and otherwise poor working conditions, the life span of the battery is shortened because of flammable materials and the recycling process is not satisfactory⁴⁵.

The aim of this paper is to research how the lithium-ion battery can be optimised by replacing the present materials at the cathode, anode and as electrolyte and separator to be more suitable as an alternative to today's use of fossil fuels. It will take a look into the chemistry of the battery to locate weaknesses about the present composition of it and also find solutions to how it can become more sustainable through its whole life from mining to recycling. New chemistries for the cathode and the anode will be suggested and discussed as potential replacements for the current electrode materials.

2 Theory

The theory on electrochemical (electrolytic and galvanic) cells, secondary batteries and charging/discharging of the LIB is referenced from Electrochemical Science and Technology²⁹.

2.1 Electrochemical cells

Electrochemical cells are devices that can either convert electric energy to chemical energy or the other way around due to non-spontaneous or spontaneous reduction and oxidation reactions respectively. These electrochemical reactions occur due to a difference in electrochemical potential between two electrodes called the cathode and the anode. The reduction reaction will occur at the cathode, while the anode holds the oxidation reaction. The difference in inner electrochemical potential, ϕ gives the cell potential, ΔE_{cell} for an electrochemical cell and is given by the equation:

$$\Delta E_{cell} = \phi_{cathode} - \phi_{anode} = -\frac{\Delta G}{nF} \quad (1)$$

where ΔG is the Gibbs free energy of the total cell reaction, n is the number of electrons transferred and F is the Faraday constant.

2.1.1 Electrolytic cells

Electrolytic cells are electrochemical cells that convert electric energy to chemical energy by adding a power source to the outer circuit that will drive a non-spontaneous reaction. The amount of potential that has to be added for the reaction to occur has to be opposite and larger than, ΔE_{cell} . In an electrolytic cell the anode is the positive electrode, whilst the cathode is the negative one, therefore the electrons flow from the cathode to the anode in an outer circuit.

2.1.2 Galvanic cells

Galvanic cells convert chemical energy into electric energy due to spontaneous red-ox reactions. In a galvanic cell, the positive electrode is the cathode and the electrons will be drawn here. Simultaneously, the ions from the electrolyte will be drawn to the negative anode to establish electric neutrality.

2.2 Secondary Batteries

The electrochemical cell is classified as a battery when it is self-contained and does not need any supplement of reactants. In a secondary battery the cell reactions are reversible. These batteries will therefore operate galvanically when they are discharged and electrolytically when being charged. Active materials are the materials in the battery that is consumed as the battery is being discharged.

The performance of a battery depends on a series of properties such as capacity, cycle life, power density and safety. The cycle life of a battery defines how many times it can be recharged before the capacity is degraded to a certain level. The shelf life of the battery refers to the amount of time it can be stored before it begins to deteriorate. The capacity is the amount of energy a battery can store and for how long it can power a device. Specific energy or energy density is the energy delivered by a given mass or volume of the battery. This is an important value for battery design since high specific energy makes them more efficient and practical for several applications. Specific energy is defined as:

$$\text{Specific Energy} = \frac{\text{Capacity} \cdot \text{Voltage}}{\text{Total Mass}} \quad (2)$$

29.

2.3 The chemistry of the lithium-ion battery

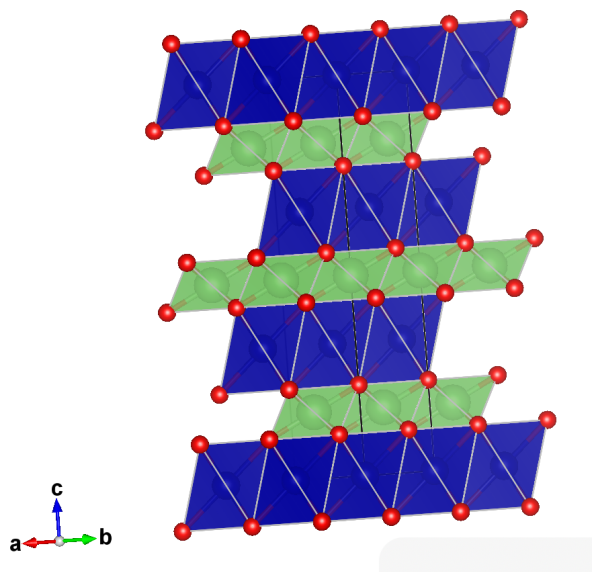
A lithium-ion battery utilise insertion reactions for both positive and negative electrodes with Li ions and is therefore also known as "rocking chair" or "shuttle" battery technology¹³. Lithium is used because it is the most electropositive (-3.04 V vs. SHE) and also the lightest ($M = 6.94 \text{ g mol}^{-1}$) metal². The conventional lithium ion battery (LIB) is built up of a cathode, an anode and a separator with an absorbed electrolyte between these electrodes¹³. The Li-ion battery technology has been under steady development and the very first stages of it used lithium cobalt oxide (LiCoO_2) as cathode active material which deintercalates and intercalates Li around 4 V⁴¹. The lithium ion is small and can therefore intercalate a crystalline solid without changing the structure of the host. Intercalation requires an inflow of electrons as well to preserve electroneutrality²⁹. The lithium-ion cell is a versatile secondary battery, and due to its high energy density it can be implemented in portable consumer electronics³⁶.

Table 1: Voltage range, specific energy, energy density, life cycle at 80% depth of charge¹¹ and average life span⁴⁵ of the lithium-ion battery

Voltage range [V]	Specific energy [W h kg ⁻¹]	Energy density [W h L ⁻¹]	Life cycle at 80% DoD	Average life span [years]
4.0-2.5	150	400	3000	3-10

2.3.1 Cathode

The cathode of the lithium-ion cell consists of lithium ions (Li^+) intercalated in a metal-oxide lattice⁵². A variety of metals have been used to gain different battery characteristics, some of them being cobalt (Co), manganese (Mn) and nickel (Ni).⁵². Elemental lithium is strongly reactive, and for this reason, Li^+ is used²⁹. Because of the ions' convenient size, they do not significantly change the crystalline structure of the metal-oxide host and this is important to maintain a battery with a long life cycle that doesn't degrade over time⁵².

**Figure 1:** The structure of LiCo_2 in VESTA with structural parameters and lattice parameters referenced from Momma et al.²⁷

2.3.2 Anode

The anode is usually made up of graphite layers which are not strongly bonded to each other and can store lithium ions between them⁵². The anode active materials accept Li ions when the cell is being charged, and release the ions when discharged²⁹. Like the cathode process, the anode insertion -and extraction of Li ions do not

significantly change the graphite structure⁵². The anode active materials generally have lower potentials than the standard hydrogen electrode (SHE)²⁹.

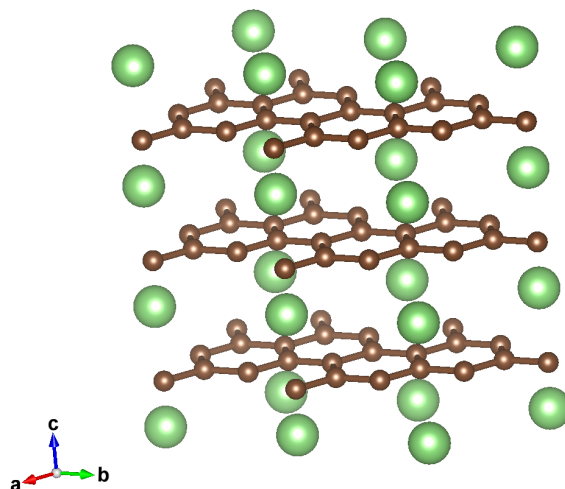


Figure 2: The structure of LiC_6 in VESTA with structural parameters and lattice parameters referenced from Momma et al.²⁷

2.3.3 Electrolyte and Separator

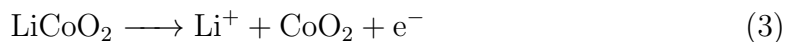
The electrolyte's role in the electrochemical cell is to create a channel between the electrodes that can transfer ions²⁹. Important properties of the electrolyte is low electronic conductivity, high ionic conductivity, thermal stability and environmentally benignity³. Another important property is the ability to form the protective solid electrolyte interface (SEI) layer on the surface of the anode materials in the LIB, due to electrolyte decomposition which improves the cyclability and safety of the battery⁴⁹. The electrolyte in today's conventional lithium-ion batteries is typically a lithium salt (e.g. LiPF_6) dissolved in an aprotic organic carbonate/polymer²⁹.

The separator is an essential component in liquid electrolyte LIBs because it physically separate the electrodes and therefore prevents electrical short circuiting²⁹. The separator is porous and absorbs the electrolyte and allows the flow of ions, and the ions only, between the electrodes⁷. It acts as an isolator since it is not electrically conductive and it ensures safety by separating the anode and the cathode⁷.

2.3.4 Charging of the lithium-ion battery

Charging of the lithium-ion cell is an electrolytic process. The lithium ions is intercalated in the metal oxide lattice at the cathode. When the charger is connected, the

electric current (I) will flow towards the cathode and lithium ions will be released from the metal oxide through the separator. A simplification of the cathodic lithium oxidation reaction during charging of the lithium-ion battery is:



At the anode the lithium ions intercalate the graphite layers and lithiated carbon (LiC_6) is formed. The electrons travel through an outer circuit from the cathode to the anode (opposite direction of the current, I). The anodic lithium reduction reaction can be written as:



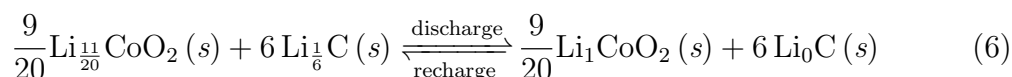
When the battery is fully charged, the graphite layers will hold all the lithium ions and the electrons. The total cell reaction for the charging of a lithium-ion cell is:



2.3.5 Discharging of the lithium-ion battery

When the power source is removed and replaced by a load (e.g. a light bulb), the cell will behave galvanically. The lithium ions will now spontaneously move from the anode towards the cathode, where lithium metal oxide is again formed. The electrons will move the same way through the cord (from anode to cathode) and produce electricity.

The lithium content in the cell will decrease progressively over time because the ions will be trapped in a layer of SEI at the anode as it is being charged and discharged repeatedly. A more accurate cell reaction will therefore be:



A great deal of research has been performed to substitute some of the materials in the conventional Lithium ion battery due to a variety of reasons, and in this paper, some of the suggestions will be presented, starting off with mentioning the issues the cell is facing today.

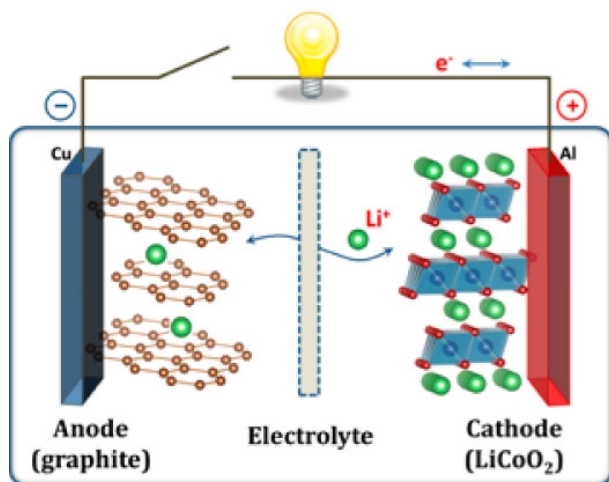


Figure 3: Standard LIB with Li-ions flowing through an electrolyte intercalating a graphite anode and a LiCoO_2 cathode¹⁰

2.4 Disadvantages and sustainability issues connected to the LIB

2.4.1 Disadvantages

Even though the lithium-ion cell is one of the more promising BESSs, it has some challenges, such as overcharging that leads to thermal runaway, gas evolution and dendritic lithium growth⁴⁴. To avoid these safety issues to occur, the internal voltage, temperature, pressure and current need to be controlled⁴⁴. For the battery to maintain safe and operative, its voltage must never rise above 4.2 V or fall below 2.0 V and it must have a constant current until the battery reaches 4.0 V²⁹. The safety issues associated with the LIBs may arise under mechanical abuse such as crush, nail penetration, drop, vibration, or electrochemical abuse behaviours such as overcharge, over-discharge, short circuits of full or half charged batteries. Mechanical abuse is likely to destroy the battery immediately due to cell thermal runaway⁴⁴.

2.4.2 Sustainability

The lithium-ion battery is the most convenient battery on the market today due to its high energy density, long lifespan and high battery capacity⁴¹. The battery gives a reduction in carbon footprint and makes us less dependent on fossil fuels while they're being used³⁰. Yet, the development and waste managing have a long way to go before we can call it sustainable. It is estimated 200-500 million tons of spent LIB and since the batteries consist of several component, the recycling is

challenging⁴⁵. There is also difficult to create sorting and recycling infrastructure for these batteries since different manufacturers uses different LIB chemistries¹⁴. Until recently, LIBs tend to prioritise electrochemical performance and costs over recycling responsibilities. There is also a significant limitation in the amount of lithium and cobalt available and a geographical imbalance in where they are found⁶. Australia, Chile and Argentina holds 80% of the world's Li reserves and 59% of the world's cobalt reserves comes from the Democratic Republic of Congo³⁸. The demand for both natural and synthetic graphite has also increased and is predicted to increase even more in the future because of the growth in the production of electric vehicles and energy storage systems³⁸. There are also several ethical questions when it comes to human exploitation related to the mining of cobalt³⁸.

Sustainability includes both environmental and social impact³⁸. The environmentally impact concerns the expected lifetime of the battery, the GHG emissions from the entire life cycle, the abundance of the materials used and the use of non-renewable resources associated with battery manufacturing³⁸. Recyclability and the potential damage to ecosystems and human health across the supply chain are also factors to have in mind regarding sustainability for the LIB³⁸. The social impact is also important to produce sustainable batteries. Human rights, health and safety is obviously important to consider as a part of the LIB manufacturing process.

3 Discussion

3.1 How to make the lithium-ion battery more sustainable

3.1.1 Cathode

As mentioned before, LiMO_2 ($M = \text{Co}, \text{Ni}, \text{Mn}, \text{Al}$) represent the current cathode. By 2015, 32% of lithium and 46% of cobalt were used for battery production²². Since the abundance of lithium is somewhat limited, new cathode materials that can match the electrochemical properties that the current LIB holds are being considered²². The cell relies on the shuttle of ions and electrons back and forth between the electrodes which means that the cathode materials need to have good electronic and ionic conductivity to ensure an acceptable rate performance³⁴.

Co is today commonly used as the metal in the metaloxide at the cathode due to its ease of synthesis, stability in air and high operation voltage⁵. It is however toxic and disputed due to a negative environmental impact and human rights violation in

conjunction with mining. Today's battery research are heading towards cobalt free cathodes⁵⁰. It is suggested to use other transition metals such as Ni, Mn and Fe which could lead to lower cost and possibly higher energy contents⁴³. The Co-free cathodes LiFePO_4 (LFP) and LiMn_2O_4 (LMO) are researched and they show good qualities when it comes to thermal stability and cost, but LFP shows poor electrical conductivity at lower temperatures, and LMO has structural challenges due to Jahn Teller distortion on Mn^{3+} which leads to significant volumetric changes²⁸. These challenges has a negative effect on the cycle life of the battery²². More promising alternatives to Co-free cathodes are lithium-rich oxides, nickel-rich layered oxides and lithium nickel manganese oxide (LNMO, spinel)⁵⁰. These materials are less expensive and more abundant than the cathodes containing Co⁵⁰. They have also shown good performance (Table 2), but with some critical disadvantages. The Li-rich cathode has problems with phase transition and transition metal dissolution, while the Ni-rich cathode struggles with structural degradation and safety issues⁵⁰. The LNMO (spinel) cathode material suffers from a side reaction at high voltage and/or temperature between LNMO and the electrolyte⁵⁰.

Table 2: Approximate values for specific energy and specific capacity for Li-rich oxide, Ni-rich layered oxide and LNMO (spinel)⁵⁰

Cathode	Specific Capacity mAh g^{-1}	Specific Energy Wh kg^{-1}
Li-rich	250	900
Ni-rich	240	800
LNMO (spinel)	145	625

3.1.2 Anode materials

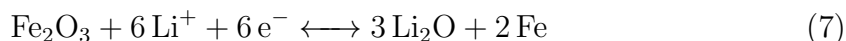
For the battery to be sustainable the anode materials must be abundant, non-toxic, have high energy density and cycling stability and also be cost-effective³⁶. The materials must therefore be able to store large amounts of ions without changing the structure and also undergo many cycles without significant performance loss³⁶. In an electrochemical point of view it should have high electrical and ionic conductivity³⁶.

Today graphite is the standard anode of the LIBs²⁹. The graphite particles are spherically shaped to achieve high packing density¹⁶. When Li ions are intercalated into the graphite structure, there is one Li ion surrounded by 6 carbon atoms (figure 2)¹⁶. This leads to good cycle life since the Li ions don't increase the volume of the structure¹⁶. Graphite can be divided into natural and synthetic graphite where synthetic graphite is more expensive because of the energy consuming production of it³⁴. Synthetic graphite is also sensitive to the solvents used to dissolve the lithium-

salts in the electrolyte and it has a relatively low specific capacity⁴⁶. There is an ongoing trend to substitute synthetic with natural graphite since synthetic graphite often is made from petroleum-based precursors which give rise to an interest in producing graphite from biomass and industrial waste instead⁴⁶. Although, natural graphite originate from ores which is geographically distributed and is actually on the 2023 list of critical materials for the EU⁶.

Silicon (Si) as a Li-alloy anode material in the lithium ion batteries is a promising alternative¹². It is the second most abundant material in the Earth crust (27.7%) and has a specific capacity almost 10 times higher (3579 mA h g⁻¹) than that of graphite which could increase the energy density of the LIB¹². With silicon, the Li ions alloy directly with silicon instead of using a host structure to store them¹². The main issue with Si as anode material is the significant volume change during intercalation and deintercalation of lithium ions¹². Volume changes lead to a shorter life of the battery due to fractures and deformation at the Si anode and destruction of the protective SEI²³. There has been a lot of research on how to implement Si as a possible anode material for lithium ion batteries, for example processing of Si to nano particles²³. The problem of processing Si is the high cost and energy consumption²³.

Different metal alloys and transition metal oxides such as SnO₂ and Fe₂O₃ have also been suggested as potential anode materials for the LIBs²⁰. The metal oxides can reversely store more than one Li-ion per metal atom which leads to higher capacity²⁰. Fe is more abundant than Sn and would therefore be a more sustainable option⁴⁸. Lithium is stored in the Fe₂O₃ as described in the following electrochemical reaction⁴⁸:



Again, the cycle life of the battery will be reduced due to severe volume changes in the Fe-oxide structure during charge/discharge process. Creating a nano structure of the anode material is yet again suggested as a solution to increase the contact area between the electrode and the electrolyte which will increase the charge/discharge rate and give a higher specific capacity⁴⁸.

3.1.3 Electrolytes and Separators

Today's electrolyte in conventional LIBs, LiPF₆, possesses high solubility, ionic mobility, conductivity, and electrochemical stability and acts as a primary option in

current LIBs³. Yet, it has naturally poor chemical and thermal stability and pose sustainability concerns³. The current challenge for the electrolyte is the potential hazard risk due to thermal instability and it's volatile and flammable nature, as well as poor recyclability³.

Ionic liquids (ILs), being liquids consisting of salts with low melting points, could replace the present carbonate solutions since they possess low flammability, a broad electrochemical window, high thermal stability and low volatility³⁵. They are stable up to 300-400 °C compared to 55°C which is the stable temperature of today's LIB³⁵. Batteries with IL electrolyte will also be a safer option since it consists of no volatile components and is therefore not flammable¹⁹. The ILs have also shown to effectively form the protective SEI layer on the anode surface, but due to higher viscosity, potentially lower conductivity and insecurities around Li⁺ transport numbers, they have not become a common choice of electrolyte yet¹⁹.

3.1.4 Solid State Electrolytes

Solid-state-electrolytes (SSE) can serve as both electrolyte and separator⁹. This provides possibilities to circumvent the safety issues of the present flammable liquid electrolytes in the LIBs⁹. Efficient and sustainable SSEs need to have high ionic conductivity, small grain boundry and be chemically stable and environmentally benign⁹. Oxides and sulfides have been researched as potential materials in SSEs, as well as hydride -and halide materials which have high ionic conductivity⁹.

For the ions to be able to travel through the SSE, they need to overcome the interactions in the lattice which means that the conductivity depends on the crystalline structure of the electrolyte⁹. The ions travel through the vacancies in the SSE that depends on the Schottky defects in the material⁹.

SSEs can be both polymeric and ceramic and the biggest interest is around polymeric solid electrolytes, since ionic polymers can be produced from renewable resources and possibly replace those synthesised from fossil fuels⁹. However, there are some concerns about the amount of energy required to produce monomers from renewable resources⁹. Various classes of ceramic solid electrolytes have been developed such as Li and Na super ionic conductor (LISICON-type and NASICON-type), garnet-type, perovskite-type, sulfide electrolyte and oxy-nitride electrolyte⁹. Their current challenges are poor compatibility with the Li anode, poor mechanical property and poor interfacial stability⁹. Production of all-oxide ceramic electrolytes is expensive and that makes it challenging to have a large-scale production of them⁹. SSEs are also facing some challenges such as lower contact between the electrolyte and the

electrodes, which is crucial since this is where the charge transfer reactions occur⁴⁰.

The separator needs to be electrochemically and chemically stable to prevent degradation and to ensure a long life⁷. The absorption of electrolyte is also a crucial requirement since that is essential for ion transport⁷. It is also important to find a material with the right thickness and porosity⁷. Polyethylene and polypropylene are dominating separators in commercial LIBs²⁹. Cellulose-based separators are of great interest since it is a biopolymer and naturally abundant³. Separators made of chitin, alginate and silk fibroin biomaterials are promising choices for the advancement of future sustainable LIBs³.

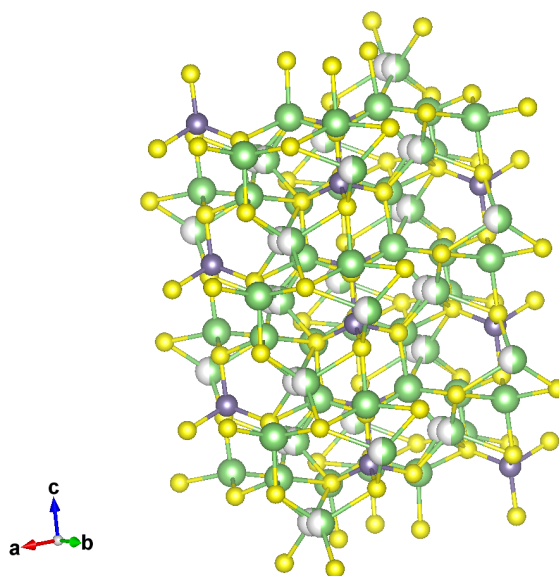


Figure 4: Structure of LISICON-type SSE (GeLi_4S_4) with structural parameters from Momma et al.²⁷

3.2 New chemistries to replace the LIB

3.2.1 Cathode materials

Sodium is one of the elements that has shown potential to replace lithium as cathode material³². The Earth's crust holds more than 1000 times more sodium than lithium³¹. Sodium is however larger than lithium and will therefore not have the same volumetric capacity, which is critical for a battery being used in portable devices³². Lithium-ions occupy space between the graphite layers when the battery is charged without significantly changing the structure. Sodium, on the other hand, is too large to leave the structure unchanged after intercalation⁴. In other words, new anode materials must be researched for the sodium-ion battery (SIB) to be a

good alternative to the LIB when it comes to capacity. There is a lot of research on the field of SIBs and $\text{Na}_3\text{V}_2(\text{PO}_4)_3$ has been tried out as a cathode material²², but it has shown some poor conductivity properties due to its large band gap of 1.735 eV²⁴. Other suggestions to cathode materials have been researched, but there has been challenges when it comes to specific capacity, cycle life and specific power⁴. Elemental doping is used to overcome these challenges and to improve the kinetics of the materials used at the cathode SIBs²⁴.

Another option is calcium (Ca), which is even more abundant than sodium³⁹. Looking at the favourable aspects, Ca will move two charges for each ion, instead of one which lithium and sodium carry³⁹. It also has an atomic radius quite similar to the sodium ion and the most negative redox potential of the multivalent elements³⁹. Because of Ca's intermediate atomic weight and density it shows some potential when it comes to both gravimetric and volumetric capacities³⁹. The problem with Ca is that it will change the chemistry in the battery³⁹. It is larger, heavier and slower than lithium and sodium and one of the greatest challenges is therefore to move Ca-ions back and forth between the cathode and anode. To solve the problems related to kinetics and efficiency, different alloys with calcium has been researched to find suitable electrode materials and electrolytes³⁹. Ca^{2+} have for example been tried intercalated into layered oxides such as V_2O_5 and MoO_3 ³⁹

Table 3: Review of ionic radius³⁹, metal/ion potentials vs. SHE² and abundance on the earth crust in mg kg^{-1} ³⁹ for Li, Na and Ca.

Element	Ionic Radius [pm]	Metal/ion redox potential vs. SHE [V]	Abundance on the earth crust [mg kg^{-1}]
Li	90	-3.04	20
Na	116	-2.71	23600
Ca	114	-2.87	41500

3.2.2 Anode materials

For sodium to replace lithium in the batteries, the cell is dependent on some other anode material that does not undergo structural change after intercalation of ions, has a high reversible capacity and have a low redox potential compared to the cathode redox potential¹⁶. Different metal oxides have also been researched as anodes for sodium ion batteries¹⁶. Some metal oxides, such as Ti-based oxides, can store Na^+ -ions by intercalation chemistry, but due to few storage sites in the Ti-oxide crystalline structure, the reversible capacity has not given satisfactory results¹⁶. Other metal oxides can store Na^+ -ions by conversion chemistry¹⁶. Sodium ions will then

react with the anode to form a new compound during charging, and decompose back to sodium ions during discharging, i.e. conversion reactions changes the structure of the electrode materials¹⁶. Octahedral SnO₂ nanocrystals have been researched as anode material with sodiation and desodiation and gave a satisfactory results for both reversible capacity and cycle performance¹⁶.

Carbon-based anode materials such as graphite is not yet off the table for SIBs. Both hard carbon (HC) and graphite has shown some promising qualities for large-scale application in SIBs¹⁶. HC is also cheap and abundant⁸. Although, the binding energy between Na and the graphite layers will be higher due to sodium's larger ion radius¹⁶. Lithium, on the other hand, will be stabilized by van der Waals forces when intercalated between the graphite layers⁸. The van der Waals forces in the sodium-intercalated graphite seems to be more or less absent, due to the structural deformation energy¹⁶.

3.3 Recycling

The battery production is, as known, growing rapidly and the demand for the materials used in the LIB with it⁴⁷. To have recycling in mind when manufacturing batteries is more important than ever. Since the LIB consists of non-abundant materials, it is crucial that they can be used more than once. For circular economy to be feasible and profitable, the recycling process must be of lower cost than the manufacturing process, something it is not today⁵¹. The LIB has a complex structure and consists of everything from plastic to heavy metals and is therefore challenging to fully recycle¹⁷. Ideally, the materials used would be separated and chemically pure after the recycling process¹⁷. Recycling is also important because the waste could otherwise pose a large risk to human health and to the environment¹⁷.

3.3.1 Recycling cathode materials

The cathode of the LIB holds the most valuable materials and is therefore of high interest to be fully recycled. The recycling of the cathode materials is the biggest challenge and takes the largest portion by both cost and mass of any spent LIB²⁵.

Hydrometallurgy is used to recycle the cathode materials in the LIBs today and focus on recovery of Co and Li contained in the cathodes²⁵. Hydrochloric acid (HCl), nitric acid (HNO₃), hydrogen peroxide (H₂O₂) and sulfuric acid (H₂SO₄) are used in the hydrometallurgy process of extracting and separating the cathode materials¹⁴. It is a four-part process including leaching, impurity removal, transition metal re-

covery and Li recovery¹⁴. The recovery processes is carried out with chemical precipitation or solvent extraction¹⁴. The methods are up to this day non-profitable. It is a high-cost process due to transportation, dissembling and important factors such as product yield, water balance and reagent consumption has to be further investigated¹⁴. Direct recycling is proposed as an alternative³⁷. It will then be possible to enable critical metals recovery without dismantling the battery package³⁷. This has shown to be profitable for low Co containing materials such as Ni, Mn/Fe based cathodes³⁷. Each direct recycling process is targeted for specific sets of electrode/electrolyte chemistries³⁷. In a process like this, pre-treatment involves spent LIBs being dismantled and their subcomponents, including anode and cathode materials, are exfoliated using mechanical or polymetallurgical methods³⁷. Direct recycling is more cost-effective and less energy-consuming than hydrometallurgy, but can be challenging because of lack of chemical information or safety classification³⁷.

3.3.2 Recycling anode materials

There is also a growing interest around the ability to recycle the spent anode materials due to the projected waste-amounts from end-of-life LIBs²¹. The graphite waste will become a threat to human health and to the environment²¹. Recycling of graphitic anode components makes up 15% of the costs²¹. Spent graphite anodes can contain Li deposits lost from the cathode and interfacial growth which makes the recycling process a bit more complicated, but graphite is still way easier to recycle than the cathode materials since it doesn't undergo structural changes and degradation at its end of life²¹. Acid treatment is used to extract Li and the extracted Li is later precipitated²¹. This is a precursor for new battery material synthesis²¹. Graphite is finally regenerated using thermal annealing²¹. The recycled graphite can be used in new LIBs or be used to prepare high-performance graphene²¹.

3.3.3 Recycling electrolyte

The electrolyte is the next largest component by cost and mass of any spent LIB¹⁸. They contain toxic organic solvents and LiPF_6 which is expensive¹⁸. The electrolytes can be recovered by distillation where the electrolyte is heated and vaporised¹⁸. The vapour can be condensed and used again¹⁸. The distillation process leaves the lithium salt behind and gives high-purity results¹⁸. It is although quite energy-consuming compared to solvent extraction where suitable solvents are used to selectively dissolve the organic solvents¹⁸. This process requires less energy, but more accuracy due to the careful choice of solvent¹⁸.

4 Conclusion and future perspectives

It will be hard to replace the LIB for some applications because of its leading energy capacity, but the critical limitation in the amount of materials used in this battery forces us to look in another direction. The LIB is developing towards Co-free cathodes and anodes consisting of more abundant materials. Solid state batteries and new chemistries such as SIBs and multivalent ion batteries are growing fields of research and could result in more cost-effective and sustainable BESSs. The recycling process of LIBs is developing to create a circular economy, but has not become profitable yet.

Something clearly needs to be done if we are going to reach the UNs goals on climate action and responsible consumption and production. I believe that battery energy storage systems are the future and that the ongoing research on them is heading in the right direction. My concern is how the global economy controls what is currently beneficial, such as the exploitation of cheap labour to mine critical elements to the production of LIBs. If we become more conscious and change our mindset from short term economy to sustainability, we might save our planet after all.

References

- [1] Lea Berrang-Ford, James D. Ford and Jaclyn Paterson. ‘Are we adapting to climate change?’ In: *Global Environmental Change* 21.1 (1st Feb. 2011), pp. 25–33. ISSN: 0959-3780. DOI: 10.1016/j.gloenvcha.2010.09.012. URL: <https://www.sciencedirect.com/science/article/pii/S0959378010000968> (visited on 14th Mar. 2023).
- [2] Allan G. Blackman and Lawrence R. Gahan. *Aylward and Findlay’s SI chemical data*. 7th edition. OCLC: 842856344. Milton, Qld: John Wiley & Sons Australia, 2014. 185 pp. ISBN: 978-0-7303-0246-9.
- [3] Heather Cavers et al. ‘Perspectives on Improving the Safety and Sustainability of High Voltage Lithium-Ion Batteries Through the Electrolyte and Separator Region’. In: *Advanced Energy Materials* 12.23 (2022). _eprint: <https://onlinelibrary.wiley.com/doi/pdf/10.1002/aenm.202200147>, p. 2200147. ISSN: 1614-6840. DOI: 10.1002/aenm.202200147. URL: <https://onlinelibrary.wiley.com/doi/abs/10.1002/aenm.202200147> (visited on 25th Apr. 2023).
- [4] Kudakwashe Chayambuka et al. ‘From Li-Ion Batteries toward Na-Ion Chemistries: Challenges and Opportunities’. In: *Advanced Energy Materials* 10.38 (2020). _eprint: <https://onlinelibrary.wiley.com/doi/pdf/10.1002/aenm.202001310>, p. 2001310. ISSN: 1614-6840. DOI: 10.1002/aenm.202001310. URL: <https://onlinelibrary.wiley.com/doi/abs/10.1002/aenm.202001310> (visited on 8th Mar. 2023).
- [5] *Co-Free Layered Cathode Materials for High Energy Density Lithium-Ion Batteries* | *ACS Energy Letters*. URL: <https://pubs.acs.org/doi/pdf/10.1021/acsenenergylett.0c00742> (visited on 21st Apr. 2023).
- [6] *Critical raw materials*. URL: https://single-market-economy.ec.europa.eu/sectors/raw-materials/areas-specific-interest/critical-raw-materials_en (visited on 26th Apr. 2023).
- [7] Valadoula Deimede and Costas Elmasides. ‘Separators for Lithium-Ion Batteries: A Review on the Production Processes and Recent Developments’. In: *Energy Technology* 3.5 (2015). _eprint: <https://onlinelibrary.wiley.com/doi/pdf/10.1002/ente.201402215>, pp. 453–468. ISSN: 2194-4296. DOI: 10.1002/ente.201402215. URL: <https://onlinelibrary.wiley.com/doi/abs/10.1002/ente.201402215> (visited on 25th Apr. 2023).
- [8] Xinwei Dou et al. ‘Hard carbons for sodium-ion batteries: Structure, analysis, sustainability, and electrochemistry’. In: *Materials Today* 23 (1st Mar. 2019), pp. 87–104. ISSN: 1369-7021. DOI: 10.1016/j.mattod.2018.12.040. URL: <https://doi.org/10.1016/j.mattod.2018.12.040>

- [//www.sciencedirect.com/science/article/pii/S1369702118310873](https://www.sciencedirect.com/science/article/pii/S1369702118310873) (visited on 27th Apr. 2023).
- [9] Zhonghui Gao et al. ‘Promises, Challenges, and Recent Progress of Inorganic Solid-State Electrolytes for All-Solid-State Lithium Batteries’. In: *Advanced Materials* 30.17 (2018). _eprint: <https://onlinelibrary.wiley.com/doi/pdf/10.1002/adma.2017051705> p. 1705702. ISSN: 1521-4095. DOI: 10.1002/adma.201705702. URL: <https://onlinelibrary.wiley.com/doi/abs/10.1002/adma.201705702> (visited on 24th Apr. 2023).
- [10] John B. Goodenough and Kyu-Sung Park. ‘The Li-Ion Rechargeable Battery: A Perspective’. In: *Journal of the American Chemical Society* 135.4 (30th Jan. 2013), pp. 1167–1176. ISSN: 0002-7863, 1520-5126. DOI: 10.1021/ja3091438. URL: <https://pubs.acs.org/doi/10.1021/ja3091438> (visited on 26th Apr. 2023).
- [11] M. A. Hannan et al. ‘Battery energy-storage system: A review of technologies, optimization objectives, constraints, approaches, and outstanding issues’. In: *Journal of Energy Storage* 42 (1st Oct. 2021), p. 103023. ISSN: 2352-152X. DOI: 10.1016/j.est.2021.103023. URL: <https://www.sciencedirect.com/science/article/pii/S2352152X21007349> (visited on 14th Apr. 2023).
- [12] Sheng Gong He et al. ‘Considering Critical Factors of Silicon/Graphite Anode Materials for Practical High-Energy Lithium-Ion Battery Applications’. In: *Energy & Fuels* 35.2 (21st Jan. 2021). Publisher: American Chemical Society, pp. 944–964. ISSN: 0887-0624. DOI: 10.1021/acs.energyfuels.0c02948. URL: <https://doi.org/10.1021/acs.energyfuels.0c02948> (visited on 24th Apr. 2023).
- [13] Tatsuo Horiba. ‘Lithium-Ion Battery Systems’. In: *Proceedings of the IEEE* 102.6 (June 2014). Conference Name: Proceedings of the IEEE, pp. 939–950. ISSN: 1558-2256. DOI: 10.1109/JPROC.2014.2319832.
- [14] Joey Chung-Yen Jung, Pang-Chieh Sui and Jiujun Zhang. ‘A review of recycling spent lithium-ion battery cathode materials using hydrometallurgical treatments’. In: *Journal of Energy Storage* 35 (1st Mar. 2021), p. 102217. ISSN: 2352-152X. DOI: 10.1016/j.est.2020.102217. URL: <https://www.sciencedirect.com/science/article/pii/S2352152X20320405> (visited on 25th Apr. 2023).
- [15] Taehoon Kim et al. ‘Lithium-ion batteries: outlook on present, future, and hybridized technologies’. In: *Journal of Materials Chemistry A* 7.7 (12th Feb. 2019). Publisher: The Royal Society of Chemistry, pp. 2942–2964. ISSN: 2050-7496. DOI: 10.1039/C8TA10513H. URL: <https://pubs.rsc.org/en/content/articlelanding/2019/ta/c8ta10513h> (visited on 14th Mar. 2023).

- [16] Youngjin Kim et al. ‘High-Capacity Anode Materials for Sodium-Ion Batteries’. In: *Chemistry – A European Journal* 20.38 (2014). _eprint: <https://onlinelibrary.wiley.com>, pp. 11980–11992. ISSN: 1521-3765. DOI: 10.1002/chem.201402511. URL: <https://onlinelibrary.wiley.com/doi/abs/10.1002/chem.201402511> (visited on 21st Apr. 2023).
- [17] Dana L. Thompson et al. ‘The importance of design in lithium ion battery recycling – a critical review’. In: *Green Chemistry* 22.22 (2020). Publisher: Royal Society of Chemistry, pp. 7585–7603. DOI: 10.1039/D0GC02745F. URL: <https://pubs.rsc.org/en/content/articlelanding/2020/gc/d0gc02745f> (visited on 24th Apr. 2023).
- [18] Shuya Lei, Wei Sun and Yue Yang. ‘Solvent extraction for recycling of spent lithium-ion batteries’. In: *Journal of Hazardous Materials* 424 (15th Feb. 2022), p. 127654. ISSN: 0304-3894. DOI: 10.1016/j.jhazmat.2021.127654. URL: <https://www.sciencedirect.com/science/article/pii/S0304389421026224> (visited on 25th Apr. 2023).
- [19] Andrzej Lewandowski and Agnieszka Świdorska-Mocek. ‘Ionic liquids as electrolytes for Li-ion batteries—An overview of electrochemical studies’. In: *Journal of Power Sources* 194.2 (1st Dec. 2009), pp. 601–609. ISSN: 0378-7753. DOI: 10.1016/j.jpowsour.2009.06.089. URL: <https://www.sciencedirect.com/science/article/pii/S0378775309011616> (visited on 25th Apr. 2023).
- [20] Chu Liang et al. ‘Lithium alloys and metal oxides as high-capacity anode materials for lithium-ion batteries’. In: *Journal of Alloys and Compounds* 575 (25th Oct. 2013), pp. 246–256. ISSN: 0925-8388. DOI: 10.1016/j.jallcom.2013.04.001. URL: <https://www.sciencedirect.com/science/article/pii/S0925838813008657> (visited on 24th Apr. 2023).
- [21] Kui Liu et al. ‘From spent graphite to recycle graphite anode for high-performance lithium ion batteries and sodium ion batteries’. In: *Electrochimica Acta* 356 (1st Oct. 2020), p. 136856. ISSN: 0013-4686. DOI: 10.1016/j.electacta.2020.136856. URL: <https://www.sciencedirect.com/science/article/pii/S0013468620312494> (visited on 25th Apr. 2023).
- [22] Tiefeng Liu et al. ‘Sustainability-inspired cell design for a fully recyclable sodium ion battery’. In: *Nature Communications* 10.1 (29th Apr. 2019), p. 1965. ISSN: 2041-1723. DOI: 10.1038/s41467-019-09933-0. URL: <https://www.nature.com/articles/s41467-019-09933-0> (visited on 2nd Mar. 2023).
- [23] Fei Luo et al. ‘Review—Nano-Silicon/Carbon Composite Anode Materials Towards Practical Application for Next Generation Li-Ion Batteries’. In: *Journal of The Electrochemical Society* 162.14 (9th Oct. 2015). Publisher: IOP Pub-

- lishing, A2509. ISSN: 1945-7111. DOI: 10.1149/2.0131514jes. URL: <https://iopscience.iop.org/article/10.1149/2.0131514jes/meta> (visited on 24th Apr. 2023).
- [24] Wei-Jun Lv et al. ‘Strategies to Build High-Rate Cathode Materials for Na-Ion Batteries’. In: *ChemNanoMat* 5.10 (2019). _eprint: <https://onlinelibrary.wiley.com/doi/pdf/10.1002/cnma.201900254> pp. 1253–1262. ISSN: 2199-692X. DOI: 10.1002/cnma.201900254. URL: <https://onlinelibrary.wiley.com/doi/abs/10.1002/cnma.201900254> (visited on 21st Apr. 2023).
- [25] Brian Makuza et al. ‘Pyrometallurgical options for recycling spent lithium-ion batteries: A comprehensive review’. In: *Journal of Power Sources* 491 (15th Apr. 2021), p. 229622. ISSN: 0378-7753. DOI: 10.1016/j.jpowsour.2021.229622. URL: <https://www.sciencedirect.com/science/article/pii/S0378775321001671> (visited on 24th Apr. 2023).
- [26] Tom K. R. Matthews, Robert L. Wilby and Conor Murphy. ‘Communicating the deadly consequences of global warming for human heat stress’. In: *Proceedings of the National Academy of Sciences* 114.15 (11th Apr. 2017). Publisher: Proceedings of the National Academy of Sciences, pp. 3861–3866. DOI: 10.1073/pnas.1617526114. URL: <https://www.pnas.org/doi/abs/10.1073/pnas.1617526114> (visited on 14th Apr. 2023).
- [27] K. Momma and F. Izumi. ‘VESTA 3 for three-dimensional visualization of crystal, volumetric and morphology data’. In: *Journal of Applied Crystallography* 44.6 (1st Dec. 2011). Publisher: International Union of Crystallography, pp. 1272–1276. ISSN: 0021-8898. DOI: 10.1107/S0021889811038970. URL: <https://journals.iucr.org/j/issues/2011/06/00/db5098/> (visited on 26th Apr. 2023).
- [28] Angelo Mullaliu et al. ‘Lattice Compensation to Jahn–Teller Distortion in Na-Rich Manganese Hexacyanoferrate for Li-Ion Storage: An Operando Study’. In: *ACS Applied Energy Materials* 3.6 (22nd June 2020). Publisher: American Chemical Society, pp. 5728–5733. DOI: 10.1021/acsaem.0c00669. URL: <https://doi.org/10.1021/acsaem.0c00669> (visited on 19th Apr. 2023).
- [29] Keith B. Oldham, Jan C. Myland and Alan M. Bond. *Electrochemical Science and Technology: Fundamentals and Applications*. Wiley, 2012. ISBN: 978-1-119-96599-2.
- [30] Jędrzej Piątek et al. ‘Sustainable Li-Ion Batteries: Chemistry and Recycling’. In: *Advanced Energy Materials* 11.43 (2021). _eprint: <https://onlinelibrary.wiley.com/doi/pdf/10.1002/aenm.202003456> p. 2003456. ISSN: 1614-6840. DOI: 10.1002/aenm.202003456. URL: <https://onlinelibrary.wiley.com/doi/abs/10.1002/aenm.202003456> (visited on 12th Mar. 2023).

- [31] *Pursuing better, cheaper and more environmentally friendly batteries - English articles*. URL: <https://www.titan.uio.no/english/2021/pursuing-better-cheaper-and-more-environmentally-friendly-batteries.html> (visited on 17th Apr. 2023).
- [32] M. Saiful Islam and Craig A. J. Fisher. ‘Lithium and sodium battery cathode materials: computational insights into voltage, diffusion and nanostructural properties’. In: *Chemical Society Reviews* 43.1 (2014). Publisher: Royal Society of Chemistry, pp. 185–204. DOI: 10.1039/C3CS60199D. URL: <https://pubs.rsc.org/en/content/articlelanding/2014/cs/c3cs60199d> (visited on 17th Apr. 2023).
- [33] Richard Schmalensee, Thomas M. Stoker and Ruth A. Judson. ‘World Carbon Dioxide Emissions: 1950–2050’. In: *The Review of Economics and Statistics* 80.1 (1st Feb. 1998), pp. 15–27. ISSN: 0034-6535. DOI: 10.1162/003465398557294. URL: <https://doi.org/10.1162/003465398557294> (visited on 14th Apr. 2023).
- [34] Bruno Scrosati and Jürgen Garche. ‘Lithium batteries: Status, prospects and future’. In: *Journal of Power Sources* 195.9 (1st May 2010), pp. 2419–2430. ISSN: 0378-7753. DOI: 10.1016/j.jpowsour.2009.11.048. URL: <https://www.sciencedirect.com/science/article/pii/S0378775309020564> (visited on 20th Apr. 2023).
- [35] Bruno Scrosati, Jusef Hassoun and Yang-Kook Sun. ‘Lithium-ion batteries. A look into the future’. In: *Energy & Environmental Science* 4.9 (2011). Publisher: Royal Society of Chemistry, pp. 3287–3295. DOI: 10.1039/C1EE01388B. URL: <https://pubs.rsc.org/en/content/articlelanding/2011/ee/c1ee01388b> (visited on 22nd Apr. 2023).
- [36] Xin Shen et al. ‘Beyond lithium ion batteries: Higher energy density battery systems based on lithium metal anodes’. In: *Energy Storage Materials* 12 (1st May 2018), pp. 161–175. ISSN: 2405-8297. DOI: 10.1016/j.ensm.2017.12.002. URL: <https://www.sciencedirect.com/science/article/pii/S2405829717306086> (visited on 14th Mar. 2023).
- [37] Steve Sloop et al. ‘A direct recycling case study from a lithium-ion battery recall’. In: *Sustainable Materials and Technologies* 25 (1st Sept. 2020), e00152. ISSN: 2214-9937. DOI: 10.1016/j.susmat.2020.e00152. URL: <https://www.sciencedirect.com/science/article/pii/S2214993718300599> (visited on 25th Apr. 2023).
- [38] Darlene Steward, Ahmad Mayyas and Margaret Mann. ‘Economics and Challenges of Li-Ion Battery Recycling from End-of-Life Vehicles’. In: *Procedia Manufacturing*. Sustainable Manufacturing for Global Circular Economy: Proceedings of the 16th Global Conference on Sustainable Manufacturing 33 (1st Jan. 2019), pp. 272–279. ISSN: 2351-9789. DOI: 10.1016/j.promfg.2019.04.

033. URL: <https://www.sciencedirect.com/science/article/pii/S2351978919305104> (visited on 18th Apr. 2023).
- [39] Lorenzo Stievano et al. ‘Emerging calcium batteries’. In: *Journal of Power Sources* 482 (15th Jan. 2021), p. 228875. ISSN: 0378-7753. DOI: 10.1016/j.jpowsour.2020.228875. URL: <https://www.sciencedirect.com/science/article/pii/S0378775320311794> (visited on 17th Apr. 2023).
- [40] Laisuo Su et al. ‘Tailoring Electrode–Electrolyte Interfaces in Lithium-Ion Batteries Using Molecularly Engineered Functional Polymers’. In: *ACS Applied Materials & Interfaces* 13.8 (3rd Mar. 2021). Publisher: American Chemical Society, pp. 9919–9931. ISSN: 1944-8244. DOI: 10.1021/acsami.0c20978. URL: <https://doi.org/10.1021/acsami.0c20978> (visited on 24th Apr. 2023).
- [41] J.-M. Tarascon and M. Armand. ‘Issues and challenges facing rechargeable lithium batteries’. In: *Nature* 414.6861 (Nov. 2001). Number: 6861 Publisher: Nature Publishing Group, pp. 359–367. ISSN: 1476-4687. DOI: 10.1038/35104644. URL: <https://www.nature.com/articles/35104644> (visited on 16th Mar. 2023).
- [42] Mike Tebyetekerwa et al. ‘Rechargeable Dual-Carbon Batteries: A Sustainable Battery Technology’. In: *Advanced Energy Materials* 12.44 (2022). _eprint: <https://onlinelibrary.wiley.com/doi/pdf/10.1002/aenm.202202450>, p. 2202450. ISSN: 1614-6840. DOI: 10.1002/aenm.202202450. URL: <https://onlinelibrary.wiley.com/doi/abs/10.1002/aenm.202202450> (visited on 14th Apr. 2023).
- [43] Natalia Voronina, Yang-Kook Sun and Seung-Taek Myung. ‘Co-Free Layered Cathode Materials for High Energy Density Lithium-Ion Batteries’. In: *ACS Energy Letters* 5.6 (12th June 2020). Publisher: American Chemical Society, pp. 1814–1824. DOI: 10.1021/acsenergylett.0c00742. URL: <https://doi.org/10.1021/acsenergylett.0c00742> (visited on 21st Apr. 2023).
- [44] Jianwu Wen, Yan Yu and Chunhua Chen. ‘A Review on Lithium-Ion Batteries Safety Issues: Existing Problems and Possible Solutions’. In: *Materials Express* 2.3 (1st Sept. 2012), pp. 197–212. DOI: 10.1166/mex.2012.1075.
- [45] Yue Yang et al. ‘On the sustainability of lithium ion battery industry – A review and perspective’. In: *Energy Storage Materials* 36 (1st Apr. 2021), pp. 186–212. ISSN: 2405-8297. DOI: 10.1016/j.ensm.2020.12.019. URL: <https://www.sciencedirect.com/science/article/pii/S2405829720304827> (visited on 8th Mar. 2023).
- [46] Masaki Yoshio, Hongyu Wang and Kenji Fukuda. ‘Spherical Carbon-Coated Natural Graphite as a Lithium-Ion Battery-Anode Material’. In: *Angewandte Chemie International Edition* 42.35 (2003). _eprint: <https://onlinelibrary.wiley.com/doi/pdf/10.1002/anie.200351203>, pp. 4203–4206. ISSN: 1521-3773. DOI: 10.1002/anie.200351203. URL: <https://onlinelibrary.wiley.com/doi/pdf/10.1002/anie.200351203>

- [//onlinelibrary.wiley.com/doi/abs/10.1002/anie.200351203](https://onlinelibrary.wiley.com/doi/abs/10.1002/anie.200351203) (visited on 24th Apr. 2023).
- [47] Xingwen Yu and Arumugam Manthiram. ‘Sustainable Battery Materials for Next-Generation Electrical Energy Storage’. In: *Advanced Energy and Sustainability Research* 2.5 (2021). _eprint: <https://onlinelibrary.wiley.com/doi/pdf/10.1002/aesr.20200102>, p. 2000102. ISSN: 2699-9412. DOI: 10.1002/aesr.202000102. URL: <https://onlinelibrary.wiley.com/doi/abs/10.1002/aesr.202000102> (visited on 12th Mar. 2023).
- [48] Lei Zhang, Hao Bin Wu and Xiong Wen (David) Lou. ‘Iron-Oxide-Based Advanced Anode Materials for Lithium-Ion Batteries’. In: *Advanced Energy Materials* 4.4 (2014). _eprint: <https://onlinelibrary.wiley.com/doi/pdf/10.1002/aenm.201300958>, p. 1300958. ISSN: 1614-6840. DOI: 10.1002/aenm.201300958. URL: <https://onlinelibrary.wiley.com/doi/abs/10.1002/aenm.201300958> (visited on 24th Apr. 2023).
- [49] Yaguang Zhang, Ning Du and Deren Yang. ‘Designing superior solid electrolyte interfaces on silicon anodes for high-performance lithium-ion batteries’. In: *Nanoscale* 11.41 (25th Oct. 2019). Publisher: The Royal Society of Chemistry, pp. 19086–19104. ISSN: 2040-3372. DOI: 10.1039/C9NR05748J. URL: <https://pubs.rsc.org/en/content/articlelanding/2019/nr/c9nr05748j> (visited on 26th Apr. 2023).
- [50] Hong Zhao et al. ‘Cobalt-Free Cathode Materials: Families and their Prospects’. In: *Advanced Energy Materials* 12.16 (2022). _eprint: <https://onlinelibrary.wiley.com/doi/pdf/10.1002/aenm.202103894>, p. 2103894. ISSN: 1614-6840. DOI: 10.1002/aenm.202103894. URL: <https://onlinelibrary.wiley.com/doi/abs/10.1002/aenm.202103894> (visited on 19th Apr. 2023).
- [51] Haiyang Zou et al. ‘A novel method to recycle mixed cathode materials for lithium ion batteries’. In: *Green Chemistry* 15.5 (2013). Publisher: Royal Society of Chemistry, pp. 1183–1191. DOI: 10.1039/C3GC40182K. URL: <https://pubs.rsc.org/en/content/articlelanding/2013/gc/c3gc40182k> (visited on 24th Apr. 2023).
- [52] Ghassan Zubi et al. ‘The lithium-ion battery: State of the art and future perspectives’. In: *Renewable and Sustainable Energy Reviews* 89 (1st June 2018), pp. 292–308. ISSN: 1364-0321. DOI: 10.1016/j.rser.2018.03.002. URL: <https://www.sciencedirect.com/science/article/pii/S1364032118300728> (visited on 18th Apr. 2023).

*[heading=bibintoc,title=Bibliography]

Appendix

A Parameters for LiCoO₂

The lattice parameters used to compute the structure of LiCoO₂ in figure 1 referenced from Momma et al.²⁷ is shown in table 4.

Table 4: Unit-cell volume [\AA^3], axial relationship [\AA] and interaxial angles for LiCoO₂ referenced from Momma et al.²⁷

Lattice parameters	LiCoO ₂
Unit-cell volume [\AA^3]	96.337
Axial relationship [\AA]	a = b = 2.814, c = 14.048
Interaxial angles	$\alpha = \beta = 90^\circ$, $\gamma = 120^\circ$

The structural parameters used to compute the structure of LiCoO₂ in figure 1 referenced from Momma et al.²⁷ is shown in table 5.

Table 5: Occupancy value (g) and fractional coordinates for the atoms in LiCoO₂ referenced from Momma et al.²⁷

Atom	g	x	y	z
O	1	0.000	0.000	0.2268
Co	1	0.000	0.000	0.000
Li	1	0.000	0.000	0.500

B Parameters for LiC₆

The lattice parameters used to compute the structure of LiC₆ in figure 2 referenced from Momma et al.²⁷ is shown in table 6.

Table 6: Unit-cell volume [\AA^3], axial relationship [\AA] and interaxial angles for LiC₆ referenced from Momma et al.²⁷

Lattice parameters	LiC ₆
Unit-cell volume [\AA^3]	58.6975
Axial relationship [\AA]	a = b = 4.28, c = 3.70
Interaxial angles	$\alpha = \beta = 90^\circ$, $\gamma = 120^\circ$

The structural parameters used to compute the structure of LiC₆ in figure 2 referenced from Momma et al.²⁷ is shown in table 7.

Table 7: Occupancy value (g) and fractional coordinates for the atoms in LiC₆ referenced from Momma et al.²⁷

Atom	g	x	y	z
Li	1	0.333	0.000	0.500
C	1	0.000	0.000	0.000

C Parameters for GeLi₄S₄

The lattice parameters used to compute the structure of GeLi₄S₄ in figure 4 referenced from Momma et al.²⁷ is shown in table 8.

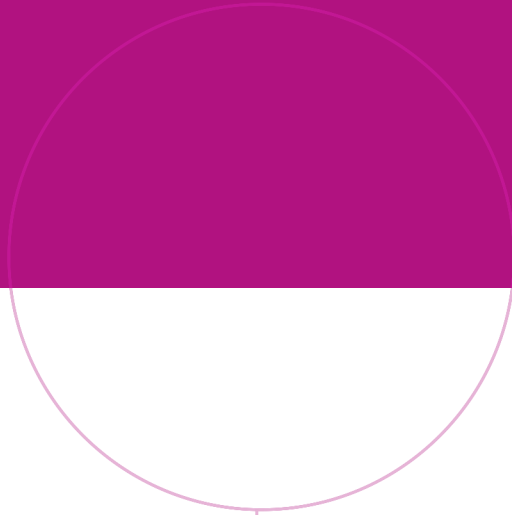
Table 8: Unit-cell volume [\AA^3], axial relationship [\AA] and interaxial angles for GeLi₄S₄ referenced from Momma et al.²⁷

Lattice parameters	GeLi ₄ S ₄
Unit-cell volume [\AA^3]	670.47
Axial relationship [\AA]	a = 14.0658, b = 7.75102, c = 6.14973
Interaxial angles	$\alpha = \beta = \gamma = 90^\circ$

The structural parameters used to compute the structure of GeLi₄S₄ in figure 4 referenced from Momma et al.²⁷ is shown in table 9.

Table 9: Occupancy value (g) and fractional coordinates for the atoms in GeLi₄S₄ referenced from Momma et al.²⁷

Atom	g	x	y	z
S1	1	0.3443	0.0135	0.7764
S2	1	0.0857	0.7500	0.7933
S3	1	0.6130	0.2500	0.7292
Ge	1	0.4103	0.2500	0.6495
Li1	1	0.1650	0.5040	0.6750
Li2	1	0.4240	0.7500	0.8290
Li3	1	0.1650	0.5040	0.6750



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