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The Effects of Different Materials Used in Non-Aqueous Lithium Air Battery Electrolytes on Ionic Conductivity and Durability

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Abstract

Batteries are increasingly important in moving away from fossil fuels towards a more sustainable future; they are currently used in many different applications, such as cars, portable electronics, and drones, but certain applications of batteries, such as in large aircraft, are limited by their gravimetric energy density. While many high energy density batteries have been synthesized, most lack high durability and are unable to withstand many charge and discharge cycles without losing considerable energy storage capacity. For example, lithium air batteries have one of the highest gravimetric energy densities of any battery, but because lithium ions are not able to flow freely from the anode to the cathode and vice versa. This paper examines recent advancements in lithium air battery materials and explores any potential developments for lithium air battery viability. It also discusses the various issues that plague lithium air battery advancement, and its place in the future of green energy and a viable source of energy storage.

Keywords: Batteries, Energy density, Lithium-air battery, Durability

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1. Introduction

Batteries are used in a variety of applications, from small portable electronic devices to EV's. This has partly been due to numerous advances in the volumetric energy densities of batteries, allowing ample amounts of energy to be efficiently stored in a small form factor. But one area where batteries have struggled to be utilized is where light weight high energy storage capabilities are required, such as in the aerospace industry. This is currently an area where battery technology is lacking, as though there have been considerable improvement in volumetric energy densities over the years, gravimetric energy density improvements have been lacking, as there has not been a pressing need for such capabilities, until now, when the world is transition into to a future of renewable energy sources.

Attempts to improve gravimetric energy densities in standard lithium ion batteries have been successful to an extent in that their weight has not hampered their use on a broad scale, but successful attempts to drastically improve gravimetric energy densities to that point where they can compete with gasoline and jet fuels have been limited due to the physical properties of electrode materials as well as the electrolytes. Lithium air batteries, however, relying on a porous oxygen based cathode, are able to yield gravimetric energy densities up to 39.6 MJ/kg, close to that of gasoline (*Lithium-Air Battery - an Overview | ScienceDirect Topics*, n.d.). However, as promising as it sounds, lithium air batteries are plagued by numerous issues, the most significant of which is their low cyclability, caused by poor ionic conductivity of the electrolytes used in them.

This paper discusses the basics of lithium ion

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batteries as well as the advantages and issues related to lithium air batteries and their real world implementation.

2. Findings

2.1 Basic Battery Electrochemistry

To generate current which can be used to power devices, a steady flow of electrons must be generated, which move through such devices, such as a lightbulb. This flow of electrons comes in the form of alternating current, where direction of flow alternates, and direct current, where the flow is in a singular direction, the latter of which batteries provide. In order to generate this flow of electrons, redox reactions must take place within the battery. Redox reactions can be divided into two half-reactions, the oxidation and reduction reactions. These reactions help to convert the chemical energy stored in batteries into electrical energy. In the oxidation reaction, atoms lose electrons and ions. In the reduction reaction, the electrons, and ions, come together, leading to an atom gaining electrons; this process uses up electrons. Essentially the oxidation reactions produce electrons, which are then used up by the reduction reactions.

Due to the opposite nature of these half reactions, they have to occur at separate places within a battery. Batteries typically have four major components; an anode, a cathode, electrolyte solution, and a casing (to protect the battery from external damage). The oxidation half reactions occur at the anode, and the reduction reactions occur at the cathode, the two of which are separated by an electrolyte solution. When the oxidation reaction occurs at the anode, the electrons produced essentially pile up and since electrons are negatively charged, they repel each other and want to move away to a less negative area which would be at the cathode where these electrons can be used up (mischa, 2016). But since the anode and cathode are separated within the battery and electrons are prevented from flowing from the anode to the cathode within the battery, an alternative route can be created where electrons can flow from the anode to the cathode and through this route is where current is created (mischa, 2016). In this route things that require power can be placed, such as a lightbulb, which the electrons will flow through on their way to the cathode (Path of an Electron through an Electric Circuit, n.d.). But if this route from the anode to the cathode is blocked, nothing occurs as no electrons are moving and no current is generated (mischa, 2016). However, this process is limited to the material available to be used in the reduction reactions within the battery; once those are used up, the redox reactions can no longer occur and the battery is essentially dead; this is what happens in non-rechargeable batteries when they are used (Every Battery Eventually Dies – Here's Why, n.d.).

2.2 Conventional Electrolytes

An electrolyte paste prevents the electrons from the anode reaching the cathode where they would be used up (mischa, 2016). The casing just holds the components in place and protects the battery from damage. An electrolyte is a substance that is found in all batteries. They can come in many different forms such as liquids, gels, as well as solids, in some solid-state batteries. An electrolyte is a very important part of the battery because it is what allows the ions to move from the anode to the cathode and vice versa. This is important for a battery's ability to be charged and recharged as the ions from the redox reaction need to be able to get back to their anode and cathode (Components of Cells and Batteries, n.d.). Charging basically reverses the ions back to their starting point, their anode and cathode, however, not all ions make it back and over time this causes the energy capacity to degrade over time, causing it to eventually become unusable (Components of Cells and Batteries, n.d.). In batteries such as lithium air batteries, the issue is with non-aqueous electrolytes, which are very resistant to allowing the products of the redox reactions through, making lithium air batteries almost unusable.

2.3 Lithium Ion Batteries

Lithium-ion batteries are relatively high energy density, and most can be recharged. The most common type of lithium-ion battery is a lithium cobalt oxide battery (A Guide to the 6 Main Types of

Lithium Batteries, 2021). In this battery, Li^+ ions, lithium in a +1 oxidation state, move from the anode to the cathode and vice versa through the electrolyte solution. The anode in lithium cobalt oxide batteries contains graphite which helps the batteries have a higher energy density, faster charging, higher durability, and a longer lifespan. The graphite used as the anode is lithiated, meaning the graphite is made of intercalated layers of lithium and graphite with a chemical formula of Li_xC_6. The cathode in lithium cobalt batteries is made of a lithiated metal oxide, specifically Li_xCoO_2. However, lithium-ion batteries can also have cathodes made of other materials, such as polyion type cathodes: $LiMPO_4$, $LiM(PO_4)F$, $Li_2MP_2O_7$, Li_2MSiO_4, $LiMSO_4F$, $LiMBO_3$. Lithium-ion batteries can also have cathodes made of spinel oxides such as $LiMn_2O_4$. The electrolyte solution should allow the Li^+ ions to move freely between the anode and the cathode. Most lithium-ion batteries today use a liquid or gel electrolyte such as $LiPF_6$. Solid-state electrolytes are not currently used commercially because many of the solid-state electrolytes do not allow the Li^+ ions to pass through freely (Bartholome et al., n.d.). During the battery's discharge, the lithium in the lithiated graphite anode is oxidized into Li^+, and an electron is freed. The chemical reaction happening is

$C_6Li \rightarrow C_6 + Li^+ + e^-$

The Li^+ ions move from the anode to the cathode through the electrolyte solution, which will get used up in the reduction reaction at the cathode. The electrons produced during oxidation want to move from the anode to the cathode. However, since electrons take the path of least resistance, they do not move through the electrolyte solution to the cathode. There is also a separating layer in many batteries which prevents the flow of electrons from the anode to the cathode and only allows the Li^+ ions to pass through. In the cathode, where the reduction reaction occurs, the cobalt is reduced to a lower oxidation state, and the Li^+ ion combines with the electrons from the oxidation reaction to form lithium cobalt oxide. Since the electron is required for the reduction reaction to occur, the reduction reaction will not occur until a path for the electrons is given from the anode to the cathode. So, no current will be generated until the battery is plugged in with its positive and negative terminals connected. The chemical reaction occurring at the cathode is

$_XCoO_2(s) + xLi^+ + xe^- \rightarrow LiCoO_2(s)$.

When the battery recharges, a current put into the battery causes the Li^+ ions to go from the LiCoO back to the lithiated graphite anode (Lithium-Air Battery - an Overview | ScienceDirect Topics, n.d.).

2.4 Solid State Electrolytes

Solid-state batteries are another very high energy-density battery type being researched. They work similar to conventional batteries, except for electrolytes. Conventional batteries use a liquid or gel electrolyte, but these have many issues, such as being more prone to catching on fire and causing them to be heavier (Balshaw2019-11-11T09:38:00+00:00, n.d.). Solid-state batteries can solve these issues with faster charging times, higher durability, and much safer (Mauger et al., 2019). However, solid-state batteries are still being researched, as creating a suitable electrolyte is still causing issues. Solid-state electrolytes, unlike liquid or gel electrolytes, make it much more difficult for ions to pass from the anode to the cathode (Uddin & Cho, 2018).

2.5 Effects of Temperature on Batteries

Materials with a higher electrochemical potential lead to higher energy-density batteries (Liu et al., 2016). Lithium is used in many batteries because of its high electrochemical potential and its lightweight (Goonan, 2012). As the temperature increases, batteries' energy storage capacity increases. However, as the temperature goes down, the energy storage capacity of batteries goes down (BU-502: Discharging at High and Low Temperatures, 2010). So, the temperature is one of the main effects of battery performance: including discharge rate, recharge rate, and energy density (Temperature Effects on Batteries, n.d.). Another major trend is that charging batteries faster can also degrade the batteries faster. Slower charging can increase the lifespan of a battery, and the number of charge cycles can also affect the battery's energy capacity. More charges lead to less energy capacity over time due to battery degradation (src="https://www.rd.com/wp-content/uploads/2020/07/Brooke-Nelson.jpg?fit=50 et al., 2019).

2.6 Lithium-Air Batteries

One type of battery with a much higher energy density than lithium-ion batteries is lithium-air batteries. Lithium-air batteries have one of the highest theoretical energy densities of any battery, about five times more than lithium-ion batteries and close to that of gasoline at 43.2MJ/Kg (Liu et al., 2020). Two types of rechargeable lithium-air batteries are being worked on, aqueous and non-aqueous. Lithium-air batteries have the same components as a normal battery: an anode and a cathode, and they rely on reduction and oxidation reactions to generate current. The anode used is pure Lithium, and the cathode consists of carbon and oxygen gas (Liu et al., 2020). The electrolytes used are non-aqueous organic electrolytes, allowing the lithium ions to pass through freely from the anode to the cathode and vice versa. Lithium-air batteries are separated based on their electrolytes: non-aqueous aprotic electrolyte, aqueous electrolyte, solid-state electrolyte, and hybrid electrolyte (Liu et al., 2020). As the battery discharges, the Li^+ ions move from the Lithium to the carbon-oxygen anode by dissolving and moving through the electrolyte. The electrons move to the cathode from the anode through an external path. This process is reversed when the battery is charged as in a conventional battery. The chemical reactions for discharging are

$2Li + O_2 \rightarrow Li_2O_2$ and $4Li + O_2 \rightarrow 2Li_2O$.

The anode half reaction, oxidation, is

$$Li \rightarrow Li^+ + e^-$$

and the cathode half reaction, reduction, is

$2Li^+ + O + 2e^- \rightarrow Li_2O_2$ (Liu et al., 2020).

2.7 Lithium-Air Battery Issues

The issue with this type of lithium-air cell is that the product of the reaction does not dissolve in the electrolyte and ends up depositing in the cathode and, over time, reduces the capacity and durability of the battery due to the usable material within the battery, reducing (Liu et al., 2020). An aqueous electrolyte can solve this problem by using a lithium-ion conductive water-stable solid electrolyte in which the discharge product is soluble within the electrolyte. However, this results in a lower energy density. The redox reaction for this type of cell is

$4Li + O_2 + 6H_2O \leftrightarrow 4(LiOHH_2O)$ (Liu et al., 2020).

The air used in these batteries can also affect performance. Moisture in the air can corrode the lithium within the battery.

When operating lithium-air batteries, the ideal conditions would be to operate them using pure oxygen (Asadi et al., 2018). But to make them viable for use on a wide scale in many different industries, they need to be able to work in an environment where they will be exposed to different environmental factors such as heat, cold, humidity, and other impurities in the air.

Humidity for lithium-air batteries can be both beneficial and harmful as well. This can be seen in a study where under the conditions of pure oxygen and zero humidity, the discharge voltage was 2.51 volts. However, under the conditions of 84% humidity, the discharge voltage increased to 2.79 volts. The energy efficiencies also increased from 66.2% to 73.8% from the dry air to the air with 84% relative humidity, respectively (Tan et al., 2016). But this can also be harmful as this can cause a buildup of hydrogen within the battery (Byrne, 2021).

The main issue right now with lithium-air batteries is that when the redox reactions occur within lithium-air batteries, the product, lithium peroxide, is not able to decompose back into lithium oxide (Tan et al., 2016), which reduces battery energy storage capacity (Song et al., 2020).

The product of the redox reactions for lithium-air batteries under high relative humidity is lithium-hydroxide, which requires less energy to decompose than the lithium peroxide created from the dry air. This allows for better batteries because lithium hydroxide allows for a higher discharge voltage when formed (Gallant, 2020).

But other problems can arise from using lithium-air batteries under high humidity environments. This is because pure lithium is highly sensitive to moisture. If excessive moisture comes into contact with the lithium, lithium hydroxide will also be formed, but also hydrogen, which makes the batteries unsafe due to it being very flammable (Byrne, 2021).

Though it may be possible to create lithium-air batteries with very high gravimetric energy densities, such as up to 11000 Wh/Kg, it may not be practical to be able to use these lithium-air batteries due to other factors that would also have to be considered other than just gravimetric energy density (Lithium-Air Battery - an Overview | ScienceDirect Topics, n.d.). Many lab tests show that lithium-air batteries can reach this gravimetric energy density. However, they have also shown that the use of non-aqueous electrolytes to be able to attain these results has led to speedy degradation in lithium-air batteries, such as only a four charge and recharge cycles, making their gravimetric energy density something that is not very useful as it cannot be sustained (Edge et al., 2021). So, a lower gravimetric energy density battery that lasts longer may be better in the real world due to lithium-air batteries' impracticalities.

But there have been some advancements that could help reduce the degradation of lithium-air batteries. One of the many factors responsible for the degradation of lithium-air batteries is lithium superoxide (LiO_2). Therefore encapsulating, or trapping, the lithium superoxide could increase the recyclability of lithium-air batteries and prevent the degradation of these batteries (Encapsulation as a Method for Preventing Degradation in Li-Air Batteries, n.d.).

But one of the other issues is the practicality of attaining the air needed for them. Many of the impurities in the air used for these batteries can also cause many problems with their durability. The air in lithium-air batteries can have trace amounts of moisture, carbon dioxide, nitrogen, etc. These impurities can cause many side reactions at the anode and the cathode in the battery, causing instability (Liu et al., 2017).

Making lithium-air batteries viable for use with ambient air is an advancement that can make lithium-air batteries viable if the durability issues are fixed. One of the components of ambient air is carbon dioxide. But it can react with oxygen, as well as with lithium, forming lithium carbonate. The advantage of lithium carbonate is that it can provide a way to make lithium-air batteries cyclable, being able to withstand multiple charger cycles due to the lithium carbonate being easier to decompose. Lithium carbonate, on the other hand, can also be detrimental to lithium-air cells as it can also cause the cells to dry out (Liu et al., 2020).

Lithium-air batteries can last many charge cycles due to recent development, even up to 1200 charger cycles (Kondori et al.). However, the above tests were run at a very low voltage, allowing the lithium-air batteries to last longer than they usually would have. But discharge voltage is vital in battery use as though a battery may be able to hold a lot of power if it cannot discharge it at a reasonable amount, then it is not of any actual use.

3. Discussion

Lithium-air batteries, as any other liquid or gel electrolyte battery, come with risks. The most major of these risks is the risk of the batteries catching on fire due to overheating or leaking, causing the release of many flammable compounds. Lithium-air batteries currently work best under very high humidity conditions, provided water can be kept from damaging certain electronics that manage the battery. The water in the air can react with lithium-ion, forming lithium hydroxide. This helps lithium-air batteries run better because lithium hydroxide is more easily decomposable within the battery, allowing the battery to last longer due to easier charge and recharge cycles being able to run. However, when lithium reacts with water, it forms both lithium hydroxide and hydrogen gas, the latter of which can be very flammable, causing many safety concerns with lithium-air batteries. So, the conditions in which these batteries function the best also yields the most dangerous conditions these batteries are used in. This must be worked out in the future by altering the material used for the electrodes, allowing safer products to form to create safer batteries.

Lithium-air batteries may be viable for use in the future. However, only if substantial research is dedicated to increasing their durability, discharge rates, and energy capacity. Lithium-air batteries cannot last for numerous charge cycles, which is very important for them to become viable. Lithium-ion batteries used in devices that require very little power, such as TVs or small appliances, can easily be replaced at a low cost. However, much larger lithium-based batteries, such as the ones used in EVs today, are much more expensive to manufacture and replace, with many costing about 15% of the price of the car itself. So, the even more expensive lithium-air batteries must demonstrate a long lifespan in order for them to be economically viable. Though they have many performance benefits, they only provide a small quantity of use other than in aerospace applications and other applications where the weight of the battery is very critical. However, for EVs, battery weight is not as much of a detriment to their range as it would be in planes, and therefore is less of a concern. Due to spatial limitations, volumetric energy density is more critical in many uses today. So, if lithium-air batteries became commercially viable, they would not be used on a vast scale, causing their costs to be much greater than traditional lithium-ion batteries.

The issues with solid-state batteries may be solved before those of lithium-air batteries. Even if lithium-air batteries' durability is improved to viability, they will still rely on liquid or gel electrolytes, which are still not as safe as solid-state electrolytes. A lithium-ion solid-state battery could have a very high energy density, not as high as lithium-air batteries, but still, a high enough gravimetric energy density for them to be revolutionary. A pairing of lithium-air batteries that use solid-state electrolytes would result in the safest and highest energy-density battery. But the cost would preclude this kind of battery as it would only be economically viable in applications of high performance or in aerospace applications where saving weight is a requirement and can result in more range, which would be more cost-effective. There are also many advancements that can result from creating a lithium-air solid-state battery, such as electric planes, which still need to be more viable due to the gravimetric energy density of current batteries being very low when compared with that of jet fuel.

4. Conclusion

The current advancement in lithium-air batteries has slightly increased their durability. There has also been much research done in all the major areas where problems are in lithium-air batteries: with the electrolytes, the redox products, and the side reactions occurring in lithium-air batteries. But solid-state batteries are currently beating lithium-air batteries to the market and do not require as much development as lithium-air batteries. Moreover, for the large-scale use of lithium-air batteries, especially in aerospace operations, these batteries would have to have a very high discharge rate and be able to sustain how much energy they can hold over long periods. Since lithium-air batteries are still struggling first to become usable, their discharge rate has yet to be explored that much, and more research would have to be done to make them more commercially viable in the future.

There is currently some successful research into making lithium-air batteries last for multiple charge cycles, making them viable for commercial use (Kondori et al.). Specifically, getting them to last for 1200 charger cycles, in some cases, is very close to how many charge cycles current lithium-ion batteries, used in, for example, electric cars, last. However, this testing was done with meager discharge rates, causing the number of charger cycles the batteries lasted to be much higher than in practical use. The discharge rate is critical because it is about how many enemies a battery can hold and how much it can release at a time. If there were an electric car with a range of 600 miles but a max speed of 5mi/hr., then it would not really be of much use. This is still the current dilemma lithium-air batteries face. However, it is progress that these batteries can at least last more than a couple of charger cycles without dying.

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