

Dendrite formation in electrochemical energy storage systems

Samson Temitayo Olatunde ^{1,*} and Peter Etinosa Igbidun-Uwuigbe ²

¹ *Chemical Engineering Department, Obafemi Awolowo University, Ile-Ife, Osun State, Nigeria*

² *Department of Chemical Engineering, Federal University of Technology, Owerri, Nigeria.*

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Abstract

Dendrites are metal microstructures that may form on the lithium battery's negative electrode during charging. When the anode's absorption capacity is exceeded and excess lithium ions accumulate on its surface, lithium dendrites form. In the field of metallurgy, dendrites are tree-like structures formed by crystals when liquid metal cools and solidifies. The crystals develop faster in the directions that are most beneficial from an energy standpoint, giving the structure its characteristic tree-like shape. Consequently, this dendritic growth has major effects on the materials' properties. The study gives an explanatory introduction of what dendrites are in batteries and innovative ways to reduce their consequent effect on storage systems.

Keywords: Dendrite; Formation; Electrochemical; Batteries

1. Introduction

In the field of metallurgy, a dendrite is a tree-like pattern of crystals that forms during the solidification of liquid metal due to the rapid development along energetically favorable crystallographic orientations. Considerable effects on the material's characteristics result from this dendrite expansion. Dendrites may develop in both unary (single-component) and multi-component systems. Undercooling, also known as supercooling, the liquid (the molten substance) below the freezing point of the solid is necessary. First, the undercooled melt develops a solid sphere at its center [1]. The stability of the sphere's morphology and the uniformity of its shape degrade as its size increases. To some extent, the solid form already shows the crystal's preferred development orientations. Anisotropy in the surface energy of the solid-liquid interface or the relative ease with which atoms attach to the interface along different crystallographic planes, or both, may be to blame for this growth direction (see hopper crystal for an example of the latter) [2]. Interface attachment kinetics are typically quite slow in metallic systems (for exceptions, see dendrite(crystal)). The solid then strives to reduce the area of the surfaces with the greatest surface energy, which is what happens in metallic systems.

As a result, the growing dendrite has an increasingly pointed tip. The dendrite may have a faceted shape if the anisotropy is sufficiently great. The equilibrium between surface energy and temperature gradient (which drives heat and solvent diffusion) in the liquid at the interface determines the microstructural length scale. To put it another way, the process becomes exothermic as more and more atoms lose their kinetic energy during solidification. In the case of a pure material, the solid-liquid interface allows latent heat to be dissipated, maintaining a consistent temperature during the melting and subsequent solidification processes [3-4]. The pace at which this latent heat may be dissipated will determine the rate of development of the resulting crystalline material. Dendrites formed from a molten material that has been allowed to cool too slowly may be seen as parabolic needle-shaped crystals that expand without deforming their original form. The grain size is determined by nucleation and growth, but in columnar development, the main spacing is determined by competition between neighboring dendrites. Nucleation of new crystals is reduced compared

* Corresponding author: Samson Temitayo Olatunde
Chemical Engineering Department, Obafemi Awolowo University, Ile-Ife, Osun State, Nigeria.

to substantial undercooling if the melt is cooled slowly. Big dendrites are the final outcome of dendrite development. In contrast, a rise in the number of nuclei and a subsequent decrease in dendrite size come from a quick cooling cycle with substantial undercooling (and often leads to small grains) [4-6].

The product's ductility increases as its dendrite size decreases. Welding is an example of an application where dendrite development and its subsequent effects on material characteristics may be seen. Etching a polished specimen may reveal the dendrites, which are also frequent in cast goods [7]. The deeper the dendrites grow into the liquid metal; the more heat they are able to extract. Overheating will cause them to melt again. Dendrite recoalescence refers to this process. Dendrites often develop when a system is not in balance [7-8]. Directional solidification uses dendrite growth in gas turbine engine blades, which are heated and pressed very hard along the axis of rotation. Grain boundaries become more fragile than grains themselves at elevated temperatures. Grain boundaries are positioned parallel to dendrites to reduce the impact on attributes. As the nickel-based alloy (MAR M-200) solidified, tungsten accumulated in the dendrites. As a result, the casting outperforms its conventionally cast counterpart in terms of strength and creep resistance along the length of the blades [9].

The present push to boost the energy density of rechargeable lithium batteries has sparked a renewed interest in the chemistry of electrodeposition at lithium metal electrodes. The battery life is shortened and safety is jeopardized due to the nucleation and proliferation of protrusions during charging. Since several non-dendrite morphologies have been recorded in the literature, we advise not to use the term "dendrite," which strictly speaking, requires a branching structure. Electrodeposition and cycling tests with a wide range of electrolytes. Whether the electrolyte is a liquid or a polymer, adding the right salt makes sure that lithium ions are present. However, lithium ions are not only present in but essential to the atomic structure of inorganic solid electrolytes. When electricity flows through a liquid or polymer electrolyte, the salt content in the electrolyte changes [10]. Contrarily, when current is conducted via inorganic solid electrolytes, similar concentration gradients do not exist. When the anion is covalently bonded to the polymer chain, the polymer electrolyte may theoretically function as a single ion conductor [11].

The dendrite morphologies formed in polymeric single-ion conductors are not as well understood as those obtained in polymer electrolytes with added salt [12]. That is why there is a focusing on salt-added liquid and polymer electrolyte systems here. Our goal is to determine what factors govern lithium electrodeposition in different media, including liquid electrolytes, polymer electrolytes, and ceramic electrolytes. Many factors, including current density, salt concentration where lithium is plated, tip radius of the protrusion, temperature, pressure, solid electrolyte interphase (SEI), ion transport, and mechanical properties of the electrolyte [13-15], all play a role in determining the morphology of electrodeposited lithium. Even though we have been discussing the anode and the electrolyte, it is well-known that spontaneous reactions between lithium metal and all known electrolytes result in the formation of an SEI layer [16].

The ever-increasing demand for energy systems combined with the precarious nature of fossil fuel supply has given rise to an urgent need for high-energy storage devices [17]. Recently, the technologies of batteries and fuel cells have ushered in a time of revolutionary transformation across a number of different sectors, including those of power tools and electric cars [18]. The use of lithium-ion batteries, which are characterized by a long lifespan, an exceptional design, and an affordable price point, has become the norm in both consumer and industrial electronics. In addition, the growing need for vehicle feedstock and alternative combustibles has necessitated an abundance of technological advancements and optimizations in the field of battery chemistry [19]. The need for a battery system with a high storage capacity has led to the adoption of large-throughput electrode materials [20]. The performance of these materials will be a decisive element in the success of future improvements, so it is important that they be continuously improved.

In addition, the cathode material and the anode material have an equal impact on the overall performance of high-energy-density battery systems [21-22]. Anodes are the materials that store electrons. It's possible that alkali metal anodes should be taken into account for these sorts of high-powered systems. When compared to those of lead-acid batteries. The valence electron, denoted by the symbol ns^1 , of the alkali metals Li, Na, and K may be readily removed, which leads to the production of a great number of cations. These metals have a high degree of reactivity. These soft metals have low melting points because they have low ionization energies, which makes them very reactive and contributes to their low melting points (Li: 181 C, Na: 98 C, and K: 63 C) [23,24]. With the exception of hydrogen, all of the alkali metals may be found in their solid state at ambient temperature. Because the metal salt ions are relatively larger than the other ions in the same period, dissolving them in aqueous electrolytes results in a drop in the charge density [25]. This causes the alkali ions to become separated from their counter anion, which is due to the fact that the metal salt ions are relatively bigger than the other ions in the same period.

Various electrode materials are used in commercial Li batteries. We can see the evolution of their individual energy use and stamina over time. With a cumulative manufacturing rate more than 100 GWh yr⁻¹, LIBs are already outpacing lead-

acid batteries in the energy industry. Within a few years, this pace of output might reach hundreds of GWh [26]. Various electrode materials are used in commercial Li-ion batteries. We can see the evolution of their individual energy use and stamina over time. With a cumulative manufacturing rate of more than 100 GWh yr⁻¹, LIBs are already outpacing lead-acid batteries in the energy industry. Within a few years, this pace of output might reach hundreds of GWh [26].

2. Experimental Causes of Dendrite development in Batteries

It is common knowledge that lithium-ion batteries, the most common type of portable rechargeable battery, power a wide variety of modern electronics. The increased energy density, increased power density, and the absence of a memory effect have all contributed to their rapid spread. You won't find these benefits in any other rechargeable battery. As a result, Li-ion batteries have seen an increase in their use across a wide range of industries since their commercial introduction in the 1990s. Graphite is typically used for the anode and lithium transition metal oxide is used for the cathode in a Li-ion battery. The term "Li-ion battery" is derived from the fact that lithium ions travel between two electrodes to store and release energy under typical operating circumstances. However, there have been numerous reports of Li-ion battery safety incidents, leading to widespread alarm [25].

One of the reported causes of battery failure was internal short circuits brought on by lithium dendrite formation. Lithium dendrite development, also known as lithium plating, is a potentially dangerous side effect of using a lithium-ion battery under atypical settings, such as overcharging or charging at a low temperature. Metallic microstructures called lithium dendrites form on the negative electrode during charging. Dendrites of lithium form when an excess of lithium ions on the anode surface are not quickly absorbed. This is because they may produce electrical shorts, which can result in system failure and even fires. Some of the Samsung Galaxy Note 7 batteries caught fire in 2016, and the ensuing research found that lithium dendrites were to blame for the internal short circuits. Lithium dendrite formation might also decrease capacity. Lithium dendrites react with the electrolyte, breaking it down and setting off the internal loss of lithium. Together with the slow development of lithium dendrites comes a cumulative loss of capacity. Knowing how lithium dendrites form is helpful for making safer batteries. However, the conventional Li-ion battery is not transparent, making it impossible to see lithium dendrite formation [25-26].

A symmetrical lithium cell was charged at a constant current to measure the pace at which dendrites grew. Both the positive and negative electrodes were constructed from lithium metal. Multiple factors, including current density, temperature, electrolyte, and electrolyte convection, affect the development of lithium dendrites. The dynamics of the electrolyte are determined by these parameters. The current step included the application of a steady current until an internal short circuit was produced by a lithium dendrite. This part of the study aimed to see whether there was a correlation between the amount of current applied and how quickly dendrites expanded. Dendrite production was detected at different current densities using an in situ monitoring technique.

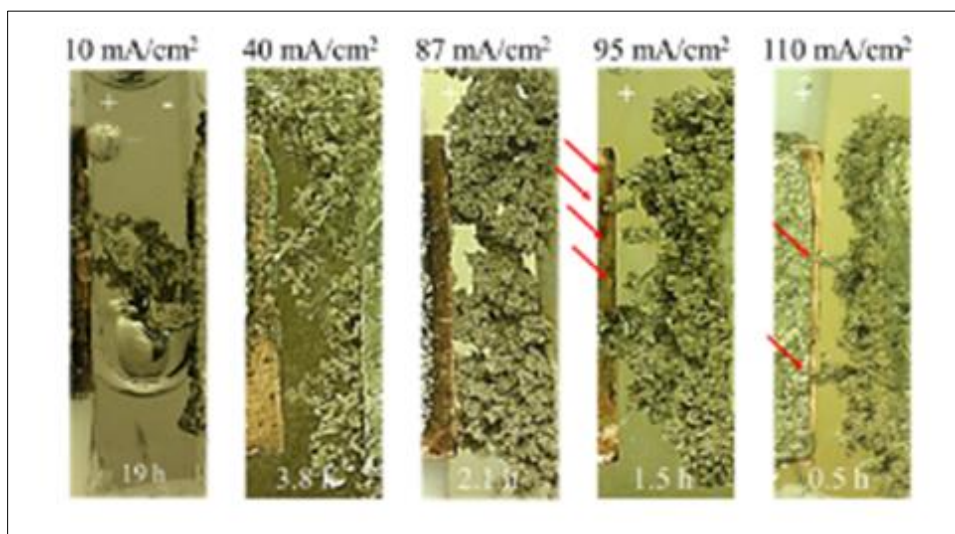


Figure 1 Dendrite development at varied current densities [26]

In Fig. 2, above, an experimental study conducted by [26] examined the dendrite morphology at different current densities. Current densities increased from 10 to 110 mA/cm². The timestamps on the images show how long into the

test the internal short-circuiting lasted. Lithium dendrites formed in this experiment with the negative electrode (right) and progressed toward the positive electrode. When the current density is raised, the lithium dendrite growth rate also increases since there is less time between short circuits. Furthermore, the lithium dendrite's shape changed. With increasing current, the negative electrode edge developed a protrusion (seen by the red arrows).

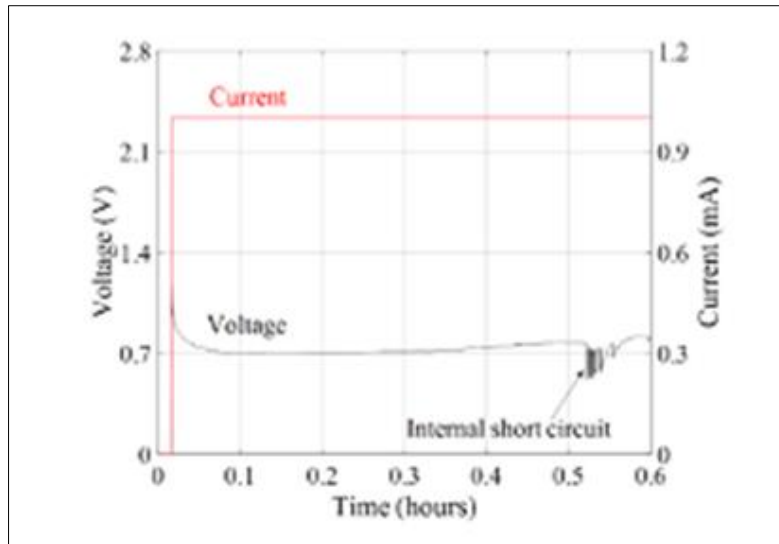


Figure 2 Typical current and voltage profile (110 mA/cm²) during lithium dendrite development test [26]

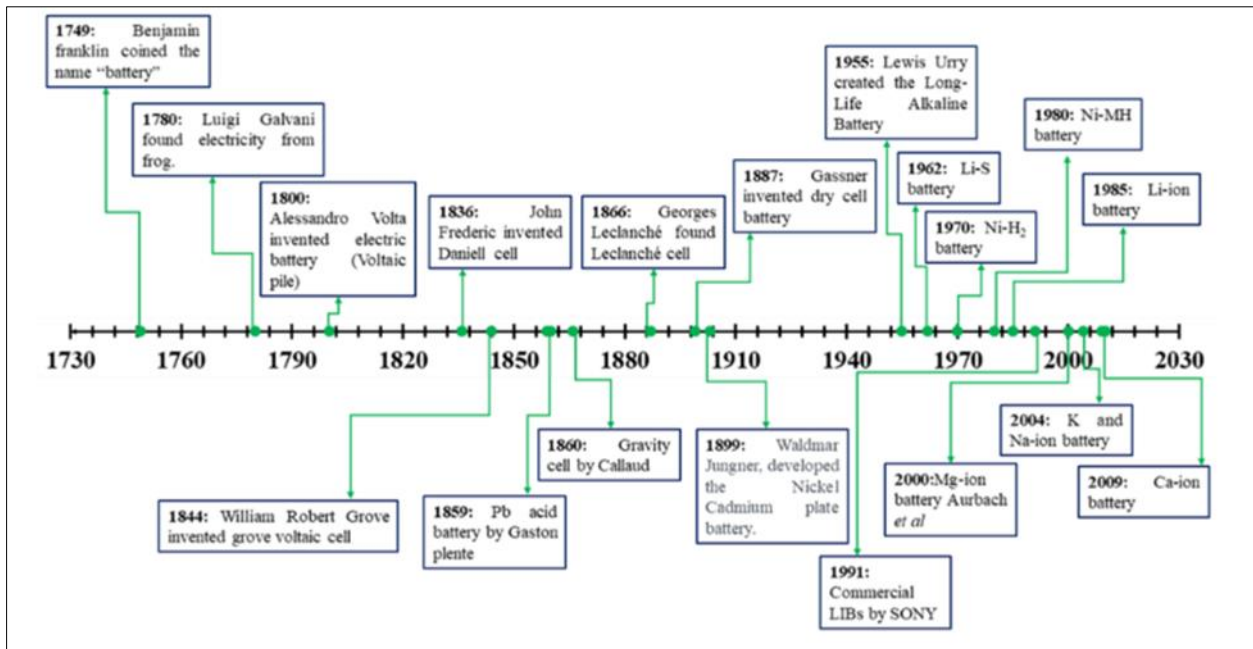


Figure 3 Evolution of batteries from 1730 through 2020

The voltage and current profile of a typical lithium dendrite development test is shown in Figure 2. Current and voltage under 110 mA/cm² are shown in this profile, which is representative of the fifth sample. Here, the current was measured to be steady and under control. Until the voltage dropped suddenly at 0.5 h, it had hovered about 0.7 V, suggesting that the lithium dendrite had triggered an internal short circuit. Dendrite shape may be affected by the current density, and when it grew, the average dendrite development rate did as well. Dendrites' growth shape changed from flat mossy to sharp needle-like when the current density exceeded 87 mA/cm². Lithium dendrite development may be monitored, and the elements that affect growth can be accounted for, to reduce the number of accidents and

malfunctions in consumer electronics. By analyzing the dendrite development rate collected at different current densities, the dendrite's length may be estimated, making it possible to detect and prevent short circuits at an early stage.

3. Material Considerations for Batteries and a historical perspective

Benjamin Franklin coined the term "battery" in 1749 to characterize connected capacitors, and the term has stuck ever since. Bioelectricity was first developed by Luigi Galvani in 1782, using frog cadavers as a source of inspiration. A variety of batteries were invented in later years by scientists and chemists. Advances in machine dynamics (e.g., random-access memory) and the Goodenough-Kanamori principles for determining the magnetic super exchange in Li-ion batteries have contributed to the development of cutting-edge technology throughout the last decade.

The introduction of the Li-ion battery in the 1990s sparked a revolution in mobile and consumer devices. Cutting-edge LIBs, in particular, with an average energy density of 250 Wh Kg⁻¹, sparked a paradigm shift toward gasoline alternatives [27]. Its energy density is undoubtedly too low to fully replace gasoline, however. As a result, there are extensive initiatives ongoing to create high-energy batteries that transcend lithium-ion batteries [28]. Insertion chemicals and carbon-based electrolytes are widely used in today's commercial LIB technologies [29-30].

Additionally, current collector additives are used to prevent anodic and cathodic corrosion. To avoid thermal runaway at high temperatures, cathode materials are tailored to reduce oxygen loss. More importantly, the kinetics of the batteries are affected by the dramatic volume expansion caused by the insertion and removal of metallic Li. A stable SEI layer must grow on the electrode surface and stay intact after several battery cycles in order to solve the issues of volume change, electrode breakdown, and side reactions [31-32]. Due to their high theoretical capacity of 1675 mAh g⁻¹, sulfur cathodes have recently been used in Li batteries. Sulfur's enormous storage capacity is the result of the cathode's electrochemical cleft and restructuring of sulfur bonds. Li-sulfur batteries have a significant advantage over Li-ion batteries since their electrode loads are lower. As a rule, sulfur is abundant, harmless to the environment, and easy to get by, especially because it is mostly extracted from the surface of the Earth [33].

In addition, Li-S batteries are often robust and resistant to harm even when subjected to extreme conditions. Li-sulfur batteries are better, but they come with their own set of challenges and restrictions. For sulfur-based cathodes, the shuttle mechanism is the main disadvantage [34-35]. Since sulfur is not conductive, the electrode must stay in close contact with a conductive layer (like carbon) if high efficiency is the goal. In addition, passivation layers raise questions about the Li electrode's security. Periodic charge and discharge cycles in Li-sulfur batteries may lead to the growth of spike-like deposits (dendrites) on the surface of the Li anode, significantly lowering the battery's performance [35].

Furthermore, spike layers may impede the current-generating migration of Li ions. Lower sulfur utilization and a significant shuttling effect that disrupts electro-chemical kinetics are the results of the sulfur electrode's slow conversion processes during charge-discharge cycles. In addition, the cyclic stability of the electrodes is diminished since the electrode material dissolves in the electrolyte. Other key downsides are the self-discharge property and instability of Li-anode insoluble lithium polysulfide, as the products of the self-discharge transmit back to the sulfur cathode, creating a re-oxidation process. Over time, a battery's overall efficiency might suffer due to a loss in capacity, lifespan, and coulombic efficiency [36-38].

Li-S batteries are being studied by a large number of scientists because of its potential as a high-capacity, environmentally friendly battery of the future. In order to provide a comprehensive knowledge of the progress made in metal-sulfur batteries, we will quickly cover some of the recent advancements in Li-S below. Using silver iodide as a host material in Li-S batteries, [39] included a halogen into the transition metal cathodes. In the electrochemical process, Li-iodide and silver have a synergistic effect that increases ionic conductivity and so blocks the polysulfide shuttle. Carbon materials exhibit high conductivity and adsorb strongly with Li-polysulfides, suggesting that they may be able to mitigate the polysulfide shuttle effect [40]. A nanoparticle-encapsulated core-shell sulfur cathode inhibits polysulfide shuttle.

There are a variety of sophisticated cathodes employed in Li-S batteries, including sulfur shells with MoS₂, FeS₃, MnO₂, TiO₂, SiO₂, Ga, Ag, PANI, and PDA and polymer core nanoparticles [41]. The regulation of dendrites in Li-S batteries is another active area of study. Currently, a 3D-printed host with nitrogen-doped Ti₃C₂ Mxenes is being considered as a dendrite suppressor. By providing additional active sites for the lithiophilic-sulfiphilic process, the porous nitrogen-rich Mxene structure extends the anode's lifetime to 800 hours at 5 milliamperes per square centimeter [42]. A dendrite-free anode and undisturbed ion transport are possible thanks to tunnel confinement provided by a graphitized, three-dimensional framework with high porosity [43]. High mass loading is made possible by the fibrous materials' enhanced

surface to volume ratio [44]. Controlling dendrites and polysulfide shuttles in sulfur batteries may be possible using knowledge gained from studying nanofibrous, porous frameworks, and 2D and 3D structures. A more practical strategy for controlling dendrite development is the use of solid-state metal-sulfur batteries. Depending on the specifics of the cell components, it may be necessary to fine-tune the electrolyte's thickness (ceramic, polymer, hybrid, gel), internal resistance, and temperature.

4. Prevention of Dendrite Formations in Batteries

Batteries at present cannot store all the energy produced by a wind farm during a very windy night for use the following day. One intriguing alternative to making a battery with a greater capacity is to replace the ordinary battery's negative electrode with one composed of lithium metal. Dendrites, which are deposits that develop on electrode surfaces while charging, cause the batteries to short circuit and pose a significant safety risk. How to stop dendrites from growing is something that has only just been figured out by scientists. Experts from the Department of Energy's Pacific Northwest National Laboratory worked together for the first time to successfully grow protective coatings around the anodes, preventing the formation of dendrites. The results of this research, conducted by the Department of Energy's Joint Center for Energy Storage Research (JCESR), may aid in the development of a secure metallic lithium anode and, in turn, pave the way for the widespread adoption of high-energy-density battery systems for use in electric vehicles and the long-term storage of renewable energy [45].

Researchers from the Pacific Northwest National Laboratory, Harbin Institute of Technology, Wuhan University, Tianjin Institute of Power Sources, and the United States Army Research Laboratory collaborated on a study to investigate the efficacy of an electrolyte additive known as cesium hexafluorophosphate (CsPF₆) in enhancing the performance of batteries and safeguarding the anode, which is the negative electrode. In order to evaluate the chemical composition and microstructure of the samples, respectively, high-resolution x-ray photoelectron spectroscopy and energy-dispersive x-ray analysis were used [46]. Both of this equipment may be found at the DOE's Environmental Molecular Spectroscopy Laboratory (EMSL). Films formed of densely packed lithium Nano rods, which look like a handful of dried spaghetti noodles, became dendrite-free thanks to the addition of CsPF₆[47]. The incorporation of the electrolyte additive made it easier to produce a layer of lithium-rich solid electrolyte interface material, which was one of the benefits of this process. This complicated layer is produced as a result of the rapid breakdown of the electrolyte, which is the component of batteries that serves as a conduit for the transport of lithium ions between the electrodes. During the charging process, a synergistic effect was produced by the Cs⁺ additive and the SEI layer, which resulted in the formation of lithium ions on the surface without the formation of any dendrites. Even after several charging/using cycles or deposition/stripping cycles, the lithium films did not develop any dendrites. The results demonstrate that CsPF₆ preserves the anode, enhances battery performance, and promotes the orderly and smooth development of lithium metal layers [48].

5. Controlling the Quality of Commercial Lithium-Ion Cells

When a Li-ion battery develops a short on the inside, it may violently disassemble itself. Only superior production techniques and stringent quality control can guarantee this. Cell makers have blamed lax quality control for the spate of Li-ion battery recalls that occurred between the years 2000 and 2006. These internal short circuits were caused by burrs from improperly cut CIDs, flakes of anodized materials from the interior of the cell can, and active material impurities adhering to the electrodes, according to destructive analysis of cells from the same "lot" or "date code." However, the cells involved in the failures were destroyed beyond analysis. Overcharging cells and batteries is another key contributor to internal short circuit creation. In the case of original equipment manufacturers, it has been determined that strict control of charge and discharge voltage, together with proper cell-voltage monitoring, balancing, and protection, is required. However, strict restrictions must be included in the battery design when commercial cells are employed in the development of specialized batteries for usage in space, underwater, and ground (electric vehicle) applications [49]. In addition to the more apparent dangers of overcharge, over discharge, and external shorts, the subtler danger of internal short generation due to use beyond their limitations should also be tested for by doing the appropriate level of testing (cell, bank, string, module, battery, etc.). Thermal runaway can happen in cells if their operating parameters are exceeded. This can be caused by localized heating, separator breakdown, imbalanced cell impedances, and finally the formation of lithium dendrites (overcharge) and electrolytic dissolution of copper with copper deposition on the anode, cathode, and separator (over discharge) [50].

The proliferation of cell producers, many of which are of extremely poor quality, is a serious problem for commercial Li-ion cells. Several after-market cells and batteries are being developed and offered at extremely cheap rates owing to the higher cost of high purity raw materials used in the creation of the electrodes and due to the demand for supply of

Li-ion cells. The aftermarket batteries are produced using low-grade raw materials and in unsanitary circumstances with little regard to quality. As a result, Li-ion cells and batteries have experienced thermal runaway in the wild. Avoid using aftermarket cells and batteries as much as possible, particularly in a human-rated setting. Cell acceptance tests performed at the manufacturing facility and initial screening of the cells during the battery buildup process are not the only ones that need to be considered; audits of the cell manufacturing facility, destructive analysis of freshly manufactured cells, and stringent screening techniques are also important. Cleanliness and humidity levels in cell-manufacturing facilities are of the utmost importance [51-52]. Electrode material powders, slurries, etc., should be filtered via a magnetic filter to get rid of any metal particles. Electrode coating sites, as well as slicing and cutting locations, should employ magnets, adhesive tape trays, and vacuum extractions. Because of the potential for electrolyte additions to cause changes in humidity and the entry of contaminants from the environment, such additions should be made with as little environmental contact as possible. X-raying cells is necessary to ensure that the electrodes and separator are properly positioned at both ends of the cell. The tabs should be X-rayed to ensure they are properly bent and not too close to the header or the electrodes, both of which might cause a short. Manufacturers of cells should conduct random sample testing for performance and safety of cells produced from each lot, as well as an independent review of incoming raw materials [53-54]. Lastly, cell traceability back to its raw materials and parts is needed for strict monitoring, finding and fixing problems before they happen, and improving the quality of cell production over time.

6. Conclusion

Many modern vehicles and mobile phones rely on Lithium ion batteries that employ a liquid electrolyte. As the battery is charged or discharged, electrons or charged ions go from the anode to the cathode via the electrolyte that separates them. Possible combustibility of liquid electrolytes is a major risk factor. But the use of lithium electrodes in rechargeable Lithium ion batteries is a very exciting prospect since it might increase the battery's capacity. Dendrites are long, thin metal projections that may grow from one electrode through the electrolyte substance to the other. In the long run, these dendrites might reach the opposite electrode, causing a short circuit that would destroy the battery and any device it powers. Dendrite production is a key source of worry in Lithium ion batteries since it has been linked to battery fires. Rechargeable Lithium ion batteries have not been developed due to the dangers of short circuiting caused by dendrite growth. Despite the fact that the use of lithium ion batteries with a solid electrolyte, like some ceramics, has many benefits over the use of lithium ion batteries with a liquid electrolyte, such as increased safety and greater energy storage capacity, the development of lithium ion batteries with solid electrolytes has been difficult. There is significant worry that dendrites may grow in these solid electrolyte batteries. Finding the optimal solid electrolyte material that is stiff and not elastic has been a primary focus of previous efforts to avoid dendrite development in lithium batteries using solid electrolyte. To prevent the dendrites from penetrating the electrolyte material, researchers looked at substances with a high enough shear modulus, a measure of how stiff or soft a substance is. In this regard, lithium phosphorous sulfide and metal oxides have been studied as two of the most promising solid electrolyte materials. Researchers have tried several different solid electrolyte materials in an attempt to stop dendrite growth, but their efforts have shown inconclusive results. Scientists headed by MIT's Yet-Ming Chiang evaluated four different types of solid electrolyte materials and analyzed their behavior throughout charge and discharge cycles to determine why stiffer solid electrolyte material could not prevent dendrite growth. Their investigation revealed a striking dissimilarity in the processes by which dendrites grow in solid and liquid electrolyte materials. According to the new findings, dendritic growth is kicked off when lithium from one electrode deposits onto pits, fissures, or scratches on the electrode surface through electrochemical reaction. Over time, this deposit builds up and spreads toward the opposite electrode, leading to disastrous results. In addition, the findings of this study demonstrated that dendritic expansion occurs at the dendrite's apex rather than its base. Therefore, developing electrodes with smoother surfaces may drastically minimize, and even prevent, dendrite growth.

Compliance with ethical standards

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Disclosure of conflict of interest

The authors declare no conflict of interest.

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