LITHIUM-ION BATTERY SYSTEMS:

A PROCESS FLOW AND SYSTEMS FRAMEWORK DESIGNED
FOR USE IN THE DEVELOPMENT OF A LIFECYCLE ENERGY
MODEL

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The Academic Faculty

by

Yukti Arora

In Partial Fulfillment
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of Master in Science in Environmental Engineering in the
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LITHIUM-ION BATTERY SYSTEMS:

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<table>
<thead>
<tr>
<th>Acronym</th>
<th>Description</th>
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<tbody>
<tr>
<td>AESC</td>
<td>Automotive Energy Supply Corporation</td>
</tr>
<tr>
<td>CAFE</td>
<td>Corporate Average Fuel Economy</td>
</tr>
<tr>
<td>CD</td>
<td>Charge Depleting</td>
</tr>
<tr>
<td>CO</td>
<td>Cobalt</td>
</tr>
<tr>
<td>COCl₂</td>
<td>Cobalt Chloride</td>
</tr>
<tr>
<td>CO₂</td>
<td>Carbon Dioxide</td>
</tr>
<tr>
<td>CS</td>
<td>Charge Sustaining</td>
</tr>
<tr>
<td>CV</td>
<td>Conventional Vehicle</td>
</tr>
<tr>
<td>DEC</td>
<td>Diethyl Carbonate</td>
</tr>
<tr>
<td>DMC</td>
<td>Dimethyl Carbonate</td>
</tr>
<tr>
<td>EC</td>
<td>Ethylene Carbonate</td>
</tr>
<tr>
<td>EMC</td>
<td>Ethyl Methyl Carbonate</td>
</tr>
<tr>
<td>EPA</td>
<td>Environmental Protection Agency</td>
</tr>
<tr>
<td>EV</td>
<td>Electric Vehicles</td>
</tr>
<tr>
<td>GIS</td>
<td>Geographic Information System</td>
</tr>
<tr>
<td>HEV</td>
<td>Hybrid Electric Vehicle</td>
</tr>
<tr>
<td>ICE</td>
<td>Internal Combustion Engine</td>
</tr>
<tr>
<td>IIG</td>
<td>Investigaciones Geologicas</td>
</tr>
<tr>
<td>LCE</td>
<td>Lithium Carbonate Equivalent</td>
</tr>
<tr>
<td>LCO (LiCoO₂)</td>
<td>Lithium Cobalt Oxide</td>
</tr>
<tr>
<td>LFP</td>
<td>Lithium Iron Phosphate</td>
</tr>
<tr>
<td>Li</td>
<td>Lithium</td>
</tr>
<tr>
<td>Li-ion</td>
<td>Lithium-ion</td>
</tr>
<tr>
<td>Abbreviation</td>
<td>Full Form</td>
</tr>
<tr>
<td>--------------</td>
<td>-----------</td>
</tr>
<tr>
<td>LiAsF₆</td>
<td>Lithium Hexafluoroarsenate</td>
</tr>
<tr>
<td>LiClO₄</td>
<td>Lithium Perchlorate</td>
</tr>
<tr>
<td>Li₂CO₃</td>
<td>Lithium Carbonate</td>
</tr>
<tr>
<td>LiNO₂</td>
<td>Lithium Nickel Oxide</td>
</tr>
<tr>
<td>LiPF₆</td>
<td>Lithium Hexafluorophosphate</td>
</tr>
<tr>
<td>Mg:Li</td>
<td>Magnesium:Lithium</td>
</tr>
<tr>
<td>Na₂CO₃</td>
<td>Sodium Carbonate</td>
</tr>
<tr>
<td>NCA</td>
<td>Nickel Cobalt Aluminum</td>
</tr>
<tr>
<td>NEC</td>
<td>Nippon Electric Company</td>
</tr>
<tr>
<td>Ni</td>
<td>Nickel</td>
</tr>
<tr>
<td>NMP</td>
<td>N-Methyl-2-Pyrrolidone</td>
</tr>
<tr>
<td>NREL</td>
<td>National Renewable Energy Laboratory</td>
</tr>
<tr>
<td>PC</td>
<td>Propylene Carbonate</td>
</tr>
<tr>
<td>PEL</td>
<td>Permissible Exposure Limit</td>
</tr>
<tr>
<td>PHEV</td>
<td>Plug-in Hybrid Electric Vehicle</td>
</tr>
<tr>
<td>PPM</td>
<td>Part Per Million</td>
</tr>
<tr>
<td>PVDF</td>
<td>Polyvinylidene Fluoride</td>
</tr>
<tr>
<td>SOC</td>
<td>State of Charge</td>
</tr>
<tr>
<td>SQM</td>
<td>Sociedad Quimica y Minera</td>
</tr>
<tr>
<td>USGS</td>
<td>U.S. Geological Survey</td>
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</tbody>
</table>
SUMMARY

The use of Lithium-ion batteries in the automotive industry has increased over the past few years, reaching 18.6% in 2013 (Sapru, 2014). The anticipated increase in demand of lithium (Li) for electric and hybrid cars entering the fleet has prompted researchers to examine the long term sustainability of lithium as a transportation resource. To provide a better understanding of future availability, this thesis presents a systems framework for the key processes and materials and energy flows involved in the complete electric vehicle lithium-ion battery lifecycle, on a global scale. This framework tracks the flow of lithium and identifies the key energy inputs and outputs, from extraction, to production, to on road use, and all the way to end of life recycling and disposal. This process flow model is the first step in developing a lifecycle energy and resource analysis model for lithium that will eventually help policymakers assess the future role of lithium battery recycling, and at what point in time establishing a recycling infrastructure becomes imminent.

Developing the systems framework in this thesis is an important step in analyzing key issues associated with lithium global supply and demand. Lithium is a critical component to batteries. However, if lithium is not recycled, a shortage of lithium is projected by 2021-2023 based on the “reserves, projected mining capacity, and forecasted demand” (Sonoc and Jeswiet, 2014). This thesis provides a systems approach and modeling framework to assess the complex relationships in the lithium supply chain. The thesis also outlines linkages to future research work, discussing how new research results can be integrated into the proposed systems framework to estimate sustainability issues.
arising from lithium battery use in electric vehicles. Outputs of these future models will help policymakers decide when lithium recycling makes environmental and economic sense.
1. INTRODUCTION

Before the introduction of Lithium ion (Li-ion) batteries, also known as 'new era' batteries, the most prominent batteries in use were lead acid and nickel cadmium. As time passed, these compositions could no longer fulfill the changing needs of the automotive industry. The latest generation of electric vehicles is more suited for Li-ion batteries because Li-ion batteries have higher energy density, are lighter, are lower maintenance, and have a longer battery life (Budde-Meiwes, et al., 2013). Alternatives, such as nickel-metal hydride and sodium nickel chloride batteries, face similar issues as lead acid and nickel cadmium batteries in terms of lower energy density, power, and performance. Furthermore, the alternative nickel batteries may also have a more significant impact on the environment, providing a disincentive for future development. On the other hand, Li-ion batteries provide a better alternative in terms of efficient energy density, costs, and environmental impact and are likely to be a forefront of new technology (Budde-Meiwes, et al., 2013). As a result lead-acid and nickel-cadmium batteries are being phased out and Li-ion batteries are capturing an increasing market share for electric vehicles.

Lithium-ion batteries are expected to become a prominent technology and dominate the battery market by 2017 (Deutsche Bank, 2009). Li-ion batteries are forecast to increase from $3.2 billion in 2013 to $24.1 billion in 2023 in light-duty consumer vehicles (Navigant Research, 2014). This increase in demand is highly dependent on the reserves and resource estimates of lithium. Even by USGS’s (U.S. Geological Survey) conservative reserve estimates of ~11 million tones as reported by Gaines and Nelson
(2009), there is only enough capacity to meet the demand until 2050 without implementing a recycling infrastructure. Therefore, it is important to not only evaluate the adequacy of future demand and supply of lithium but also ponder whether Li-ion batteries can sustainably power the future generation of motor vehicles (Gaines and Nelson, 2009).

“For a successful new technology to persist into the future, it is important to evaluate the reserve quantity, lifecycle economics, and potential security issues associated with the resource. The first step in assessing the technology is to develop a comprehensive understanding of the system in which the technology and resources reside. Once the system can be modeled, it becomes possible to assess the potential impacts that changes in other technologies, market demand, disruptions in component supply, labor, transportation, and other factors may play in the acceptance of that technology over time. In a resource-constrained world, especially when resources are not uniformly geographically distributed, it is important to be able to assess how the potential availability of scarce input resources will impact the long-term viability of the technology. For any constrained resource, recycling applications may alleviate pressures on the natural environment and improve the economic competitiveness of a technology that uses the resource. Therefore, a comprehensive understanding of the system in which the technology and resources reside is necessary to establish resource security, assess the benefits of resource recycling, and assess future viability of the technology (Guensler, 2014).”
The objective of this thesis is to identify the elements that should be included in a lithium process flow model and systems framework for the use of Li-ion batteries in motor vehicles. The thesis will identify and assess the key processes and flows involved in the lithium demand and supply on a global scale. The framework is based on the information derived from the literature review which is divided in the form of five chapters evaluating the important concepts all throughout the paper. Establishing the systems framework requires the identification of all elements that contribute to energy and resource consumption along the Li-ion battery lifecycle chain. The thesis also describes how the resulting framework can be adapted by others to develop a full energy model that can be used to quantify the lifecycle energy impacts of using Li-ion batteries to power future electric vehicle or hybrid vehicle fleets.

Chapter 1 provides a brief introduction of the electric vehicles and their types in the market, followed by Chapter 2, which provides a related background on the subject matter. Chapter 3 covers an extensive, in-depth literature review of the system, including: battery chemistry, components, and inner workings of the battery; the uses of lithium in the industry; sources and distributions of lithium resources; advantages and disadvantages of Li-ion batteries; and the fate of Li-ion batteries and few potential recycling options that are available in the industry. Chapter 4 introduces the elements and relationships in the lithium systems framework, which is built upon the research conducted in literature review. Chapter 5 outlines the next steps that are required to convert the process flow model and systems framework into an energy and resource consumption model. The chapter discusses data sources, variable relationships, and
programming requirements. The Chapter 6 concludes the paper, summarizing the major findings, discussing the broader impacts, and identifying next steps for future research.
2. BACKGROUND

Due to stricter laws and regulations governing vehicle production, vehicle manufacturers are under pressure to produce fuel efficient cars that limit air pollutant emissions. Under Corporate Average Fuel Economy (CAFE) standards of U.S. Environmental Protection Agency (EPA), legislation requires the car manufacturers to lower CO\textsubscript{2} emissions to 250g CO\textsubscript{2}eq/km (CO\textsubscript{2}eq is used to measure different greenhouse gases in same unit) by 2016 for the overall fleet average (The International Council on Clean Transportation, 2011). The focus of such legislation has propelled research in a direction where the battery system makes an integral part of the automotive system (Budde-Meiwes, et al., 2013).

The battery system depends on the various requirements of the vehicle, unique to its size, make, and model. Vehicles should install an appropriate battery size and composition to ensure their safety, lifetime, and performance. Li-ion batteries typically make up 25% (by weight) of the vehicle and are equipped with a variety of safety features. The lifetime of these batteries highly depends on their performance. Better performance ensures longer battery life, an incentive crucial to both consumers and manufacturers.

Battery performance is governed by two very important factors: energy, which generally deals with the driving range, and power, which is revealed in acceleration and top speed. There is usually a trade-off between range and performance. Batteries can either have higher energy or higher power, but not both (MIT Electric Vehicle Team, 2008). For example, batteries in an electric vehicle (EV) are generally energy based to
ensure a longer driving range; whereas, batteries in Hybrid Electric vehicle (HEV) are generally power-based for performance, given their ability to fully charge while driving. Plug in Hybrid Electric vehicle (PHEV) batteries use a combination that is both energy and power based. For shorter driving trips, they are energy-based and when battery becomes depleted, they are power based. These performance characteristics are shown in Table 1. To complement the performance of the batteries in electric vehicles, battery sizing is also shown in the Table 1.

Battery condition is another important criterion that helps ensure battery’s optimum functionality and is generally measured as a state of charge (SOC). The SOC is expressed as a percent of “maximum battery capacity” (MIT Electric Vehicle Team, 2008). There are two operating modes associated with SOC: charge depleting (CD), in which the vehicle activity is continuing to decrease the battery charge, and charge sustaining (CS), which retains a relatively constant charge in the battery for each mode of vehicle (Pesaran and Markel, 2007). The state of charge of batteries varies across different applications of EV, HEV, and PHEV. EVs generally run in CD mode, HEVs predominantly run in CD mode, and PHEVs run in both CS and CD mode.

Batteries are the governing part of the vehicle where their selection, sizing, design, disposal, and recycling are all crucial features that can impact the reliability, lifetime, and safety of the vehicle (Budde-Meiwes, et al., 2013).
Table 1 Performance Characteristics of Li-ion Batteries in EV, HEV, and PHEV (Lowe, et al., 2010)

<table>
<thead>
<tr>
<th></th>
<th>EV</th>
<th>HEV</th>
<th>PHEV</th>
</tr>
</thead>
<tbody>
<tr>
<td>Performance (Energy-based or Power-based)</td>
<td>Energy-based due to longer driving range</td>
<td>Power-based because batteries do not fully charge while driving</td>
<td>Energy-based for shorter driving trips and deriving energy from electric motor and stored battery power. Power-based upon battery depletion and acts as a HEV</td>
</tr>
<tr>
<td>State of Charge (SOC)</td>
<td>CD</td>
<td>CD</td>
<td>CD (~10-40 mi range) and CS @ 25% SOC</td>
</tr>
<tr>
<td>Power/Energy</td>
<td>2</td>
<td>15-20</td>
<td>3-15</td>
</tr>
<tr>
<td>Battery size</td>
<td>&gt;HEV and &gt;PHEV Ex: 24kWh (Nissan Leaf)</td>
<td>1-2 kWh Ex: Toyota Prius</td>
<td>5-15 kWh Ex: Nissan Leaf</td>
</tr>
</tbody>
</table>

Due to increasing greenhouse emissions and growing threat to resource security currently powering the transportation sector, there is an intense pressure on automakers to devise a new technology that can respond adequately to changing needs of the economy (Ford Sustainability Report, 2010). The development of Li-ion batteries employed in electric and hybrid cars are the result of that new advancement in the economy. The battery is a critical and a crucial component of the electric vehicle. The better the battery performs, the greater the utility derived by both consumers and manufacturers. Different battery chemistries serve unique needs to make and model of the car. However, a common factor across all battery technologies is the need to ensure the long term security of the materials used in a battery. That is, there needs to be enough material to meet the current and future demands of the market. Adoption of Li batteries is a function of battery characteristics, such as performance, state of charge capabilities, and size. As
with other battery technologies, Li-ion batteries pose some uncertainty with respect to the availability of Li as a resource. Ultimately, the systems framework will prove to be a useful tool to determine the amount of Li we need and how Li will be used to ensure resource efficiency.
3. LITERATURE REVIEW

This Chapter provides a literature review for lithium (Li) and the use of Li-ion batteries. This literature review is organized into eight sections: 1) types and configuration of electric vehicles; 2) types of Li-ion battery systems and their advantages and disadvantages; 3) Li-ion battery structure; 4) Li-ion battery mechanics; 5) key battery players in the market; 6) Li resources in nature; 7) Li global reserves and 8) fate of Li-ion batteries at the end of their lives. Each section contributes in developing a framework that will be described later in Chapter 4.

3.1 Types and Configuration of Electric Vehicles

Electric vehicles (EV) are playing an important role in changing the nature of the on-road vehicle fleet, especially for consumer automobiles. The latest generation of electric vehicles serves as a promise to a cleaner environment and a better fuel economy. Based on specific features and characteristics, EVs are modified and classified into general classes of Hybrid Electric vehicles and Plug-in Hybrid Electric vehicles. Each type of EV is reviewed below along with unique advantages and disadvantages.

3.1.1 All-Electric Vehicles

All-electric vehicles, known as EVs, run solely on electric motor without the use of internal combustion engine (ICE). The power is derived from the chemical energy in the battery pack and is capable of recharging from an electric grid (Nemry, et al., 2009).

- Advantages: EV’s are advantageous over CV’s because they use electricity as a fuel source rather than gasoline. Electricity is cheap and widely present in some countries.
This benefit is magnified if electricity is produced by renewable means (GoElectriveDrive.org, 2014). Generally, EVs require lower maintenance compared to conventional cars because electric motors work “without attrition” (Budde-Meiwes, et al., 2013). Therefore, electric vehicles can compete in the market given their lower maintenance and lower fuel cost, despite higher initial battery costs and that is possible because of government subsidies that are closing the gap and reducing the long payback period (Budde-Meiwes, et al., 2013). Performance factors such as “quiet motor, stronger acceleration, and smooth operation” make EV a viable option in the market (US Department of Energy and US Environmental Protection Agency, 2014b). Regenerative braking recovers energy during deceleration that is generally lost by brake heat in conventional cars to charge the batteries “via the reverse operated power generator” (Budde-Meiwes, et al., 2013). The energy “normally wasted during coasting and braking” of the vehicle is converted and stored in the battery until that energy is “needed by the electric motor” (US Department of Energy and US Environmental Protection Agency, 2014b). This function is not noticeable to the drivers but very crucial for the hybridization (Budde-Meiwes, et al., 2013).

Electric vehicles can also provide local environmental benefits by burning no gasoline and emitting no tailpipe emissions, thus reducing local pollutant concentrations. However, the total emissions of EV or hybrid cars today are highly dependent on the source of electrical power generation. Vehicles powered through renewable power source of wind, solar, nuclear, etc. can further reduce emissions and burn cleaner than non-renewable source of coal. From the political standpoint, the
domestic generation of energy from renewable sources will reduce the country’s
dependence on fossil fuels currently powering the transportation system.

- Challenges: There are many benefits of adopting EVs but their battery structure
  imposes some of the bigger challenges on their future. The Li-ion batteries costs
  anywhere from $5,000-$40,000 depending upon the model. Batteries are one of the
  most expensive parts of the car. The large battery packs installed in electric cars
  affects not only cost but also reliability and lifetime of the vehicle, therefore
  increasing the overall price of the car and potential battery maintenance expenses.
  Moreover, these battery packs are heavier and bulkier, taking up a considerable
  amount of vehicle space and increase the parasitic energy demand associated with
carrying extra weight. Charging such batteries can also prove hassle to drivers, as
drivers can spend around 4-8 hours to fully charge. Even 80% charge can take up to
30 min, unlike CVs which require only few minutes of refueling (US Department of
Energy and US Environmental Protection Agency, 2014a). The driving range of EVs
is still lower than that of CV’s. Most EV’s can travel up to 100-200 miles without
recharging, whereas, a gasoline powered vehicle can travel up to 300 miles without
refueling as reported by EPA’s fuel economy website (US Department of Energy and
US Environmental Protection Agency, 2014a). The lifecycle cost of EV (EV, in this
case, is equivalent to Nissan Leaf, 100 mpg-equivalent) is 6% higher compared to CV
(CV, in this case, is equivalent to Nissan Versa, 31 mpg) and 21% higher compared to
HEV (HEV is equivalent to Toyota Prius, 50 mpg), based on initial and usage costs
over 15 year period and 180,000 miles lifetime, discounting $7,500 in government
subsidy (Aguirre, et al., 2012). Comparing the usage cost for EV, CV, and HEV for
the same lifecycle period, an EV consumer (including $7,500 in government subsidy) would spent the same amount in electricity usage as a CV consumer for gasoline usage at the end of 13 years compared to a HEV consumer for gasoline at 8 years. Although, electric vehicles have lower fuel cost, the payback period is usually longer because of the higher initial costs. Therefore, improvement in battery development and decrease in cost can definitely affect the future of EV in upcoming years.

3.1.2 Hybrid-Electric Vehicles

Hybrid electric vehicles, or HEVs, more commonly known as Hybrids consists of an ICE, a fuel tank, an electric motor, and a battery pack (to provide electricity) as seen in Figure 1 below. The system contrasts with a conventional vehicle which only uses ICE as a single power source. HEVs were designed using a combination of gasoline engine and electric motor. HEVs can be found in three basic configurations: Series PHEV, Parallel PHEV, or Blended PHEV. Different combinations can operate in a parallel, series, or combined configuration as discussed in the next section.

- Advantages: Hybrid vehicles improve fuel economy, and increase power for electronic devices and power tools by incorporating advanced technologies in form of regenerative braking, electric drive, and Automatic start/shutoff (US Department of Energy and US Environmental Protection Agency, 2014a). The electric motor is an important step in hybridization process and allows for smaller and efficient engines to be used in a parallel HEV design by providing additional power to boost the engine during acceleration. One of the most widely known hybridization function is known as stop-start function or stop-n-go function. The combustion engine automatically
shuts off when vehicle is at a halt especially at traffic lights and starts again when the vehicle is accelerated with the help of an electric motor (US Department of Energy and US Environmental Protection Agency, 2014b). When the engine is turned off, all the electricity requirements are supplemented by the battery (Budde-Meiwes, et al., 2013). This function is designed to limit the idling time which uses more fuel standing than while moving; therefore, the vehicle generates fewer emissions.

- Disadvantages: The HEV battery is used only in the high power application as discussed in the Table 1 above. One of the major limitations of HEV is that the driving range for the pure electric portion is limited but is a reasonable option for silently cruising in residential areas (Budde-Meiwes, et al., 2013).

![HEV Components](http://www.fueleconomy.gov/feg/hybrid_diag.gif)

**Figure 1: HEV Components**

Source: http://www.fueleconomy.gov/feg/hybrid_diag.gif

### 3.1.3 Plug-in Hybrid Electric Vehicles

Plug-in Hybrid Electric vehicles, or PHEVs, are a crossover between an EV and a HEV. The PHEV combines the characteristics of both plugging in to charge directly from the grid and having an electric motor and an ICE. The pure electric range for PHEV
is generally higher than that of an EV because the vehicle can be charged by plugging into an electric outlet or at a charging station. On the other hand, when the battery level reaches SOC of ~20%, ICE can be used as a power source that allows PHEV to behave like a HEV (Budde-Meiwes, et al., 2013). Similar recharging characteristics apply to PHEVs as to EVs and PHEVs can achieve maximum driving range through both conventional gasoline and charging.

PHEVs are also found in three basic configurations: Series PHEV, Parallel PHEV, or Blended PHEV, of which different combinations can be found in research and in the market. Both Series and Parallel PHEV are shown in the figures below.

- Series: Series PHEV are also called Extended Range Electric Vehicle or EREVs (US Department of Energy and US Environmental Protection Agency, 2014c) because they use the internal combustion engine (ICE) to power the generator, which delivers the electric power to the electric motor and to charge the battery (Budde-Meiwes, et al., 2013). In this configuration, the ICE operates at an optimum efficiency (Budde-Meiwes, et al., 2013) and to reduce emissions as low as possible by decoupling engine and vehicle speed (Autonomie, 2013). The batteries in series drivetrain are assembled in form of building blocks and this design allows for a higher range of SOC and overall greater efficiency (Budde-Meiwes, et al., 2013). Because the electric motor is the only component directly attached to the wheels, this configuration generally requires a larger storage system and other components that add unnecessary weight and inefficiencies to the system (Autonomie, 2013). General Motors’ Chevy volt is designed using this system, see Figure 2 (Martin, et al., 2014).
• Parallel: In a parallel design, the ICE directly transfers mechanical power to the wheels via the gearbox and recharges the battery, like in series, but generally operates in a manner to maintain a lower SOC level as seen in Figure 3. Most European car manufacturers have focused on this design but U.S. and Japanese car manufacturers have mainly explored the extension of this system known as “power-split drivetrain” (or “series-parallel hybrid drivetrain”). In power-split system, two electric motors are used, one for powering and other for recuperating (Budde-Meiwes, et al., 2013). The engine power is divided along the two paths where “one goes to the generator to produce electricity” and the other goes through a mechanical gear system to drive the wheels (Autonomie, 2013). Both motors and the ICE are connected directly to the axle by a gear set (Budde-Meiwes, et al., 2013). Moreover, the power split configuration allows for freedom because engines, generator and motor speed are decoupled (Autonomie, 2013). An example of power split configuration is seen in Toyota’s Toyota Prius in Figure 4 (Martin, et al., 2014).

![Figure 2: Series Hybrid Drivetrain (Martin, et al., 2014)](image-url)
Depending upon the configuration of the drivetrain, different technical capabilities can be achieved but from the battery’s point of view either configuration is useful and does not make any difference (Budde-Meiwes, et al., 2013).

- Advantages: One of the major advantages of PHEV is that PHEV shares the characteristics of both the hybrid electric car and the electric car, thereby reducing the use of ICE and consumption of liquid fuel (Greenlight Initiative, 2007). This feature serves both consumers and the economy. By charging the car with electricity, consumers can save money on fuel and the nation can reduce dependence on imported fossil fuel (oil), lower greenhouse gas emissions, and improve in air quality.
According to Elgowainy, et al. (2009) at Argonne National Labs, PHEVs use 40-60% less fuel (gasoline and diesel) than conventional vehicles. The performance of PHEVs is dependent on the battery packs which are charged by electricity, and if the electricity is produced renewably, PHEV’s are even more environmentally friendly. Another potential benefit is that PHEV’s can sell the electricity back to the grid during peak hours when they are not using the electricity and can recharge again during off peak hours.

- Disadvantages: PHEV’s major challenge is the use of battery technology. The large battery pack used to propel PHEV is expensive and heavy compared to that of HEVs. Additional cost is likely to accrue when the battery needs to be replaced (Greenlight Initiative, 2009). PHEV’s generate zero tailpipe emissions, but emissions are now shifted and added to the electric plants. As a result, PHEVs still cause air pollution. The very basis of PHEV, a plug, is difficult to find outside one’s garage and there are limited options for recharging. For PHEV’s to be commercially viable, a charging infrastructure will need to be put in place. Also, vehicle performance depends on driving and charging patterns, and efficiency in CS and CD mode (Nemry, et al., 2009).

3.2 Different Types of Li-Ion Battery Systems and their Advantages and Disadvantages

Li-ion is the fastest growing battery chemistry in today’s market. Having already been used in consumer electronics in form of laptops and cell phones, they seem promising to power this era’s electric cars, provided they meet the challenges in
automotive applications. “Lithium ion batteries’ is an umbrella term for a variety of material combinations used to form batteries” (Budde-Meiwes, et al., 2013). These unique combinations help establish safety, lifetime, power, and other technical measures to offer varying performance. Different designs within the battery allows for the optimizations towards either high powered cells or high energy cells, discussed in chapter background and leads to a tradeoff in terms of advantages and disadvantages.

- **Advantages:** Li-ion batteries have been around since 1991, even though their introduction in automotive industry has only been recent (Golubkov, et al., 2013). Their lightweight and high capacity utilization at high current rates makes them suitable for such applications (Budde-Meiwes, et al., 2013). The high energy density of Li ion is twice that of NiCd currently and if technology continues to improve, there is a likelihood that Li ion energy density of triples to that of NiCd (Battery University, 2010). One of the major advantages of Li-ion batteries over other chemistries is that they are “a low maintenance battery”. Based on the market research conducted by Goriparti, et al., (2013), the cells in Li-ion batteries show the highest gravimetric energy and power densities among all other commercial rechargeable chemistries. Li ion is capable of self-discharging at a rate half of NiCd and NiMH, thus, making it suitable for use as a rechargeable batteries in evolving transportation sector (Goriparti, et al., (2013). Moreover, their disposal cause little harm compared to other non Li-ion chemistries based on the current information and is still under further research.

- **Disadvantages:** One of the major limitations Li-ion batteries possess is their high manufacturing cost. Lithium-on batteries cost twice as much as Nickel metal hydride
batteries for the same battery capacity (Urken, 2009). The high cost of battery manufacturing coupled with battery replacement impacts both manufacturers and consumers. Also, these batteries are subject to aging, a phenomenon where battery capacity is diminished over time, use, and temperature (Motorola Solutions, 2014) even when they are not in use. Extreme weather conditions such as hotter temperatures are also known to affect the lifetime of Li-ion batteries through battery degradation (Axsen, et al., 2008). The optimum temperature to charge Li-ion battery is 0 – 45°C, according to the Panasonic technical handbook on Lithium-ion batteries (2007). Li-ion also requires protection during over charging or discharging cycles to ensure safety and longer lifetime. Li-ion batteries are still considered an immature technology in the market today as the battery chemistries and compositions continually change with time. A great deal of research and development is taking place to overcome both known and unknown limitations and to establish uniformity in battery infrastructure (Battery University, 2010). Though, mass production of this technology at a feasible rate can be expected in the near future (Battery University, 2010).

The unique battery technologies in the market today all have limitations in one form or another. The Table 2 below shows the advantages and disadvantages of 4 battery technologies: Lead-acid, Nickel-metal hydride, Sodium-nickel chloride, and Lithium-ion, when employed in different types of hybrids. More advanced features can be found at varying levels of hybridization.

In Table 2, micro-hybrids are conventional vehicles, with an internal combustion engine that reduces fuel consumption and CO₂ emissions through simple stop-and-start
functions (Budde-Meiwes, et al., 2013). Mild-hybrids constitute the next level of hybridization, generally providing a boost during acceleration. At the top of the hierarchy are full-hybrids and plug-in-hybrid. These two types generally use larger batteries, different drivetrains (to transmit power), and bigger power-assist features to improve efficiency and extend battery life. As outlined in Table 2, the traditional lead-acid battery is more advantageous for micro-hybrids because of low cost and safety features. Nickel-metal hydride appears to be better suited for full-hybrid vehicles given their longer lifetime, power density, and maturity. Sodium-nickel hybrid battery technologies don’t show any clear advantages for hybrids. Lithium-ion has a higher density and higher power which is suitable for both micro-hybrid and mild-hybrid cars; however, with improvements in battery management system, Lithium-ion batteries can be used in full-hybrids and plug-in-hybrids as well (Budde-Meiwes, et al., 2013).

<table>
<thead>
<tr>
<th>Battery Technology</th>
<th>Micro-Hybrid</th>
<th>Mild-Hybrid</th>
<th>Full-Hybrid</th>
<th>Plug-in Hybrid</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lead-Acid</td>
<td>Charge acceptance</td>
<td>Charge acceptance and power density</td>
<td>Power density</td>
<td>Weight and power density</td>
</tr>
<tr>
<td>Nickel-metal Hydride</td>
<td>Low deep temperature performance</td>
<td>Limited cost reduction</td>
<td>-</td>
<td>Cost and weight for larger batteries</td>
</tr>
<tr>
<td>Sodium-nickel chloride</td>
<td>Heat loss and low power</td>
<td>Heat loss and low power</td>
<td>Heat loss and low power</td>
<td>Too high heat loss, power limited small batteries</td>
</tr>
<tr>
<td>Lithium-ion</td>
<td>Cost</td>
<td>Cost</td>
<td>Battery management system</td>
<td>Battery management system</td>
</tr>
</tbody>
</table>

Source: Budde-Meiwes, et al., 2013
3.3 **Battery Structure**

This section focuses on the four main components of the battery (cathode, anode, electrolyte, and separator) and how they contribute to inner workings of a battery. Batteries can accept different combination of chemistries that are mainly unique in their cathode composition or anode composition. Lithium is used in battery manufacturing in the cathode and electrolyte, primarily in the form of lithium carbonate or lithium chloride, depending upon the chemistry and the composition (Legers, 2008). The information presented in this section helps to identify and assess the flow of lithium in the framework that will be described in Chapter 4. Tracking the flow of lithium throughout the components of the battery can provide great insights into economics of lithium recycling and material constraint that may present in future.

A battery comprises of four main components: cathode, anode, electrolyte, and separator, all of which are discussed in detail below along with their advantages and challenges.

### 3.3.1 Cathode

For production of the cathode, lithium in the form of lithium oxide is used instead of the metallic form of lithium. Cathode material paste consists of lithium carbonate (or other Li oxides), a binder (poly vinylidene fluoride (PVDF) or such), some carbon material in form of graphite or fiber etc., and solvent such as N-Methyl-2-pyrrolidone (NMP) which is coated on aluminum foil and “serves as a current collector” (Gaines and Cuenca, 2000; Lowe, et al., 2010). All the parts are carefully assembled to achieve a precise structure of cathode.
Different composition of material has successfully provided four types of cathodes in the market. All of these combinations have their unique pros and cons and are shown in Figure 5. Lithium manganese oxide (LMO) is one of the most commonly used cathode material in hybrid and electric cars today. Lithium cobalt oxide (LCO) dominated the consumer electronics because of high energy density before being used in the cars. Due to growing safety concerns and rising prices of cobalt, manufacturers may be opting for a cheaper alternative such as LMO and lithium iron phosphate (known as LFP). Different chemistries using Nickel have also been widely used before growing interest in Lithium. NCA (nickel cobalt aluminum) and NMC (nickel manganese cobalt) are two nickel-based chemistries amongst which NCA uses some lithium content whereas NMC doesn’t use any. Each chemistry has pros and cons and is used depending upon the requirements of the vehicle use and the conditions of the market. The pros and cons of different chemistries are shown in Figure 5 below (Lowe, et al., 2010). Gaines and Cuenca (2000) of Argonne National Lab state that, “The different electrode materials have different current-carrying capacities, and this affects the storage capacities of the resultant cells.” Therefore, researchers are developing high voltage cathode material to achieve better vehicle performance.
3.3.2 Anode

The anode, or negative electrode, is generally made from graphite (carbon) and is coated on a copper foil. The anode receives Li-ion charge during charging cycle and emits it to cathode during discharging cycle (Lowe et al., 2010). The active material on anode paste consists of graphite, binder, solvent, and carbon to obtain two types of electrode structures: “highly crystallized natural graphite and randomly crystallized artificial carbon” (Lowe et al, 2010).

Anodes made of graphite allow a single lithium ion to be intercalated (inserted between layers) in its hexagon structure at a full charge of LiC₆ composition. Currently in best practices, a 2.5 Li ion can be intercalated for each hexagonal carbon structure reducing the amount of anode in comparison to cathode and can achieve the theoretical capacity of 750 milliAmphours/gram; twice that of the LiC₆ composition (Gaines and
Additional research and development advancements are being focused in obtaining more capable and stable material (Gaines and Cuenca, 2000).

### 3.3.3 Electrolyte

The electrolyte is a solution of lithium salts and organic solvents, where organic solvents are used to increase the solubility of lithium salts and decrease the viscosity of the electrolyte. The function of an electrolyte is to act as a conductor to pass Li-ions between cathode and anode (Lowe, et al., 2010). If Li-ions are mobile in the solution, the battery can perform better. Organic solvents such as ethyl methyl carbonate (EMC), dimethyl carbonate (DMC), diethyl carbonate (DEC), propylene carbonate (PC), and ethylene carbonate (EC) can be used in combination of lithium salts such as lithium hexafluorophosphate (LiPF₆), lithium perchlorate (LiClO₄), and lithium hexafluoroarsenate (LiAsF₆) (Lowe, et al., 2010). Other types of electrolytes such as gel electrolyte and solid polymer electrolytes have also been developed. This new class has been successful in providing enhanced safety, lighter weight, and design flexibility, but has been unable to achieve the required performance (Gaines and Cuenca, 2000).

### 3.3.4 Separator

The separator is a “micro-porous membrane” (Lowe, et al., 2010) that allows Li-ions to pass through the pores in the separator. The separator is usually made of polyethylene or polypropylene. The most important function of separator is to act as a safety device in the battery. In the event of the battery becoming too hot, the separator melts closing off the pores on the electrodes, preventing ions from travelling back and forth to conduct electricity (Gaines and Cuenca, 2000).
3.4 **Mechanics of Batteries**

Li-ion batteries, like any other batteries, store electrical energy that can be delivered through an electrochemical reaction. The battery is composed of individual cells that produce electricity that travels through four main components: anode, cathode, electrolyte, and separator. Electrons flow from the anode, a negatively charged electrode to the cathode, a positively charged electrode, through the electrolyte solution when connected by a wire or any electrical conductor, thus creating an electric current. The electrodes: anode and cathode are chosen in such a way that they are compatible with each other. An anode should have the tendency to donate electrons, creating cations or positive ions in the electrolyte that the cathode can easily accept, creating anions or negative ions. Such tendency to donate or accept electrons is expressed as standard electric potential and the difference between the electrode potentials of anode and cathode gives the cell, voltage. In the case of Li ion batteries, Lithium acts as a cation travelling from anode to cathode. Being the third smallest element in the periodic table, Li can be easily ionized to Li$^+$. During the battery’s charging cycle (shown in Figure 6) lithium ions move from cathode to anode through the electrolyte and stick to the carbon on the anode (American Physical Society, 2014). In the discharge cycle shown in Figure 7, ionized lithium is emitted to the electrolyte and travels back to LiCoO$_2$ on the cathode. LiCoO$_2$ is one of the most common cathode material used (Lowe, et al., 2010). This movement of Li ions produces a high voltage of 3.6 volts, more than twice to that of alkaline battery, and higher density. Li-ion batteries are rechargeable type of batteries and are “recharged by running the anode and cathode reactions in reverse (American
This ability of the battery to be recharged continuously with little loss of capacity proves very advantageous in vehicle applications.

Figure 6: Li-ion Battery Charge Cycle (Brain, 2006)

Figure 7: Li-ion Battery Discharge Cycle (Brain, 2006)
3.4.1 Safety

Li ion batteries are manufactured with various safety features. In case of high temperatures, the charging voltage is restricted and battery shuts down. If there is excess buildup of the pressure, safety protections prevent a deeper discharge cycle further inhibiting battery recharge. This safeguard allows for the safer batteries “but it also reduces the fraction of the battery that is used to store energy, and also slowly drains the battery even when the device is off (American Physical Society, 2014).”

3.4.2 Challenges and Future Research

One of the major challenges faced by Li-ion batteries in automotive applications is lower energy densities. Efforts are being made to develop electrode materials that can hold more charge in a given volume. Thus, research groups are focusing on using silicon as an anode material instead of carbon based graphite. Silicon has a high storage capacity of up to ten times that of graphite, but has a shorter life as a material and can cause pulverization (American Physical Society, 2014). This drawback can be overcome by using silicon nanowire technology developed by Yi Cui and her colleagues at Stanford University (2007). Furthermore, using Germanium made anode wires may double the recharge capacity of Li-ion batteries (Kennedy, et al., 2014). With a breakthrough in material development, such as mentioned above, it is difficult to predict the future battery compositions and how effectively they will change the market structure. Advancements are being made in the electrolyte material as well. The founder of Seeo, a Lithium-ion battery start-up company at University of Berkley, has developed polymer based electrolyte “that offers unprecedented safety and lifetime” (Department of Chemical and
Bimolecular Engineering: Tulane University, 2011). In 2009, Hiroyuki Nishide of Waseda University in Tokyo was developing a “totally organic battery” using polymer electrodes made of organic radical molecules” (Popkin, 2009). As of 2014, researchers have been focused on exploring different organic chemistries for electrolyte as well as improving the electrochemical performance of organic Li-ion battery electrodes.

3.5 Key Lithium-Ion Battery Players

The global supply chain for Li-ion batteries is presented in Figure 8 (Lowe, et al., 2010), showing the key players in the lithium ion battery component market.

A variety of players are involved in manufacturing of key materials: cathode, anode, electrolyte, and separator, manufacturing of cell components and electronics and integrating them into cell and battery packs so they can be sold to the players in automotive sector. In the key materials section of the chart, many different players are involved in producing cathode related compounds, anode related compounds, and electrolyte related compounds, all of which use lithium in some form or another. Each company is listed under their specialized skill in each section. There are companies that only focus on producing key materials and those are listed relevant to the materials they produce. The companies that specialize in producing the components of the cell and attached electronics such as wires are listed under cell components and electronics section. These cell components are integrated by other key players and are sold to vehicle manufacturers to be incorporated into the vehicles. Figure 8 also highlights in the bottom right hand corner players unique to U.S. (Lowe, et al., 2010). The framework in chapter 4 only tracks Nissan Leaf’s lithium flow is based on a similar idea as the chart
below but can be expanded to include lithium flow for other vehicles when the systems
model is further developed.
3.6 Lithium Resource Base

The goal of this section is to identify how lithium is obtained and major and minor end uses of lithium in the market. Lithium is found in mainly in brines and minerals but this chapter further explores where and how much lithium is locked in different types of brines and minerals and other lesser known sources of lithium deposit, such as Jadarite.

3.6.1 Uses of Lithium

Li is one of the lightest metals in the periodic table and serves an important economic and industrial purpose. Lithium is mostly used in ceramics and glass industry as it comprises 35% of usage distribution in 2013 as shown in Figure 10. About 29% of Li is used in consumer electronics, especially in batteries for cell phones and laptops. Recently, Li has showed a significant potential for use in electric and hybrid cars (Bradley and Jaskula, 2014). The other lesser known uses of Li range from lubricants to glass to manufacturing to even in pharmaceuticals. Li is used in glass and ceramics to provide strength and temperature resistant, in greases and lubricants to produce heat resistant and alloyed with aluminum and copper to produce lightweight structural airframe components. In pharmaceutical industry, Li is used in dental ceramics and in few psychiatric medications. Li can also be used in production of nuclear weapons, provided that Li is in a different isotopic form (Bradley and Jaskula, 2014).
Figure 9: Lithium Industrial Market Segments in 2013

The lithium distribution has definitely changed over last six years as the distribution shifted from 25% batteries in 2007 to 29% batteries in 2013. Batteries in 2007 made up the largest piece of the distribution compared to ceramics and glass in 2013. Use in lubricating grease, pharmaceutical and polymers, air conditioning has decreased by 3%, 2%, and 1% respectively. Aluminum use on the other hand has slightly increased by 2% (Goonan, 2012).

3.6.2 Sources of Lithium Deposit

Li, one of the most widely distributed metals on earth’s surface, does not occur naturally in pure elemental form due to lithium’s energetic reactivity with water. Li of commercial value is primarily found in two forms, brine deposits and hard rock minerals (Brown, 2010) Brine deposits are classified into three types: continental, geothermal, and oilfield with continental being the most common of three (Pistilli, 2012). These brine
deposits hold 66% of the world lithium reserve and can be found in salars (salt flats) of Chile, Argentina, China, and Tibet (Pistilli, 2012). The Global lithium reserves are reported at 39 million metric ton (MT) according to Pistilli (2012).

3.6.2.1 Continental Brine

Continental brines are one of the most common brine forms containing lithium and are known by variety of names: continental saline desert basins, salars, salt lakes, and salt flats, depending upon the region. These brines are composed of “sand, minerals with brine, and saline water with high concentration of dissolved salts (Pistilli, 2012). Li in these brines is derived mainly from the leaching of volcanic rocks found in desert areas and vary in their lithium content largely due to solar evaporation (Evans, 2008). The Li content ranges in highly concentrated brines at high altitude of Chile, Bolivia, Argentina, China, and Tibet from 100-7,000 ppm (Kunasz, 2006) to modest Li content of 100 - 300 ppm (Kunasz, 2006) in subsurface brines in Silver Peak, Nevada, to low concentrations ranging from 28 to 60 ppm (Kunasz, 2006) in Great Salt Lake, Utah. The low Li content occurs as a result of a low evaporation rate and constant dilution rate caused by the “high volume of fresh water influx” (Evans, 2008; Brown, 2010). If the brine deposit surface consists of silts and clays exhibiting “less salt than a salar”, it is known as a playa (Pistilli, 2012).

3.6.2.2 Geothermal Brine

Geothermal brines consist of hot, concentrated, saline solution that passes through crustal rocks in high heat flow. The solution is enriched with lithium, boron, chlorides of Sodium, Potassium, etc., that leaches from the crustal rocks (Allaby and Allaby, 1999).
Dissolved metals are often found in geothermal brines where they form “intermediary in the deposition of ore deposits” to extract lithium at a commercial value (Allaby and Allaby, 1999). Low quantities of these brines are present at Wairakei, New Zealand, Reykanes Field, Iceland, and El Tatio, Chile. The largest known domestic source is in the Salton Sea in California. A reverse osmosis process is used to extract Lithium carbonate from the Salton Sea, eliminating the need for solar evaporation (Pistilli, 2012).

Geothermal mines comprises of 3% of known global lithium resources according to Melissa Pistilli, author of ‘Lithium Deposit Types” an online article on Lithium Investigating News (Pistilli, 2012).

3.6.2.3 Oilfields

Brines containing lithium are also found in subsurface oil accumulation. There is estimated 0.75 million ton of lithium at an average concentration of 0.15% is owned by Smackover Formation on the US Gulf Coast where the concentration is as high as 700 milligrams/liters. Oilfield brines spans the US states of Wyoming, North Dakota, Oklahoma, Arkansas, and East Texas (Kunasz, 2006; Pistilli, 2012).

Brine deposits depend on multiple key geologic and geographic factors such as lithium grade, magnesium to lithium ratio, evaporation rate, elevation, surface area, porosity, depth, and density, all of which are listed in the Figure 10 below. The table compiles the data from the available basins in the world and shows the geologic factor pertaining to each. The basic brine basin information table below is taken from the journal article titled ‘Lithium Resources and Production: Critical Assessment and Global Projections in 2012’ (Mohr, et al., 2012). The key points to focus in the table are the
lithium grade, Mg:Li ratio, and evaporation rate. Atacama Salar in Chile has the highest lithium grade by weight of 0.15% and highest evaporation rate of 3-3.7 meter/year. Higher lithium grade and higher evaporation rate decreases the time the brine is exposed in the evaporation ponds cutting down the overall time and labor in the process. One of the lowest Magnesium to lithium (Mg:Li) ratio is found in Zhabuye deposit in China. Low Mg:Li ratio makes it easier to separate Li from Mg, reducing production cost (McNulty and Khaykin, 2009).

![Figure 10: Brine Basin Characteristics (Mohr, et al., 2012)](image-url)
3.6.3 Rocks

Li in rocks is determined primarily on two specifics: the Li grade of the deposit and associated iron content. Generally, in batteries technical grade Li is used and is high in purity. On the other hand, iron content is treated as an impurity and is not useful to end users. Thus, lower the iron content; better the quality (Mohr, et al., 2012).

3.6.3.1 Pegmatite or “Hard Rock”

Variable amount of Li concentrations are also found in a specific type of igneous rocks known as pegmatite. Pegmatite consists of coarse-grained igneous rocks that are formed from the crystallization of magma below the earth’s surface. Pegmatite mainly contain feldspar, mica, and quartz along with exotic elements such as lithium, tin, tantalum, cesium, etc. (Kunasz, 2006) and accounts for ~ 26% of known global lithium reserves (Pistilli, 2012). The most common rock forming mineral in pegmatite is spodumene, which contains lithium that can be extracted via an acid fusion process. Other minerals known to hold Li are lepidolite, petalite, and amblygonite.

Minerals such as spodumene, petalite, and lepidolite serve “as the feedstock for the production of lithium chemicals” (Brown, 2010) as an end use besides producing Li. If the iron content is low they can be used in glass and ceramic industry. Spodumene is also used as a concentrate for production of lithium chemicals and mining developments in China (Brown, 2010).

Hard-rock or pegmatite ore is extracted using conventional mining operations in open pit or in underground mines. The extracted ore is then processed and concentrated
using various methods before it is commercially usable (Western Lithium Corporation, 2009). In comparison to lithium produced from brine deposits, the Li extracted from hard rock is expensive. If the lithium content is higher in hard rocks, the extraction process can be justified as economical. Moreover, the production of other rare and commercial elements can help offset the increase in production cost.

3.6.3.2 Spodumene

The largest commercial source of Li is found in this particular mineral and Li content can range anywhere from 1.35% to 3.56%. Theoretically, Li content can go slightly higher to 3.7%. Typically, Li concentrations have a narrower window and operate from 1.9% to 3.3% (Kunasz, 2006). The actual concentrations are higher because sodium and potassium are replaced by lithium. Spodumene occurs all around the world and was conventionally used to extract lithium before brine deposits were discovered and utilized. Spodumene mines can be found in Sweden, Austria, Brazil, Argentina, Canada, Australia, Russia, etc., some of which may or may not be operational (Kunasz, 2006). Australia appears to have the largest spodumene producing pegmatite operation, with an estimated 560,000 tonnes of Li ore in reserves at average concentration of 1.6% Li (Pistilli, 2012).

3.6.3.3 Clay Deposits

Significant concentration of Li also occurs in hectorite minerals which are found in specific class of clay deposits known as smectite (Western Lithium Corporation, 2009). The name Hectorite is derived from a place in California, called Hector where 0.7% (by weight) Li deposits are found. The Li concentration itself in hectorite is 0.53%
(Kunasz, 2006). Hectorite is high in lithium and magnesium (Pistilli, 2012). According to the geologist Keith Evans (2008), the straddles of Nevada/Oregon border in the U.S. possess the largest known clay deposits of Li. Western Lithium is currently carrying out mining and drilling operations to obtain Li in the area (Pistilli, 2012). Another place where hectorite can be found is in Clayton Valley, Nevada (Kunasz, 2006). Figure 11 provides a pie chart based upon data compiled by Western Lithium and Evans (2008) reflecting the distribution of the lithium sources. Most of the lithium comes from the continental brines (57%), followed by mining from hard rock at 25%. Hectorite deposits contribute 7% to lithium extraction whereas geothermal brines, oilfield brines, and Jadarite provide 3% each.

3.6.3.4 Lacustrine Evaporites

Lacustrine (lake) evaporites (crystals resulting from evaporation) arise from “direct precipitation in saline lakes (Starkey, 1982).” The most commonly known deposit, Jadarite, is named after its appearance in Jadar Valley in Serbia. Jadarite is high in lithium and borate and has been ranked as the largest lithium deposits in the world (Rio Tinto Minerals, 2011). The Jadarite deposit reportedly contains 125.3 million tons of resources with an average 1.8% lithium dioxide concentration and 16.32 million tones \( \text{B}_2\text{O}_3 \) (Boron Trioxide). The company also believes that the deposit “is one of the largest undeveloped lithium sources in the world, with the potential to supply more than 20% of global lithium demand” (Rio Tinto Minerals, 2011).
Yaksic, et al. (2009) developed a chart (Figure 12) that sums up the lithium resources, reserves, products, and major end-use applications, in which they identify four types of lithium resources: minerals, brines, clays, and sea water, and two types of lithium reserves: minerals and brines. Lithium products are produced from these reserves that are used in industries and markets depending upon the composition of the product. Lithium hydroxide can be extracted directly from minerals and produced indirectly from lithium carbonate and is used in greases, lubricants, batteries etc. Lithium carbonate is produced from both minerals and brines and is used in aluminum, continuous casting, pharmaceuticals, batteries, glazes and frits, etc. Lithium carbonate, in turn, can produce lithium metal which upon further processing produces butyl lithium currently used in synthetic rubber, polymers and organic derivatives. Lithium chloride is obtained from
brines and is only used in dehumidifier systems. Minerals also produce concentrates used in glazes and frits.

Figure 12: Flowchart of Lithium Resources, Reserves, Products, and Major Und-use Applications. (Yaksic, et al., 2009)
3.7 Li**thium Global Reserves**

In 2013, more than 75% lithium is produced by Chile (38.6%) and Australia (37.1%) collectively. China produced 11.4%. Argentina and U.S. production were far lower at 8.6% and 4.3% respectively (Figure 13).

![Figure 13: Global Lithium Production in 2013](http://mcgroup.co.uk/researches/lithium)

A world map of global Li reserves (tons) can be seen in Figure 14, and the major geographic sources of lithium are discussed in the sub-sections that follow. A more in-depth table detailing the global distribution of lithium, lithium characteristics, and deposit type for each country is provided in Figure 15. Figure 15 shows various brine across globe and their characteristics, such as Lithium grade, ratio of Magnesium to Lithium, evaporation rate, elevation, surface area of the brine, porosity, depth, density, and when the brine started.
Figure 14: 2010 Global Lithium Reserves (tons)

Figure 15: Brine Basin Information (Mohr, et al., 2012)
3.7.1 Latin America

Currently, more than half of the Li production of the world comes from the continental brines of Chile, Argentina, and Bolivia, or more commonly known as the “Lithium Triangle.” These three South American countries holds vast majority of lithium reserves. Figure 16 illustrates the location of lithium brines in three countries of Chile, Bolivia, and Argentina in the South America “lithium triangle.” (Robles, 2013)

3.7.1.1 Chile

One of the major lithium producing countries, Chile, holds approximately one third (~ 27%) of global lithium reserves (Jaskula, 2012) at Salar de Atacama. This salar is
largely operated by Sociedad Quimica y Minera of Chile, South America. (SQM). This single operator produces one third of total capacity of lithium compounds from the salar, including both Lithium carbonate and Lithium hydroxide. According to the Chilean Institute of Investigaciones Geologicas (IIG), Salar de Atacama contains high concentrations of Potassium and lithium at salar’s periphery (Kunasz, 2006). The lithium content ranges from 1,000-7,000 ppm and the ratio of Magnesium to lithium (Mg:Li) is 6.6:1, which is almost 4.5 times higher than that of at Salar de Hombre Muerto in Argentina, as can be seen in table below. The higher the Mg:Li ratio is, the higher the processing cost of lithium is. The brine is located at an elevation of 2,300 m where it covers the surface area of ~ 3,000 km². Due to the high elevation, the Salar de Atacama receives high amount of sunlight which increases the rate of solar evaporation of the solar ponds (Kunasz, 2006). Rockwood Holdings also has lithium mining rights on the brine. Both companies transport their products to a lithium carbonate plants in Antofagasta for further processing (Ober, 2001; Bradley and Jaskula, 2014).

3.7.1.2 Argentina

Argentina is one of the major exporters of lithium compounds, especially, lithium chloride, lithium carbonate, and lithium hydroxide (Jaskula, 2012). Most of the lithium found in Salar de Hombre Muerto in Andes Mountain at an elevation of 4,000 m, covering the area of 280 km² of salt nucleus is recovered (Jaskula, 2012). The lithium content in this salar is relatively low compared to that of Salar de Atacama in Chile and Salar de Uyuni in Bolivia but has a low concentration of “impurities” such that Mg:Li ratio is 1.37:1 (Jaskula, 2012). The lower ratio of Mg:Li and fewer impurities means the salar can be harvested at lower processing costs, ensuring higher benefits.
3.7.1.3 **Bolivia**

Recently, it was claimed by Estuardo Robles (2013) in his article “Lithium-The Mineral of the Future”, that Bolivia holds the largest deposit 50-70% of Lithium in the world at undeveloped brine called Salar de Uyuni. According to the USGS (2014), these mega salt flats contain 100 million tonnes of lithium over the area of more than 11,000 km² and covers ~55% of the total global reserves. The anticipated full scale production capacity of this brine is 20-30 kiloton Li/year as reported by Steve Mohr and et al in their article assessing lithium availability. It is interesting to note that the composition specific to this region of Salar de Uyuni has exceedingly high ratio of Magnesium to Lithium, three times to that of at Salar de Atacama; thus; refining salt into lithium carbonate is less economical and more time consuming (Brown, 2010).

This untapped lithium resource in Bolivia has been attracting massive press attention as “the Saudi Arabia of Lithium” because of salar’s capability to fulfill any imaginable production demand (Romero, 2009). But due to troubled political relations and current unfavorable interest of Bolivian administration in foreign mining rights itself; the resource might be sheltered from potential use.

**3.7.2 United States (U.S.)**

Most of the lithium in U.S. comes from a brine operation in Silver Peak, Nevada. Currently Rockwood Holdings, American Lithium Minerals, and Rodinia Minerals operate this facility (Brown, 2010). The Silver Peak facility is the only active brine facility in Nevada, U.S., and has Li reserves of 118,000 tons in a 20-square-mile area (Daly, 2013). According to Jaskula, (2014) the Nevada mine’s production capacity was
expanded in 2012 to install a new lithium hydroxide plant in North Carolina in response to the increased price of imported Li compounds. Prior to the existence of this operational facility, U.S. produced Li from the minerals, primarily from spodumene in North Carolina, but there was a higher cost associated with obtaining Li. The introduction of cheaper brine operations in Chile in 1998, had led to the shutdown of North Carolina rock operation as other brine related operations were explored and promoted (Wallace, 2012). Roughly 28,000 tons of Li was found in a 25 square mile area of Wyoming, which upon extrapolation represents 18 million ton of Li in a 2000 square mile area worth up to $500 billion at 2013 market prices (Surdam, 2013). The numbers look promising enough to supplement the demand for U.S. production, provided that demand for lithium continues and can be recovered economically (Daly, 2013). The important thing to note about the brine operation in Silver Peak is low Mg:Li ratio of 1.5, compared to that of Salar de Atacama and Salar de Uyuni (see Figure 17), making Silver Peak ore very viable for lithium extraction.

<table>
<thead>
<tr>
<th>Cation</th>
<th>Silver Peak, Nevada</th>
<th>Salar de Atacama, Chile</th>
<th>Salar de Uyuni, Bolivia</th>
</tr>
</thead>
<tbody>
<tr>
<td>Li</td>
<td>0.023</td>
<td>0.14</td>
<td>0.025</td>
</tr>
<tr>
<td>K</td>
<td>0.53</td>
<td>1.87</td>
<td>0.62</td>
</tr>
<tr>
<td>Na</td>
<td>4.43</td>
<td>6.92</td>
<td>9.1</td>
</tr>
<tr>
<td>Mg</td>
<td>0.033</td>
<td>0.91</td>
<td>0.54</td>
</tr>
<tr>
<td>Mg:Li</td>
<td>1.5</td>
<td>6.6</td>
<td>21.5</td>
</tr>
</tbody>
</table>

Figure 17: Partial Cation Chemical Analyses (weight%) of Brines in US, Chile, Bolivia (Kunasz, 2006)
3.7.3 Canada

Canada is involved in producing Li compounds from mineral processing of spodumene (Jaskula, 2012). The total estimated production capacity is 550 metric tons and estimated reserves are 500,000 metric tons in 2013 (Howard, 2014). The USGS literature (2014) and Howard (2014) estimate 1,000,000 metric tons of Li resources in Canada. In 2012, Brian Jaskula, the author of “Lithium” published by USGS.gov, reported that Canada Lithium Corp at their Quebec plant produced Li carbonate from an open pit mine at a full production capacity at 20,000 tons/year and was also involved in agreements with China and Japan to produce “battery grade Li carbonate” at varying production rates. Nemaska Lithium, Inc. (2012) measured lithium hydroxide resource of “10.2 million metric tons (Mt)” where lithium oxide was graded at 1.53% and additional 9.4 Mt graded at 1.45% lithium oxide. This Quebec plant is expected to produce up to 20,700 tons/year of lithium hydroxide and 10,000 tons/year of lithium carbonate (Jaskula, 2012). From the feasibility analysis conducted in 2011 by Lithium One Inc., another Li mining company in Canada, it was disclosed that their processing plant in Quebec could produce battery grade lithium carbonate of 17,000 tons/year (Jaskula, 2012). Thus, the land in Quebec is rich when it comes to Li mining in Canada.

3.7.4 China

Unlike other countries, China obtains Li from a combination of sources. China holds interest in both rock based and brine based specifically, continental brine sources. The large quantities of lithium carbonate is produced domestically and also from the imported spodumene. One third of the country’s lithium production comes from brines,
out of which only 13% was used in 2011 (Jaskula, 2012) and the rest two third of lithium production comes from minerals extracted domestically which is 8,750 tonnes of lithium carbonate equivalent (LCE) as Li₂CO₃. Lithium carbonate (Li₂CO₃) can be converted to lithium by multiplying lithium by a conversion factor of 5.3. Spodumene is found mainly in Sichuan Province but is also seen in other places such as Hunan, Jiangxi, and Xinjiang provinces. The increase in China’s demand spiked lithium production to 140 % from 2011-2012 where it produced 49,000 t of LCE domestically and relied on imported spodumene for 70% (Merriman 2012; Jaskula, 2012). Various companies have invested in mining and extraction process in China. In 2012, Galaxy Resources opened a lithium carbonate plant and claimed to produce “1,200 tons of technical grade lithium carbonate and 290 tons of battery grade lithium carbonate” The plant is designed to not only produce large amount of Li carbonate and Li hydroxide but to also become a supplier for the Asia-Pacific region (Galaxy Resources Ltd., 2012a; Jaskula, 2012). China’s 2013 reserves are estimated at 3.5 million ton and resources are estimated at 5.4 million tons (USGS, 2014). The production is reported as 4,000 metric tons (Howard; 2013).

3.7.5 Russia

Russia is another lithium mineral producer along with China, Argentina, Australia, USA, and Chile. Not much information is available in terms of resource capacity of Li in Russia. Howard (2013) has roughly estimated a production capacity of 3,000 metric tons per year and reserves of 1,000,000 metric tons. According to Evans (2008), Russian lithium is stored in the form of pegmatite and is estimated at the same 1 million ton of capacity as reported by Howard (2013). However, the cost effectiveness of
brine operations and difficulty in extraction has forced Russia to import materials to meet their demands.

### 3.7.6 Australia

Most of the lithium produced in Australia is derived from pegmatite. Talison Lithium, the largest producer of low grade iron content in spodumene (Evans, 2008) produced 399, 000 t of Li equivalent to ~ 34% of global supply of lithium just in Australia itself in 2012 (Jaskula, 2012). The company produces Li concentrate in two forms: Chemical grade of 6% lithium oxide content and technical grade of 5%-7.5% lithium oxide content. Chemical grade Li is used for conversion to lithium in lithium batteries and technical grade Li is used in other non-automotive applications such as glass, ceramics etc. According to Talison, almost their entire chemical grade Li concentrate is sold to China because of China’s rising development in electric vehicles; whereas, technical grade Li concentrate is still distributed globally. China is the largest consumer of Talison produced Li concentrate (Wheatley, 2012; Jaskula, 2012). Consequently, Talison has doubled chemical grade production capacity from 315, 000 tons/year to 740, 000 tons/year and is also setting up more plants in other parts of Australia in upcoming years (Talison Lithium, Ltd., 2012a; Jaskula, 2012). Australia has an estimated Li production of 13000 metric ton, reserve of 1000,000 metric ton, and resources of 1,700,000 metric tons (Howard, 2013).

### 3.8 End of Life-Recycling

Recycling of Li-ion batteries recovers useful products, such as CO (cobalt), Nickel (Ni), etc. These batteries are multi-component products that contain basic
materials and some battery grade materials depending upon the recovery process. Currently, there is enough information on only two recycling processes used in the industry and are of value to this report (Gaines, et al., 2011). They are discussed below.

3.8.1 Umicore V’al eas Process

Umicore is a European materials technology company whose one of the core businesses is recycling and refining of precious and other non-ferrous metals from electronic and catalytic applications (Umicore Recycling, 2014). Umicore claims that they excel at recycling such that they are the world’s largest recycler of precious metals. For battery recycling Umicore uses the V’al Eas process. The V’al Eas process “is a single-furnace pyrometallurgical treatment method” which accepts both mixed and separated battery packs (Gaines, et al., 2011). The materials are fed directly into the smelter without preprocessing (Gaines, et al., 2011) to extract slag and molten metals (Vadenbo, 2009). The smelter is fueled by burning cathodes, anodes, and electrolytes (organic components) and carbon is used as a reducing agent for some metals (Gaines, et al., 2011). Two precious metals Co and Ni are recovered from this process minimize energy for primary production by 70%. These metals are then sent to a refinery in Belgium to form Cobalt chloride (CoCl₂) and then sent to Korea where Lithium Cobalt oxide (LiCoO₂), one of the most common cathode materials, is produced for car batteries. Lithium and aluminum from the smelter are used as slag in secondary markets. The waste gases are treated with plasma torch to prevent emission and formation of toxic dioxins or furans (Gaines, et al., 2011). Umicore claims high recovery of 93% for Li-ion batteries where metals constitute 69%, carbon is 10%, and plastics are 15% (Gaines, et al., 2011). Figure 18 provides the flowchart for the V’al Eas process (Vadenbo, 2009).
3.8.2 The Toxco Process

Toxco, Inc. primarily recycles, reuses, and disposes of contaminated material.

Toxco is headquartered in California and has offices in Canada as well (Toxco Materials
Management Center, 2014). Toxco claims to be the only company in the world that can recycle lithium-based batteries of any shape and size (Toxco, Inc., 2014). The company uses a proprietary combination of mechanical and chemical process, but Figure 19 shows the general steps taken in lithium processing, as derived from an Environmental Assessment for proposed lithium recovery in a battery recycling facility (Vadenbo, 2009). Li-ion batteries are first discharged to ensure safety and then circuits and wires are removed for recycling. The battery packs are disassembled and sent through a process of mechanical and chemical treatments to recover Li-ion fluff, copper cobalt, and cobalt filter cake (Gaines, et al., 2011). Fluff mainly consists of plastic and steel that can be used for steel recovery or disposal, depending upon the value of the steel content (Vadenbo, 2009). Lithium is extracted along with the fluff where it is mixed with sodium carbonate (Na₂CO₃) to produce lithium carbonate as a final product (Vadenbo, 2009). Copper cobalt product yields “salable metals” such as copper, cobalt, nickel, aluminum (Gaines, et al., 2011) at separation and recovery during their metallurgical operations (Vadenbo, 2009). Cobalt filter cake is also exposed to metallurgical operations and can be used as coating for appliances (Vadenbo, 2009).
Figure 19: Process Flow Chart for Toxco’s Recycling Process for Lithium-ion Batteries (Cheret, et al., 2007; Vadenbo, 2009)
4. LITHIUM-ION SYSTEMS FRAMEWORK

In this section, a process flow diagram was created after compiling the literature review. The process flow is divided into five sections: Mining/Extraction, Battery production and Assembly, Vehicle Manufacturing, Consumers, and End of Life. Each section details the lithium inflows and outflows throughout the individual stage, and the flows can be seen visually in Figure 20 to Figure 24. A complete process flow model is presented at the end of this section in Figure 25, putting the individual pieces together to provide a ‘big picture’ effect.

4.1 Mining/Extraction

FMC lithium, one of the major producers of lithium in the market today operates a brine facility in Argentina to produce lithium carbonate and lithium hydroxide using an advanced proprietary technology (FMC Lithium, 2014). The high grade lithium such as lithium carbonate/lithium hydroxide produced from this brine operation is then sold to the components of battery making: cathode, anode, and electrolyte. Apart from the extraction of lithium compounds, FMC has also been successful in developing advanced technologies for efficient cathodes (FMC Sustainability Series, 2012). As claimed by FMC, they are not only the innovators in the industry but are also the no.1 supplier of lithium compounds and salts (FMC Lithium Market Review, 2012). According to the Lithium Market Review published by FMC, they are the only company in the world that is a “mine-to-metal producer.”
4.1.1 Resource Extraction

FMC started their brine operations in Argentina in 1997 and currently operates four facilities within the country. One of the facilities, Salar Del Hombre Muerto, is known for “high concentration of lithium” ranging from 200-2,000 ppm and can be processed further to obtain higher ranges (FMC Lithium, 2014). FMC brine technology is proprietary; therefore, it is difficult to determine the exact process. Hence, estimating the energy and resource costs associated with this step in the system will be difficult.

4.1.2 Evaporation

Generally, brine extraction involves pumping brine from aquifers to a series of evaporation ponds designed to collect lithium and associated co-products such as potash. The process is relatively simple and low cost compared to other extraction techniques (Rodinia Lithium, 2014). Lithium goes through several refining and processing steps where it is fed into the processing plant directly from the ore (Western Lithium Corporation, 2012), before it is packaged for shipment. Lithium recovery is the final goal of the process (Western Lithium Corporation, 2012).

4.1.3 Purified or Refined Lithium

Purified or refined lithium in the form of lithium carbonate and potash is shipped either in solid form or as a powder, where it is packaged into 25 kg bags or 11 pound ingots and stored on pallets for international shipment (Canis, 2013). Recovered products are then shipped internationally to battery cell manufacturing companies through a
combination of ship and rail. Salar De Hombre Muerto is located comparatively closer to both rail lines and seaport that makes it is convenient to ship the product internationally.

Figure 21 outlined the process flow of lithium extraction, evaporation, and refining to produce a final lithium compound suitable for shipment and use as battery materials.
Figure 20: Mining/Extraction Portion of the Process Flow Model

- Extraction
  - Brine pumped from Salar de Hombre Muerto

- Evaporation
  - Harvested Potassium Salts
  - Lithium ponds

- Li and Potash Recovery
  - Lithium Carbonate plant
  - Lithium Hydroxide plant
  - Potassium plant

Products
- Lithium Carbonate
- Lithium Hydroxide
- Potash
  Shipped as powder or 11 lbs ingots
4.2 Battery Production and Assembly

Shipped lithium carbonate is then sold to the companies specializing in making the battery cell materials using lithium compound in anode, cathode, and electrolyte powder as seen in Figure 22. These companies produce basic material in any form.

4.2.1 Battery Cell Materials

Battery manufacturing requires combination and collection of cell materials to be incorporated. These basic materials and components such as cathode, cathode material, manganese, copper foil, etc., are manufactured by specific companies specializing in their respective skills. Cell materials and electronics are produced by variety of companies and below is the list of cell material suppliers for Nissan Leaf obtained from multiple sources.

- Cathode Material: Nippon Denko
- Manganese: Nippon Denko: Rebranded as NEC
- Cobalt: Supplier unknown
- Aluminum foil: Nippon Denko
- Anode (Copper foil): Furukawa Electric
- Anode Graphite: Hitachi Chemicals
- Binder: Supplier unknown
- Electrolyte: Ube Industries
- Electrolyte solution: Tomiyama
- Separator: Cellgard/Ube Industries
This list of material suppliers was compiled from data and key facts Rolland Berger presentation slides (2011), and Goldman Sachs Global Tech batteries documents (2009).

4.2.2 Battery Cell Fabrication and Production

The cell material companies supplies the necessary chemicals and components (Canis, 2013) further up in the process to cell fabrication and production companies that specializes in manufacturing actual cells using the desired components.

4.2.3 Battery Final Pack Assembly

After the cells are formed, they are assembled in the batteries by another set of specialized companies, depending upon the final system. It is apparent from Canis (2013) and Lowe’s report (2010) that very few U.S. manufacturers are involved in this stage. Most of the players are concentrated in the Asian market and these companies often export supplies to U.S. manufactures. A list of such players both in U.S. and globally has been produced by Marcy Lowe and is presented in Figure 8.
Figure 21: Battery Production and Assembly Portion of the Process Flow Model
4.3 Vehicle Manufacturing

Nissan and Nippon Electric Company, Limited (NEC), a Japanese multinational electronics corporation entered into a joint venture to form a company called Automotive Energy Supply Corporation also known as AESC to focus on mass developing cells and battery packs. For this reason most of battery cell production and assembly takes place outside U.S. (NEC, 2008). Batteries are assembled by AESC at Japan and shipped to Tennessee, U.S. to be put into the vehicle. But, in 2013, Nissan decided to move production from their Sagamihara plant in Japan to their Smyrna Plant in Tennessee to focus on models in the U.S. (Davies, 2010). This partnership has been very valuable and strategic to Nissan and NEC. NEC’s expertise in cell technology and electrode manufacturing combined with Nissan’s real world automotive application has been extremely helpful in dominating markets in U.S. (NEC, 2008).

4.3.1 Installation

Once the batteries are assembled, they are delivered to the automaker, in this case Nissan’s plant in U.S. (see Figure 22), to be mobilized in the vehicle. These hybrid or electric car batteries have to be critically circuitted into the vehicle unlike the lead acid batteries which are generally dropped in their particular section. Due to this intricate and complicated system of installing batteries in the car, the automakers are very closely intertwined with the whole design and production process. This is another advantage for Nissan and NEC venture which makes them “functionally optimal (Canis, 2013).”
4.3.2 Warehouse Storage

Batteries and cars are assembled at the automaker’s assembly plant. Before the final product is ready to be launched in the market and sold to the consumers, the cars are stored at a warehouse storage which can be at a plant site or at an offsite, closer to the market it is being shipped to.

4.3.3 Dealership

The cars that are ready to be sold to consumers are shipped to dealerships either through trucks, rails, or even ships depending upon the location of warehouse storage. Thereafter, dealership keeps the car on shelf and takes care of any maintenance charges until the car is sold. After the car is sold to the consumers, dealership is no longer responsible for the car and consumer bears all the cost. However, damaged batteries can also be sold or replaced at the dealership.
Figure 22: Vehicle Manufacturing Portion of the Process Flow Diagram

4.4 Consumers

Consumers are the users of the batteries deployed in vehicles. Consumers typically buy new cars directly from dealerships or brokers, based on their demand and goals for the product. Consumers may own the product until the product is no longer
usable, or they may sell that product to other consumers during the product’s useful life. Consumers bear the initial expenses and those that accrue during the ownership period.

Initially, a new car is directly sold to its first owner (Consumer 1), typically at a dealership. Consumer 1 uses the vehicle until that value of the product is lost and he passes the vehicle onto to next consumer, and so on, through Consumer n whereupon the product has reached the end of its useful life (see Figure 23). During the ownership period, the vehicle is also subject to costs associated with wear and tear, regular maintenance, and insurance coverage.

4.4.1 Service Station

Throughout the lifetime of the vehicle, from consumer 1 to consumer n, it is subject to repairs and maintenance at service stations. Battery replacement is one such maintenance expense. Additional services can cause higher strain on the consumers and it could lead them to dispose or discard off the vehicle before it reaches end of life.
Figure 23: Consumer Portion of the Process Flow Diagram
4.5 End of Life

The lifespan of batteries depends upon their application, but when they reach their end of life, their ultimate fate must be determined. Batteries will either be recycled or dumped in the landfill (see Figure 24). Batteries that are sent to landfill are likely to be not recycled due to low economic value; whereas, batteries that are sent to be recycled are likely to recover profitable individual components. Recycling efforts had always been focused on recovering specialty metals such as Co and Ni that have high economic value. However, to reduce the cost of Li-ion batteries it may be necessary for lithium to be recycled and reused. Consequently, manufacturers are exploring different characteristics that will easily fulfill their economic and performance goals (Vadenbo, 2009). Future demand for batteries and technological developments is likely to affect the course of recycling process and feasibility.

4.5.1 Landfill

Cars that end up in landfill are most likely to not be recycled or have a second life where they can still be used alternatively in other forms. Batteries that lack second life use will usually be disposed or discarded in unsustainable way that won’t benefit environment and society. According to the health and safety document prepared by Vimmerstedt, Ring, and Hammel for National Renewable Energy Laboratory (NREL), lithium and lithium compounds as lithium ions are in fact hazardous but less hazardous than metallic lithium (1995). Li-ion battery chemistry such as LFP (lithium iron phosphate), LiCoO$_2$ (lithium cobalt oxide), LiNO$_2$ (lithium nickel oxide), LiMnO$_2$ (lithium manganese oxide) have been subjected to permissible exposure limit (PEL) to
prevent workers from excess exposure. The oxides of nickel, manganese, and cobalt decompose into hazardous products at the end of their lives such that nickel and manganese chemistries can form strong base compounds and cobalt chemistry can “decompose into hazardous cobalt oxide” in form of dust and fumes (Vimmerstedt, Ring, and Hammel, 1995).

4.5.2 Third Party Recycling

Third-party recycling firms become involved at the end of the useful life of the product. Recyclers often extract electronics, wiring, and commercial chemicals from batteries. Although there is limited information about current recycling operations in the literature, the procedures should be similar to those discussed in Section 3.8. Used car batteries can be sold back to dealership, if the manufacturer is intending to incorporate useful parts back into the manufacturing process. Reusing useful parts may reduce the financial burden that comes with rise in cost of harvesting virgin material in the production.

4.5.3 Hazardous Waste Site

Most amount of waste is generated at the vehicle’s end of life. This waste primarily comes from the used car batteries. Chemicals used in battery production can release hazardous or toxic waste, if not disposed of properly, at the end of their use. Limits and bans have been introduced in various countries to minimize the risk of exposure from these harmful chemicals. Moreover, some car companies have invested in ‘take-back’ programs to ensure safety and risk management for their vehicles at the end of their lives.
Figure 24: End of life/Recycling Portion of the Process Flow Diagram
Each framework component discussed above is now shown as a part of a complete flow model for Nissan Leaf in Figure 25. The arrows between the components show clear links with each other. The products obtained from extraction and processing can be sold either to the companies that develop cell materials for batteries or the companies that use those materials to build cell components. Battery packs are formed by assembling all the electronics and components and are shipped to installation plant, in this case, Tennessee, US, to be put into the vehicle. After the batteries are installed in vehicles and are ready to be sent at dealerships, they are sold to consumers. These batteries spend their lifecycle in-use, in repairs, services, and maintenance passed down to different consumers. At the end of their useful lives, these batteries are either recycled by Third-party recycling companies or they are dumped in landfills where they are not recycled and pollute the ecosystem. When battery materials are recycled and recovered, they can be used in secondary markets, such as cement, or they can be sold back to cell manufacturing companies to be used in the battery parts. The reusing of valuable materials in the new product eases the financial burden on automakers and consumers, and resource burden on the natural environment.
Figure 25: A Complete Process Flow Model
5. A PROPOSED LITHIUM LIFECYCLE ASSESSMENT MODEL

Now that a the process flow for lithium has been explored and a systems framework has been defined in Chapter 4, the next logical step is the creation of a lifecycle energy model based upon the systems framework. This chapter outlines the procedures for creating an Excel spreadsheet model, wherein each element of the lithium process flow and systems framework is defined within a series of worksheet ‘modules.’ Each worksheet will take user inputs, such as tons of ore mined, type of construction equipment used, and kilometers traveled from quarry to processing plant, and provide an estimate of the energy consumption and emissions expected to result from the process component. The content of the boxes in the process flow diagram will be represented by a series of calculations in the worksheets, and individual cells in the worksheets will be used to provide assumptions and data values required by the calculations. At each step in the flow diagram within the worksheets, user input variables determine output parameters that feed into the next system element. A user interface page will allow the user to specify the processes that will be employed and various assumptions made in the model. For example, the user will be able to use a drop-down menu to select the locations at which Lithium will be mined and processed and the types of mining and transportation equipment that will be employed in each step. When complete, the user will also be able to essentially work backwards through the process flow and spreadsheet calculations, by specifying the number of Li-ion batteries required. The assumptions within each module will then provide an estimate of the amount of ore to be quarried (based upon ore richness, processing plant location, process type, process efficiency, equipment characteristics at each stage, etc.) and the energy consumed in each step. Each module is
linked to the subsequent module via the transportation network, requiring that a transportation modeling network track fuel consumption, time, and cost of flow of lithium battery product components as they travel step-to-step from quarry to final disposal site. This model will be an open access model for anyone to use so that data and assumptions can be improved over time and used in modeling efforts.

5.1 Resource Extraction Module

The first module, known as the ‘Resource Extraction Module’, will consists of processes involved in obtaining lithium through extraction or mining, based upon the source of the lithium. Different lithium process flows sub-elements are available within this module, depending upon whether the lithium is obtained through physical or chemical processes. Lithium is mainly found in brines and hard rocks and in low amounts in clay deposits and physical or chemical processes would be modeled accordingly. Brine sources are further classified as continental, geothermal, and oilfieds and allow users to make a selection. Similarly, hard rock is classified into a single type, known as pegmatite, which contains different minerals such as spodumene, hectorite, jadarite, etc. The user will be able to specify the source of the ore and the equipment involved in mining and processing as model assumptions.

5.1.1 Brines

Users focusing on brines process flow will have the opportunity to input the location of the brine, which is necessary to establish the transportation network associated with shipment of the raw materials. Brine basin characteristics such as lithium grade (weight%), magnesium to lithium ratio, evaporation rate (meter/year), elevation
(km), surface area of the brine (km²), porosity (%), depth (meter), and density (g/cm³) will be provided as they are unique to the location of lithium deposit (Mohr, et al., 2012).

For hard rock mineral sources, users will also input location and associated parameters embedded in the model will provide mining characteristics, such as type of mineral, lithium grade (%), mine type (open cut, or room and pillar), and iron content of the raw ore (%) (Mohr, et al., 2012).

5.1.2 Mining and Processing Operation

A significant amount of energy and water are used in the extraction of minerals, through washing, separating, and secondary use in recovering other minerals. This water generally comes from groundwater, but can also come from surface water. Water consumption (gallons/ton of resource produced) and energy use can be estimated as a function of tons of ore processed for various operations through a case-study analysis across various processing plants.

Labor cost ($/hr) and capital costs ($/year) will also be estimated per ton of ore processed, by mining location. This includes labor crew, hourly wages, and capital consisting of fixed and financial capital, some of which user will be able to input. Fixed capital costs include equipment used in crushing and grinding of rocks, and at processing plant, etc. that helps in producing the product. Financial capital includes assets and liabilities associated with the setting up and operating the plant.

Extraction and delivery of valuable rare metals (such as cobalt and nickel required for the manufacturing of cathode in the battery) can put a strain on the process flow if
these rare metals are in short supply. Production capacity (ton/year) and content of natural resources will be calculated.

To calculate energy consumption (MJ/ton and kwh) during mineral production and processing from electricity, users can input electricity use and mix as well (electricity from coal, natural gas, nuclear, etc.) and power plant efficiency.

5.1.2.1 Machinery and Equipment

Mining is an energy intensive process and energy consumption occurs in extraction operations such as blasting, digging, and drilling using heavy machinery. In mineral processing, operations such as crushing, grinding, and separation consume energy and produce emissions. Chemical and physical processes (brine and hard-rock, respectively) use heavy equipment and industrial machinery. In this section of the model, users will be able to select the list of equipment and machinery for each process, as well as fuel type and other parameters, and the model will estimate energy consumptions, emissions, and capital costs per ton of ore processed.

5.1.2.2 Products

From the inputs made in the model, the brine process flow yields lithium chloride and lithium carbonate as products and potash as a by-product; whereas the minerals process flow yields lithium concentrates, lithium hydroxide, and lithium carbonate, along with by-products. Inputs and outputs from both brine and hard-rock are provided in Table 3 below.
Table 3: Input and Output Quantities from Brine and Hard-Rock Process

<table>
<thead>
<tr>
<th>Process</th>
<th>Input</th>
<th>Output</th>
</tr>
</thead>
<tbody>
<tr>
<td>Brine</td>
<td>Location, type of brine, type of equipment, labor operating hours and</td>
<td>Water consumption, energy consumption, labor and capital cost, lithium</td>
</tr>
<tr>
<td></td>
<td>wages, equipment operating hours, fuel type, electricity use and mix</td>
<td>chloride, lithium carbonate, potash</td>
</tr>
<tr>
<td>Hard-Rock</td>
<td>Location, Type of mineral, type of machinery, labor operating hours</td>
<td>Water consumption, energy consumption, labor and capital cost, lithium</td>
</tr>
<tr>
<td></td>
<td>and wages, machinery operating hours, fuel type, electricity use and</td>
<td>concentrate, lithium hydroxide, lithium carbonate, potash</td>
</tr>
<tr>
<td></td>
<td>mix</td>
<td></td>
</tr>
</tbody>
</table>

Users will have the option to choose from a drop down menu or directly input quantitative data into applicable cells. For example, location of the brine, type of the brine, type of the equipment, fuel type, electricity mix and use will be available as drop down menu parameters. Labor operating hours, labor cost, and equipment operating hours will be calculated based upon ore input (or lithium output, if the implementation starts with battery demand and works backward). The outputs will be in tons or kg of material produced which can be used in T&D to model the transport of raw materials.

5.1.3 Transportation

Material handling, distribution, and shipment will be accounted for under ‘Transportation Mode Network’ section which will be modeled with using a GIS (Geographic Information System) and the GREET model.
The GREET (Greenhouse gases, Regulated Emissions, and Energy use in Transportation), a full life cycle model developed by Argonne National Lab will be a useful tool in estimating air emissions and energy impacts throughout the life cycle of Lithium-ion batteries. GREET is divided in two parts: GREET 1 series evaluates the fuel cycle from well to wheels and GREET 2 series evaluates the vehicle cycle including pump to wheels, vehicle disposal, and material recovery.

GIS can help map the domestic rail and road layers to establish the transportation logistics chain to minimize time and cost. A variety of embedded transport chain options will be available for users to specify in the model within each module. For example, processed ore can be moved by truck, truck-rail-truck, container ships, etc. This portion of the model will be complex but will provide users with a lot of flexibility. Furthermore, there are associated excise and custom tax duties with the shipment that will be taken into consideration, depending upon the weight of the product and the destination where it will be delivered.

GREET has a built in transportation and distribution (T&D) module to account for transporting raw materials to and from the site. Trip distance, amount of material transported, and fuel type are specified based on the mode of transportation: truck, tanker/barge, and rail used during the process to determine the energy consumption (Btu/ton of material transported), energy intensity (Btu/ton-mile), total emissions (grams/ton of material transported) and urban emissions (grams/ton of material transported) by mode. Fuel cost and material transport costs are also accounted for in the process. The GREET T&D modeling elements will be employed every time material is transported from one module to another.
5.2 Battery Production and Assembly Module

In the ‘Battery Production and Assembly Module’, all the material and components required to form a battery pack will be modeled. The inputs in this module are the outputs from the previous module 5.1. Lithium products and co-products are sent as inputs to the battery companies specializing in developing cell materials and outputs from those companies will be inputs to cell fabrication and production companies. A battery pack is assembled from the outputs of fabrication and production companies.

This module will consist of a detailed list of potential key companies involved in developing various components of the battery based on Figure 8: Global Value Chain of Li-ion Batteries for Vehicles, with Major Global Players and U.S. Players with Current and Planned Facilities (not exhaustive) (Lowe, et al., 2010). Companies will be classified based on their expertise in developing a battery component and will be sorted into categories such as: 1) making key materials for battery, such as Chemetall which develops lithium compounds; 2) building cell components and electronics, such as A123 Systems which develop cathodes; and 3) integrating battery packs, such as NEC. Users will be able to select the key battery players and their location to compute for transportation energy use and cost.

The GREET 2 model can be used to estimate energy use and emissions for battery material processing and fabrication, where users can input percent share of composition of battery material, percent share of key material composition in specific fluids (Burnham, et al., 2006), and share of electricity use and mix in the production of battery materials. Water used in dissolution, filtration and, separation of material during
production is also accounted for. Assembled battery packs are sent as inputs to the vehicle manufacturing process in the next module, producing vehicles as outputs for consumers. Energy consumption, cost, and emissions from transporting products to installation plant is modeled using T&D in GREET as well and can be seen in Table 4.

<table>
<thead>
<tr>
<th>Process</th>
<th>Input</th>
<th>Output</th>
</tr>
</thead>
<tbody>
<tr>
<td>Battery manufacturing</td>
<td>Lithium compounds</td>
<td>Cell material</td>
</tr>
<tr>
<td></td>
<td>Cell material</td>
<td>Battery pack</td>
</tr>
<tr>
<td></td>
<td>Key players, electricity use and mix, battery material composition</td>
<td>Energy consumption at plant and transport of materials, cost, water use</td>
</tr>
</tbody>
</table>

Lithium compounds (tons or kg) obtained from the ‘Resource Extraction Module’ will act as input in this module along with other battery material for production of cell material and number of cells required to make a battery pack will be the input to estimate the battery pack production. Electricity use and mix will also be computed throughout for the production and manufacturing processes.

5.3 **Vehicle Manufacturing Module**

In this module, GREET 2 is used to estimate energy use from vehicle production and vehicle assembly. Material composition and production processes need to be identified and are provided in GREET. Users can modify and input composition of material used in vehicle production, weight of the materials, weight of the vehicle, battery
specific power and specific energy, and electricity used at installation plant and operating efficiency of the plant.

Manufactured vehicles are sent to warehouse or yard storage until they are ready to be sold at dealership. This element will again be modeled again using GREET T&D. Additional energy consumption and cost associated with storage of components at the warehouse versus at the dealership will be accounted for via electricity use and electricity mix.

Energy consumption associated with the delivery of vehicles to dealerships via freight or car carrier trailers (depending upon the location) will also be modeled using T&D in GREET. When the cars reach the dealership, energy consumption and cost will also be modeled for time spent at dealership, until purchase by consumers. Users can include vehicle sales costs for the vehicle in the modeling process if desired, along with any subsidies or benefits provided by the government or by the dealership as seen in Table 5.
Table 5: Input and Output Quantities from Vehicle Manufacturing Module

<table>
<thead>
<tr>
<th>Process</th>
<th>Input</th>
<th>Output</th>
</tr>
</thead>
<tbody>
<tr>
<td>Installation</td>
<td>Vehicle material composition, material weight, battery weight, battery specific power and energy, electricity use and mix, plant operating efficiency</td>
<td>Energy use</td>
</tr>
<tr>
<td>Storage</td>
<td>Electricity use and mix, maintenance</td>
<td>Energy consumption, cost,</td>
</tr>
<tr>
<td>Delivery</td>
<td>Actual vehicle cost, additional subsidies/benefits, time spent at dealership</td>
<td>Energy consumption, cost of not being sold</td>
</tr>
</tbody>
</table>

5.4 Consumer Module

The battery in-use will be modeled in fourth module, known as ‘Consumer Module’. Vehicles transported from dealership to consumer will generate emissions and consume energy; therefore, T&D parameter will be used to estimate those calculations. Users will have the option to input the number of consumers that vehicle was passed along to in its lifetime, starting from the original owner of the car as ‘consumer 1’, to the second consumer, and so on to consumer n; where n represents the total number of owners in the lifetime of the vehicle. The model will be equipped to calculate the depreciated value of the original car as it passed down and repaired throughout car’s useful life, based on the final price of the vehicle obtained after modeling accidents, damages incurred, battery repair and replacement costs, and other servicing costs, all of which comes from user input. The calculated final monetary value of the battery, as well
as the vehicle, will be compared to the original price of both to estimate and analyze the overall financial gain or loss the vehicle had subjected to in its lifetime. In-use emissions can also be calculated based on miles travelled, efficiency of the battery, lifetime of the battery, and fuel intake. Input and output from this module are shown below in Table 6.

<table>
<thead>
<tr>
<th>Process</th>
<th>Input</th>
<th>Output</th>
</tr>
</thead>
<tbody>
<tr>
<td>In-use</td>
<td>Number of consumers, vehicles per mile travelled, battery efficiency, lifetime of the battery, fuel intake, number of vehicle accidents, vehicle and battery repairs maintenance</td>
<td>Cost of the car after use, energy consumption, in-use emissions</td>
</tr>
</tbody>
</table>

5.5 End of Life Module

The end of life of the batteries when they are no longer in use is modeled in fifth module as ‘End of Life Module’. Vehicles after their useful lives are sent to junkyard where they are either sent to landfill, third party recycling or to hazardous waste site and have associated transportation cost, emissions, and energy consumption from T&D in GREET.

5.5.1 Landfill

Batteries (and therefore lithium) that are transported to landfills (i.e. are not recycled) have environmental impacts and costs associated with landfill operations. Energy and cost estimated per ton will be delivered through supplemental research.
5.5.2 Third Party Recycling

Batteries that are sent to third part recycling companies, such as Umicore or Toxco, are reprocessed to harvest lithium and rare metals. Because recycling processes are still proprietary, the model can incorporate a general process flow consisting of hydrometallurgical and pyrometallurgical treatment processes and energy cost estimates for these steps, but some uncertainty is inherent. Currently, specialty metals such as cobalt and nickel are mainly recycled and model will follow the same current trend. The model will also track the prices of these valuable recycled metals and their second use after they are recycled. Also, lithium recovered from the treatment process will be modeled for cost and second life, similarly to cobalt and nickel. These batteries could be sold back to dealership and will be modeled based on the before and after cost and condition of the battery. The model can definitely be expanded based on the future recycling needs and demands.

5.5.3 Hazardous Waste Processes

Batteries that are likely to end up in hazardous waste site may generate harmful air emissions and can be modeled for their environmental impact based on the type of emissions and concentration of pollutants from user input, and effect on environment and human health from the model itself as reported in Table 7.
### Table 7: Input and Output Quantities from End of Life Module

<table>
<thead>
<tr>
<th>Process</th>
<th>Input</th>
<th>Output</th>
</tr>
</thead>
<tbody>
<tr>
<td>Landfill</td>
<td>Number of batteries in landfill, pollutant type and quantity</td>
<td>energy use, emissions, cost</td>
</tr>
<tr>
<td>Recycling</td>
<td>Reused and recovered material composition, material’s second life,</td>
<td>Price of recycled/recovered material, recycling</td>
</tr>
<tr>
<td></td>
<td>electricity use and mix at plant</td>
<td>efficiency, type and quantity of recycled material,</td>
</tr>
<tr>
<td></td>
<td></td>
<td>energy intensity and use, water use</td>
</tr>
<tr>
<td>Hazardous waste site</td>
<td>Pollutant type, concentration of pollutants</td>
<td>Emissions, environmental and human health impact</td>
</tr>
</tbody>
</table>

Once all of the inputs are entered into the model, outputs provide total energy and material consumption from extraction to disposal of Lithium-ion batteries. Users will be able to analyze the feasibility of various process elements and explore ways to minimize the impact of the outputs. The proposed modeling system will help policymakers and stakeholders in understanding the material supply and demand and to assess the economic feasibility associated with recycling and development of regulations associated with Lithium-ion battery technology.
6. CONCLUSION

Lithium-ion batteries are becoming a dominant battery chemistry to power the transportation sector because of their technically sound characteristics and their application in electric and hybrid cars. As these batteries become more promising with passing time and extensive research, the long term availability of lithium used in manufacturing of Li-ion batteries might become a source of concern. Various government and industry experts such as USGS, Keith Evans, William Tahil, etc. have claimed vast reserves of lithium globally all with varied reporting. The discrepancy in the data and lack of accuracy in reported data is likely to cause supply demand constraints in the future and hamper the progress of this technology.

The objective of this thesis is to understand the Lithium-ion battery system and develop a process flow diagram for lithium resources that can serve as a basis for developing a lifecycle model to predict the costs, energy consumption, and other resource consumption associated with the use of Li-ion batteries. Ultimately, such a model could be used for predicting Li-ion battery recycling feasibility. The process flow diagram in the form of a systems framework tracks the flow of lithium from extraction, to battery production, to end of life. When coupled with estimates of Li-ion battery demand, the model will be useful in assessing whether there is enough lithium to power the future global demand and at what point it makes sense to implement Li-ion battery recycling.

As a result, a lifecycle energy model will be created based upon the systems framework. The model will be useful in quantifying the lithium material flow and identifying key energy inputs and outputs on Lithium-ion batteries throughout their life.
A proposed model is devised in this thesis to show how the framework will be modeled with the help of Excel and an already established GREET model. There will be five modules in the model, each module corresponding to each segment of the framework. Throughout these modules energy consumption and material consumption will be estimated at each step. Shipment of raw materials and final products locally or internationally between modules along with fuel consumption can be modeled using GIS and GREET model under ‘Transportation Mode Network’.

First module, known as’ Resource Extraction Module’, allows users to input parameters pertaining to brine location, brine type, equipment type, labor hours, fuel type, electricity use and mix, etc. and calculates lithium compounds and co products as raw material outputs along with water use, labor and capital costs, energy consumption during extraction process and during transporting raw materials. The lithium compounds serve as an input to next module called ‘Battery Production and Assembly Module’ to produce a battery pack as an output. Energy consumption is calculated from user input electricity use and mix and from transport of materials. Material consumption is calculated using user input key material composition in battery manufacturing. After the battery packs are formed, third module, known as ‘Vehicle Manufacturing Module’ will allow users to estimate energy consumption, material consumption, and cost at each stage: installation, storage, and delivery by inputting variables such as material composition, battery weight, electricity use, plant operating efficiency, etc. The battery in-use will be modeled in fourth module, known as ‘Consumer Module’. In this module users can input vehicles per mile travelled, battery efficiency, lifetime of the battery, fuel intake, number of accidents, battery repairs maintenance, etc. to obtain the depreciated
cost of batteries, emissions, and energy consumption. In module five, ‘End of Life Module’, the fate of Li-ion batteries will be explored, whether they are recycled, they end up in landfill or they are sent to hazardous waste site. If the batteries are recycled by third party recycling companies, users can calculate the price and quantity of recycled or recovered material, energy consumptions, and emissions throughout the process. If the batteries are sent to landfill where they are not recycled, users can determine the cost of not recycling the spent batteries and emissions that are generated in the environment. Batteries that are sent to hazardous waste site can be estimate for their emissions. Second use of recycled and recovered materials will also be incorporated into the model. Finally, the model will provide the total consumptions and emissions from all the inputs in the model for users to analyze the feasibility of this new technology.

Even though Li-ion batteries are a very promising technology, the researchers should focus not only on battery production, but also on fate of these batteries at the end of life. Unless a recycling system is in place, when batteries reach the end of their useful lives, they will end up in landfills given that the lithium compounds are not classified as hazardous waste. Currently, there are only a few companies involved in recycling of specialty metals, such as cobalt and nickel. Further research is needed to explore the costs and benefits of lithium recycling. Recycling battery materials and components may reduce pressure on natural resources by requiring the extraction of less virgin material, decreasing manufacturing costs by incorporating recycled materials, and reducing overall energy consumption associated with production. As a result, this model could prove useful as a tool for policymakers to implement and manage the recycling infrastructure.
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