Review

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Electrolytes: From a Thorn Comes a Rose, and from a Rose, a Thorn

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Abstract: In this article we take a stroll through the metaphorical rose garden, part of the flower garden, that is electrochemical storage systems. We take a closer look at batteries as the center-pieces – the roses – and especially at the system component of batteries, the electrolyte and its development, throughout history. Just as a rose can only develop its unique beauty when taken good care of and provided with clean water and suitable nutrients, a battery cell cannot function without its electrolyte; the seemingly trivial component that has to juggle a multitude of requirements in order to make batteries truly bloom.

"*For millions of years flowers have been producing thorns. For millions of years sheep have been eating them all the same. And it's not serious, trying to understand why flowers go to such trouble to produce thorns that are good for nothing? It's not important, the war between the sheep and the flowers? It's no more serious and more important than the numbers that fat red gentleman is adding up? Suppose I happen to know a unique flower, one that exists nowhere in the world except on my planet, one that a little sheep can wipe out in a single bite one morning, just like that, without even realizing what he'd doing - that isn't important? If someone loves a flower of which just one example exists among all the millions and millions of stars, that's enough to make him happy when he looks at the stars. He tells himself 'My flower's up there somewhere…' But if the sheep eats the flower, then for him it's as if, suddenly, all the stars went out. And that isn't important?*"
– Antoine de Saint-Exupéry, The Little Prince

1. Introduction

Inspired by the term "Rosarium", denoting a rose garden as a metaphor for a collection of wise philosopher's sayings, the word "rose", beautiful to look at, fragrant to smell, sweet to taste and enjoyed for thousands of years, directly evokes. You can quickly pick most kinds of flowers and take a big smell and feel good for a little while after. But trying that with a rose, you might just get a finger pricked with one of its thorns along the stem and end up asking yourself, why do roses have thorns? Roses developed thorns as a defense mechanism, after a while of taking abuse from the creatures that would not leave them alone, as the only way to protect themselves from being destroyed.

Even the rose, beautiful and enticing, is not without flaws. So too, something as stunning as a rose is also not perfect, for within, there are also flaws reflecting the cycle of life, from life to death, death to life in each system known. Something that begins as an imperfection or flaw can grow to be as beautiful as a rose. A garden of flowers stands as great metaphor for electrochemical energy storage systems, batteries as their main representatives, they are the roses and thus the show-pieces within the variety of different flowers. After all, nature is a great teacher providing many lessons learned by spending time out in the natural world. In line with this, when planting a garden, a clear vision has to be made in respect to the type, the purpose it has to serve and how it should be laid out. In other words, things must be prioritized in order to prevent possible problems and challenges that would inevitably become unmanageable in time, and manage those by minimizing their footprints. Only the seeds that will bear the fruits that are most essential should be planted, followed by other seeds, however, avoiding to plant too many flowers at the same time. Whatever it is that was decided to plant in the garden, it won't grow well unless there is a healthy soil available. A scientist, as an experienced gardener, spends a lot of time, energy, and expense to advance "the soil of the garden" and generate a strong foundation and environment that is conducive to future "vibrant growth". Creating a truly great soil might take years, however by laying down this strong foundation it will be much more likely that the garden will yield a beautiful, strong crop and significantly improve its productivity. The main goal is to plant the adequate/right seeds

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in order to achieve a targeted result. In addition, the garden's conditions have to be carefully assessed as different plants call for different environments to thrive. When setting the primary goals, the given environment and the circumstances must be thoroughly analyzed. A "pest-free garden" requires a fence around it. By setting boundaries and being wary of "weeds" inevitably present in each system, a clear path should be constructed to effectively suppress or even eliminate them. This process requires patience as illustrated by the quote by John Wenger: "You can't pull on the plants and expect them to grow faster." In this process, one has to learn to deal with things outside of control. In the end, the harvest reap is what counts, as after all the hard work, the garden should start producing a bountiful crop, ready to be picked. On the other hand, if the harvest wasn't as bountiful as desired, or if some plants failed to bear fruit, it should be analyzed what went wrong and why. The obtained answers, should lead to a different strategy and new trial for the next generation of crops in order to optimize the properties and overall performance in the considered system foreseen for targeted application.

This manuscript extends an invitation to the readers, who will follow us in a walk through a rose garden comprising the colorful historical aspects and bright scientific, technological and engineering achievements in electrolyte for energy storage applications.

1.1 Definition of Electrolyte

If you ask chemists to define "electrolyte," most of them might say that an electrolyte is a compound which produces mobile ions with positive and negative electrical charge when dissolved in a solvent such as water. In more rigorous manner, electrolytes could be either covalent compounds that chemically react with the medium to produce ions, or ionic compounds that dissociate to yield their constituent cations and anions when dissolved, or simply the molten state of an ionic compound when its crystal lattice energy is overcome by heating. In electrochemistry, an electrolyte, also called electrolytic conductor, implies any substance that conducts electricity *via* the movement of ions. The word "electrolyte" was coined by Faraday, who derived the etymology from Ancient Greek ἠλεκτρο- (*ēlectro*-), prefix related to "electro", and λυτός (*lytos*) meaning "able to be untied, unfasten or loosened" (from PIE root *leu- "to loosen divide, cut apart"). Faraday believed that the conducting particles called "ions" were produced by electrolysis, and this belief was apparently reflected by the suffix of the word "-lytos".[1] This miscomprehension was corrected by Svante Arrhenius, who proposed in his dissertation the concept of the dissociation of solid crystalline salts into paired charged particles when dissolved, for which he was awarded the Nobel Prize in Chemistry in

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1903.[2] Arrhenius argued that, even in the absence of an electric current, solutions of salts already contain ions, and that chemical reactions in solution are reactions among ions.

Today we know that typical liquid electrolytes are formed in a process called solvation, where the components in a salt compound dissociate into ions, solvated by solvent molecules. With the application of voltage on an electrolyte *via* a pair of electrodes, the electrolyte conducts electricity. As electrons alone are not able to pass through an electrolyte that only allows ionic conduction, electrochemical reactions take place at the electrode | electrolyte interfaces, that is, at cathode due to consumption of the free or extra electrons and at the anode where another chemical reaction that leads to electrons being transferred to the cathode. During this process, a negative charge excess in the cathode and a positive charge excess in the anode will be developed, thus causing the ions in the electrolyte to move either way according to their electrical charge and neutralize resulted charges in order to keep the flow of electrons and the reaction going on.

An electrolyte is often a liquid. However, electrolytes may also come as a polymer, solid ceramic and molten salts, or as a hybrid (Figure 2).[3–6]

1.2 Electrolytes as Key Components

It is common wisdom, that materials science in the field of electrochemical energy storage has to follow a systemic approach as the interactions between active materials, the electrolyte, the separator and various inactive materials (binder, current collector, conductive fillers, cell housing, etc.), are of similar or even higher importance as the properties and performance parameters of the individual materials only. In particular, of utmost scientific interest are the numerous chemical, electrochemical and mechanical interactions of all components of electrochemical energy storage systems determining their operation, safety and life. Batteries are considered the systems of choice for many portable, mobile and stationary applications.[7] However, existing technologies still face diverse challenges as the overall performance of batteries is limited by the fundamental behavior of the materials used, among which electrolytes are often blamed as the shortest board in the bucket.

Most current commercial batteries use liquid electrolytes and in all types of these current and possible future batteries, the electrolyte plays a key role in terms of design and control of the redox processes as well as regarding material interactions, performance, long-term stability, cost and last but not least the safety of a battery. Being the component bridging while interacting with all other active and inactive battery cell parts, the electrolyte has a significant impact on many chemical and technological aspects of a battery. The scope ranges from conductivity, electrochemical and thermal stability, wettability, flammability and vapor pressure, to the essential interphase film-forming abilities, i. e., the formation of the so-called solid electrolyte interphase[8–9] (SEI) on the anode and the cathode electrolyte interphase (CEI). Although the electrolyte role is often considered as trivial due to the superficial belief that energy is decided by electrodes only, the right choice based on criteria that differ depending on the battery chemistry and targeted application, is rather crucial. After many decades of research, the electrolyte and its chemistry are still limiting and critical factors in the field of electrochemical energy storage systems. The development of new and the improvement of existing electrolytes is by no means non-trivial because of the complexity of interactions between electrolyte components with each other and with other battery cell components. Straightforward approaches for the optimization of existing electrolyte systems thus often lead to situations where specific targeted properties can only be improved at the expense of other relevant properties. This implies that current electrolytes are already close to their optimum performance in terms of a property compromise and that major gains can only be achieved by moving to substantially altered electrolyte systems.

For liquid electrolytes, a special focus is set on tailored syntheses of innovative and ultrapure electrolyte components, particularly conducting salts, organic solvents/co-solvents and (multi-)functional additives supported by selected theoretical calculations, simulations and modeling. Comprehensive characterization of electrolytes by means of emerging *in situ* and *operando* physicochemical, electrochemical, analytical, spectral and structural methods enables establishment of structure-property-performance relationships that provide insight in understanding and elucidation of main operation and failure mechanisms taking place in a battery comprising electrolyte | electrode interfaces and interphases.[10]

2. Electrolytes through History – Overview

In the garden of electrochemical storage devices, a main electrolyte solvent is water, as without it, each and every plant will wither away. When the water is tainted, the garden cannot thrive. Likewise, not every electrolyte is suitable for every electrochemical device. Quite the opposite is true. Each device has a specific set of requirements that the electrolyte needs to fulfill. As new electrochemical energy storage systems were developed with time, the required properties of the electrolyte also shifted and changed.

2.1 Aqueous Electrolytes

As an integral part of any electrochemical power source, the history of electrolytes shares its point of origin with the history of what we today call battery; in the research conducted by Luigi Galvani in the late 18th century. When he connected a piece of copper to a piece of zinc and touched them to different parts of a (dissected) frog's leg, he induced twitching in the muscles. Albeit known, the phenomenon of electricity was not understood at that time. Therefore, Galvani attributed

the movement of the dead frog's leg to a form of "animal electricity".[11] What we know today is that he built a simple setup, which is now named after him: Galvanic cell. The frog's muscles, or more precisely the salt containing cellular fluids in the frog's muscles, acted as the electrolyte for the two different metal electrodes. The muscle's movement was merely a side effect of the generated voltage. That animal entities were in fact not needed to generate electricity was later shown by Alessandro Volta, who stacked several alternating layers of zinc, brine soaked cardboard and copper to the famous voltaic pile and thereby built the first battery.[12] The realization that the combination of two different electrically conductive materials in a salt solution was enough to generate electricity led to the development of more advanced setups like the Daniell-cell in 1836,[13] or the zinc-carbon battery in 1866[14] that can still be found as a commercial system today. However, it was not until 1884 that Svante Arrhenius' suggestion of the dissociation of salts into electrically charged constituent parts[15] firstly enabled understanding of the working principles of the electrolyte's role as a medium for ion transport in batteries.

The understanding of a battery's governing principles led to the development of a plethora of different battery types like the aforementioned zinc-carbon battery, or the more modern alkaline battery[16] that was derived from it. Another type of battery operation principle that branched off early on, but has become the most important by far for our modern society is the rechargeable secondary battery, often called accumulator.[17] Amongst them is the "timeless classic"; the lead-acid battery,[18] nickel-cadmium and nickel metal-hydride batteries[19,20] as well as the revolutionary lithium-ion battery.[21–23]

From all individual battery units produced in 2019 in Japan, 56% (all except the lithium containing ones) use the same electrolyte solvent that was already used in the very first batteries ever constructed: Water (Figure 1). Water has a number of beneficial properties in terms of battery application. First and foremost, it features a unique combination of a high dielectric constant[24] with a low viscosity.[25] The high dielectric constant enables it to dissolve salts, which, in combination with the low viscosity, makes it an excellent ion conductor. Furthermore, it is cheap, abundant, non-flammable and non-toxic, which makes it an excellent choice in terms of

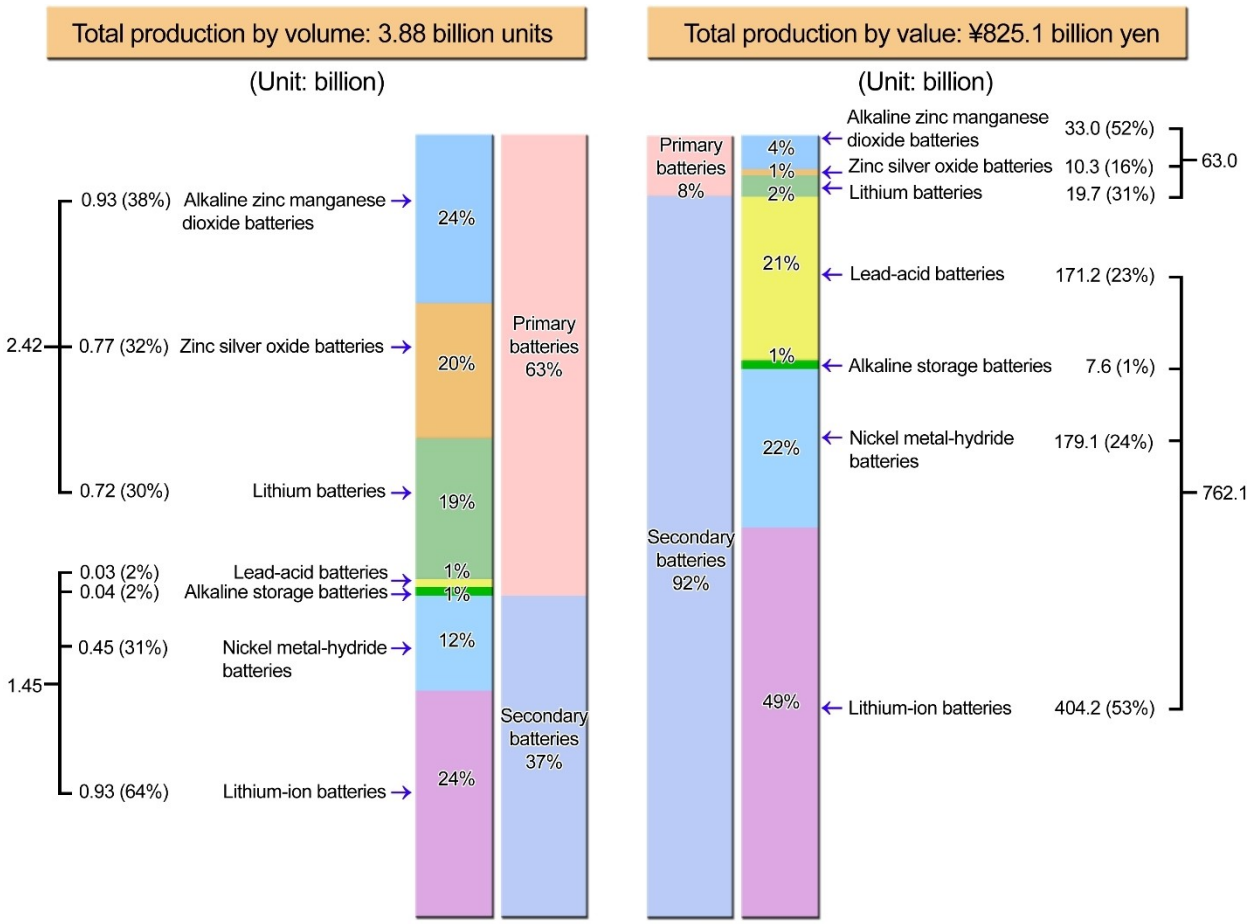

Figure 1. Total battery production statistics for Japan in 2019. Reproduced from [27].

end-user friendliness and safety. However, it does have a big disadvantage in electrochemical applications. Water has a relatively narrow electrochemical stability window of thermodynamically 1.23 V, which is the difference between its oxidation and reduction potentials at a given pH value.[26] If an electrode material with an operating potential outside of the stability window is used, water decomposes and hydrogen and/or oxygen gas will be produced in competition with the desired electrochemical reactions. This problem is mainly encountered when charging an aqueous electrolyte battery. It can be alleviated by sophisticated kinetic measures slowing down the water decomposition reaction, that manageably low amounts of hydrogen or oxygen are produced. As consequence, even thermodynamically impossible rechargeable battery systems, like the lead-acid battery or the nickel-metal hydride system operate very successfully in practice.

Since electric power is defined as the product of current and voltage, while battery energy as product of cell capacity and voltage, an increased battery voltage will deliver more power and energy for the same current and capacity. With rising popularity of portable electronics and especially with the dawn of electromobility, the battery's specific energy (energy per unit mass in Wh/kg) as well as deliverable power (in W) have become more important than ever. But to overcome the limitations imposed by the narrow electrochemical stability window of aqueous electrolytes, new battery concepts in non-aqueous media had to be developed. However, in recent years it was found that utilizing highly concentrated salt solutions such as so called "water-in-salt" electrolytes could significantly extend the electrolyte stability window while still utilizing water as solvent in minor amounts.[26]

2.2 Non-aqueous Liquid Electrolyte

It was realized already very early that lithium would make an ideal anode material due to its very high theoretical specific capacity of 3,860 $mAh\,g^{-1}$ (compared to *e.g.* zinc with 820 $mAh\,g^{-1}$)[28] as well as the lowest redox potential at 3.04 V vs. SHE.[26] The specific capacity is the amount of charge per unit mass that can be utilized for a given material. The difference in redox potential of both electrodes determines a galvanic cell voltage. Therefore, materials with a large specific capacity and either a strongly negative or strongly positive redox potential are beneficial for realizing high energy densities.

However, with lithium being an alkali metal it is obviously incompatible with aqueous electrolytes, thus requiring suitable electrolytes, for instance using water-free organic solvents. The task of finding a suitable solvent sounds simple enough, but turned out as quite challenging. Besides additional drawbacks further down the line like flammability and toxicity, organic solvents usually feature either a high dielectric constant or a low viscosity but seldom both,[29] unlike in the case of water, and thus typically suffer from greatly reduced ionic conductivity.[30] Furthermore, the typically low dielectric constant necessitates the use of conducting salts featuring weakly coordinating anions to achieve a reasonable solubility. From the multitude of investigated solvents, organic carbonates were soon identified as featuring the best compromise of properties and are still used today, half a century after the introduction of lithium into batteries.[31] While primary lithium metal batteries secured a niche where a long lifetime at high energy content is the prime requirement, the secondary battery market is dominated by the lithium-ion battery (Figure 1), a lithium metal free systems, which is however, at rough approximation facilitated by similar electrolytes and solvents as the lithium metal battery.

Both, lithium metal and graphite in its lithiated state, which is typically used as a negative electrode material in lithium-ion batteries, are thermodynamically unstable in the respective electrolytes.[32] Battery operation is only enabled by the formation of an effective protection layer that is known as the SEI.[33] SEI formation is indispensable for successful operation. In lithium-ion batteries it is formed in the first charge/discharge cycles while in a lithium-metal secondary battery solvent degradation occurs whenever fresh lithium comes into contact with the electrolyte (*i.e.* after assembly and on each charge), leading to continuous consumption of lithium, electrolyte and buildup of decomposition products.[34] As the battery is a closed system, and the amount of lithium sealed at the original state of the cell determines its capacity, the result of persisting consumption by SEI formation is usually a rapid, irreversible loss of cell capacity during operation. This is also the case when a given electrolyte fails to form an effective SEI under operation conditions, imposing an additional challenge in electrolyte development. Furthermore, lithium metal often tends to deposit as needle-like high surface area structures, *i.e.*, the so called dendrites, rather than as a uniform layer.[35] These dendrites can easily penetrate from the anode to the cathode and lead to internal short-circuits, which can potentially kick-off a thermal runaway of the battery cell and thus pose a severe safety risk.[36] In lithium-ion batteries, on the other hand, no metallic lithium occurs under normal operation and thermal runaways are rare events if the batteries are not subject to irregular operating conditions, abuse or damage. In fact, a study published in 2017 by the National Highway Traffic Safety Administration of the U.S. Department of Transportation concluded that "(…) the propensity and severity of fires and explosions from the accidental ignition of flammable electrolytic solvents used in lithium-ion battery systems are anticipated to be somewhat comparable to or perhaps slightly less than those for gasoline or diesel vehicular fuels."[37] Nevertheless, continuously improved safety is an important driving force of current research on lithium-ion batteries.

2.3 Polymer Electrolytes

The potential benefits of a polymeric electrolyte were already recognized in the 1970s.[38] With polymeric electrolyte, the

specific energy can be increased, especially in combination with a lithium metal negative electrode, where dendrite formation can be suppressed in polymeric electrolytes with high enough mechanical strength.[39–40] Furthermore, the absence of volatile organic solvents greatly reduces the fire hazard in case of a catastrophic failure and avoids liquid electrolyte leakage. However, as ion conduction in a polymer is usually coupled with segmental motion of the polymer chain rather than migration of ions between individual coordination sites, dry solid polymer electrolytes (SPE) often suffer from low ionic conductivities.[3] The batteries therefore need to either be operated at elevated temperature or the polymer needs to be gelled with plasticizers to form a quasi-solid as in the commercial LiPo lithium ion polymer batteries, that are often found in modern mobile electronics. From the need of improving the overall cycling performance of polymer electrolytes to make them feasible in application, the research field of single ion conductors (SIC) emerged, that aims to increase the performance by eliminating the anion contribution to charge transport. The anions move opposite to the lithium ions and accumulate at the negative electrode because they are not taking part in the battery reaction mechanism. This polarization has a detrimental effect on battery performance.[41] To eliminate anion migration, rather than using a simple blend of polymer and salt, polymers are used where the salt anions are covalently immobilized on the polymer chain, leaving only the lithium ions for charge transport. While much higher lithium ion conductivities can be achieved with polymer electrolytes in some cases, other challenges like poor interfacial contact between the solid electrodes and the also solid polymer remain to be tackled. This clearly underlines that overcoming the multitude of challenges in development of a suitable polymer electrolyte is far from easy, and more research on the fundamentals of these challenges is highly required.

2.4 Solid Ceramic Electrolytes

Besides solid polymer electrolytes, there are the purely inorganic solid ceramic electrolytes. Their thorough investigation began in the 1960s with the discovery of the surprisingly high room temperature ionic conductivity of silver sulfide iodide.[42] Even though most solid ionic conductors show conductivities several orders of magnitude lower than that of silver sulfide iodide, their principle feasibility was demonstrated with the success of the lithium-iodine pacemaker battery, an all-solid-state battery with a lithium iodide-based solid electrolyte.[43] The ionic conductivity stems from vacant (*i. e.* defects) or interstitial sites in the crystal lattice that enable cations to move through the solid matrix, making them intrinsic single ion conductors.[44] The development of the lithium-ion battery sparked new interest in solid electrolytes for the same reasons (higher energy density, better safety, etc.) as discussed for polymer electrolytes. Nowadays, the two main classes of compounds under study are (i) sulfides, that feature high ionic conductivities, but are highly moisture sensitive and therefore difficult to handle along with less electrochemical stability, and (ii) oxides, that are less sensitive and more electrochemically stable but typically feature lower ionic conductivities and are more difficult to process.[4] Differing from the SPEs, both classes of solid electrolytes still possess a certain level of electronic conductivity, which was believed to be the fundamental reason that encourages the formation of Li dendrite and dead Li.[45] Generally, inorganic solid ceramic electrolytes are also more or less brittle and suffer from high grain-boundary resistances,[46] which makes them especially susceptible to formation of fissures and contact loss during charge/discharge.[47] In addition, electrodes change shape during operation, hence continuous "wetting" of the electrode surfaces by the electrolyte via a conformal interface remains a challenge for long-term operation (by the way also to some extent for solid polymer electrolytes). Still, the assumed possibility of lithium dendrite suppression due to their mechanical properties as well as improved safety due to the absence of flammable compounds, make solid ceramic electrolytes a promising field of study for the development of advanced high energy batteries. Finally, it should be mentioned, that glassy electrolytes may be considered as solid electrolyte compromise between crystalline ceramic inorganic electrolytes and amorphous polymer electrolytes, which provides not only single cation conductivity but also essentially zero electronic conductivity, in addition to high mechanical strength that can prevent Li dendrite formation.

2.5 Hybrid Electrolytes

In hybrid electrolytes, at least two ion-conducting phases from different electrolyte classes exist (*e. g.* ceramic and polymer). In gel polymer electrolytes, however, the ionic conductivity only happens in the liquid phase and they are thus usually not considered to be hybrid electrolytes.[6] As solid polymer electrolytes and ceramic solid electrolytes were developed simultaneously and show mostly opposite tendencies in terms of conductivity and mechanical processability, it did not take long until efforts were made to combine both in order to overcome their individual limitations. While early attempts of creating hybrid electrolytes were to merely blend polymers and ceramics,[48] modern attempts include more sophisticated approaches like grafting ionic liquids to the surface of inorganic solids or multi-layer electrolyte structures.[49] A unique aspect of hybrid electrolytes is the possibility of constructing nanostructures and tailoring them to have desired properties, which is much more difficult to achieve in a purely polymeric or purely ceramic matrix.[50] Finally, mixtures of liquids and polymers (often named 'polymer gels') can be also considered as hybrid electrolytes (Figure 2).[51–52]

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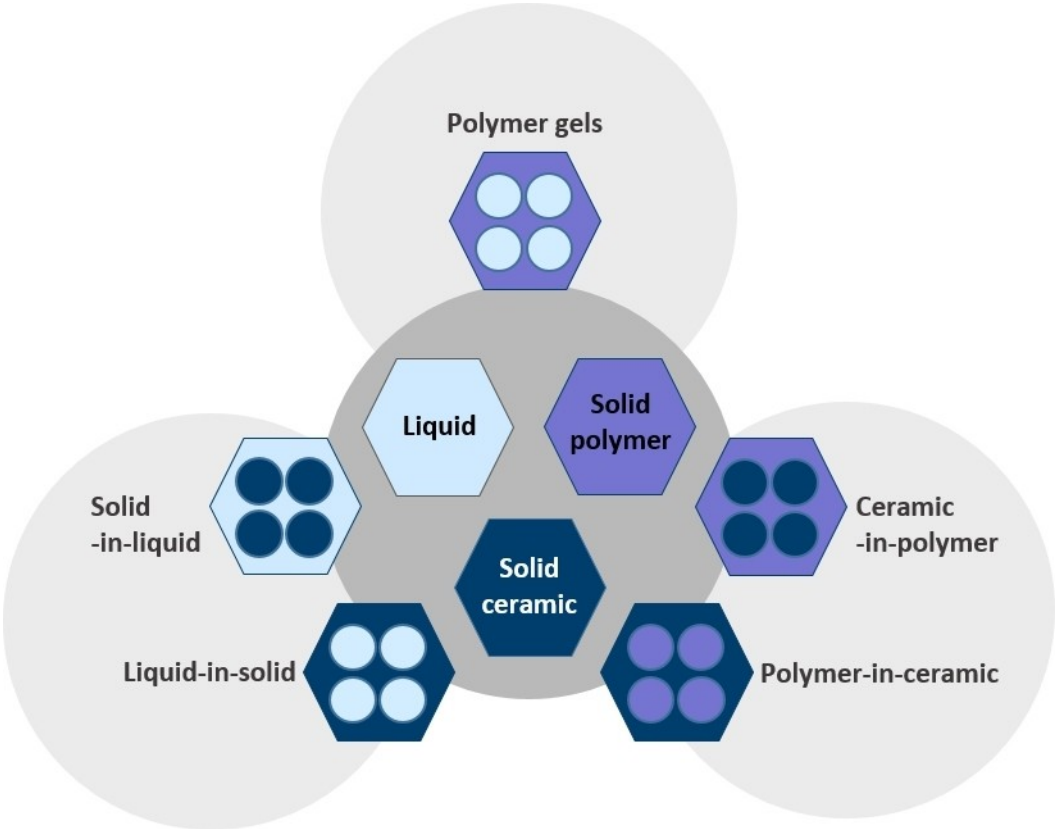

Figure 2. Overview on different types of hybrid electrolytes and schematic representation of their constituents.

3. Open Questions to be Addressed

With all its benefits and drawbacks, the electrolyte plays a key role in every electrochemical storage system and has a decisive influence on overall performance. The chemistry of electrolytes and their interaction with anode and cathode materials is complex and yet not fully understood, thus representing a delicate balance of various properties. The lack of suitable alternative electrolyte components hinders further exploration of novel functional electrolytes for targeted battery cell chemistry and its application(s). For the commercialization of large-scale applications, further advancements in energy and power characteristics must be achieved without sacrificing safety and neglecting the goal of achieving lower costs. Increasing life and energy of lithium-based batteries requires the development of novel and improved electrolyte components as well as a detailed understanding of the stability of them with the respective electrodes, in particular at high voltages. It is common knowledge that when developing new battery chemistries or introducing new functionalities into an existing battery technology, electrode|electrolyte interfaces often hold the key to exploit the full potential of the electrode materials and advance battery chemistry and performance.[53–60] Uncontrolled interfacial reactions may stimulate continuous parasitic reactions, thus limiting the life of the battery. Many of the key impediments to the development of next-generation energy storage systems can be resolved by smart designs of the electrolyte coupled with a smartly engineered electrode (surface) configurations that control interactions and reactions between the electrolyte and electrode. A slight modification of electrolyte as well as the presence of impurities may induce a complete reversal from the ideally controlled scenario to an uncontrolled one, hence driving significant research efforts in studying such interfacial processes. Furthermore, fundamental knowledge on underlying principles dictating the basic function, operation and performance limitations as well as failure of battery systems, is alarmingly required. In this underpinning way, considerable improvements and new concepts concerning electrolyte design can be achieved, thus enabling electrochemical energy storage systems with high energy density, high power, long life and adequate safety at a competitive material and manufacturing cost.

Due to the tremendously increasing research interest in electrochemical energy storage worldwide, the available literature in the field of electrolytes is steadily increasing; this is particularly true for lithium ion conducting electrolytes. Considerable progress and new concepts in respect to the advanced electrolyte formulations and active materials have been achieved leading to electrochemical systems with long life, high energy density, high power and adequate safety at

competitive manufacturing cost.[31,61–62] Manuscripts reporting novel electrolytes and/or advancement of existing formulations are appearing in large numbers literally every day, thus providing further insight into already reported materials. For example, current liquid electrolyte formulations, fulfilling a wide variety of important requirements primarily including a broad electrochemical stability window, a wide liquid range, a high thermal stability, a low vapor pressure, a low viscosity, a high lithium ion conductivity, a high capacity as well as high cycling rates, are result of perennial, however still ongoing research and development processes. In addition to that, different (multi)-functional electrolyte additives have found their applications in advancing liquid electrolyte performance, however not enough is understood about their function and effectiveness. As essential and key component of any electrochemical device, the electrolyte is concomitantly married to 3-D interphases (SEI and CEI) that originate from the high reactivity of the electrodes and the intrinsic instability of electrolyte components. In most cases, electrolyte formulations and their *ad hoc* interfacial chemistries dictate and govern the fate of each electrochemical energy storage device chemistry and its performance. Nonetheless, interphases forming at electrolyte|electrode interfaces still remain the most important but the least understood components in alkali metal/ion batteries, including lithium-based batteries.[63] For this reason, both a fundamental understanding comprising relevant structural/compositional characteristics, chemical/electrochemical reactions as well as thermodynamic/kinetic behavior thereof, hand in hand with practical strategies for enhancing the properties of specifically designed interphases are of paramount importance for enhancement of the overall performance for targeted application.[31,64–67] With this in line, the focus is set on comprehensive understanding of the ionic (and in part electronic) transport phenomena as well as elucidation of the governing mechanisms of charge transport. Due to the complexity involved in interface and interphase behavior, profound research and development require collaborative efforts involving the disciplines of chemistry, physics, materials science, nano-science/nanotechnology, engineering, as well as computational modeling/simulation that will beneficially impact the current lithium ion technologies and future generation solid state lithium and lithium ion batteries, based on ceramic solid electrolytes or polymer electrolytes. This is for instance institutionally realized in Helmholtz-Institute Münster (HI MS) in Germany and Joint Center of Energy Storage Research (JCESR) by DOE Basic Energy Science in USA.[68–69]

To answer the ever-growing scientific questions, various sophisticated *in situ*/operando and *post mortem* methods were developed to investigate the physical and electrochemical properties of electrode and electrolyte materials and their interfaces under defined model conditions or real battery working conditions. A comprehensive systematic and systemic approach allows to understand the structure-reactivity-performance relationships of elected compounds, their impact on the overall electrochemical storage device, and will help to further tailor the properties of electrolyte components for the desired application. In this complex mechanistic game, victory is assured to those approaches which are best able to piece together the gathered conclusions in view of most comprehensive scientific understanding and best capabilities in application.

4. The Rose Garden of the Future

Like roses, opportunities come with a beautiful fragrance, but also with thorns, as the majority of research and development on electrolytes is still based on a trial-and-error approach, in which slight alterations to the basic chemistry rarely offer significant advantage. In this way, development of novel and/or advancement of existing high-performance battery electrolytes calls for considerable effort, expense, and time. Traditionally, it takes many years from initial laboratory steps to commercialization and this is highly dependent on cost reduction by accelerated manufacturing and engineering. The identified necessity for rapid advancement in the fundamental research on battery electrolytes and their interfaces requires joining forces of accelerated experimental and computational methods. Artificial intelligence-orchestrated experimentation and machine learning accelerated computational methods will open new frontiers towards development of highly performant, sustainable and smart electrolyte classes for cost effective, benign and environmentally friendly cell chemistries thus expanding in view of an intensive deployment of lithium-based electrochemical energy storage systems.

In the end, a work of science is truly like a rose. The rose is not beautiful because it is like something else. Neither is a work of science. Roses and scientific work are beautiful in themselves. The uniqueness of roses may be compared with originality and creativity in scientific work.

And: If roses did not suffer thorns on their journey to beauty, they would lose out on becoming masterpieces. After all, one should count the roses and not the thorns.

"*Roses do not bloom hurriedly; for beauty, like any masterpiece, takes time to blossom.*"
– Matshona Dhliwayo, philosopher

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