# An Argument for Basic Battery Science: Each Time an Application Demands a New Battery Chemistry To Achieve Previously Unrealized Functionality, a New Fundamental Understanding Is Required

*Guest Editorial for the Accounts of Chemical Research special issue on "Energy Storage: Complexities Among Materials and Interfaces at Multiple Length Scales".*

Batteries are the critical enabler of potentially new applications of electronics, ranging from an increasingly diverse set of portable applications to stationary grid level energy management. However, due to a paucity of fundamental understanding correlating atomic to mesoscale level phenomena with a full complement of bulk electrochemical properties, present day battery science is not sufficient to provide battery technology with the agility to address the unique set of challenges of power, capacity, capacity retention, safety, and other issues associated with each new potential application. Historically, battery design and engineering involved intuitive strategies of increasing size or component number or adding electronics to the battery. However, this strategy of optimization quickly exhausts any particular battery science, as the numerous documented cases of battery failures in the field attest. It is critical that battery science be explored both fundamentally and with an appreciation for application, to achieve previously unrealized functionality and to enable deliberate design of systems with the desired characteristics. This special issue of Accounts of Chemical Research highlights some of the notable current investigations in basic battery science.

A primary consideration of energy storage devices is energy content per mass or volume. Strategies to extend length of operation without paying a penalty in size or weight are significant research considerations. Metal oxide materials remain in favor due to the availability of high oxidation state metals with little extraneous mass. Multimetal systems such as those incorporating nickel, manganese, and cobalt provide lower cost and

the opportunity to deliver almost double the practical capacity of lithium cobalt oxide alone. Strategies for multielectron transfer at a single metal center are also being explored as a method to increase usable energy content. Detailed studies of materials demonstrate that multiple physiochemical aspects must be considered in addition to bulk structure. For instance, nanosized materials can offer benefit in terms of shortening diffusion lengths as well as mitigating physical degradation of the material due to cracking. Surface characteristics are notable as the composition of a material surface may be different than that of the core, particularly with multimetal structures where segregation can occur. Further, defects influence the electrochemistry where factors such as oxygen vacancies can have profound impact on the observed electrochemical kinetics. While the need to consider and control multiple factors can be considered a complication, it also provides material designers a portfolio of tools that can be used to fine-tune and adapt material properties for specific needs. Even greater gains in energy content can be realized by employing materials that involve phase change or chemical conversion reactions. A notable example is the use of sulfur as an active cathode material with a theoretical energy content ~10 times higher than most insertion materials. In considering the negative electrode, present-day lithium ion batteries use various forms of carbon with a typical capacity of ~360 mAh/g. Next generation negative electrodes under exploration for rechargeable batteries include lithium metal and silicon, which have theoretical capacities approximately 10-fold higher than carbon.

Battery lifetime is an important and challenging area of investigation. Portable electronics demand battery lifetimes of 3 to 5 years; however, batteries for electrified vehicles may demand longevities of 7 to 10 years. Integration with renewable forms of

energy or the power grid would demand lifetimes measured in decades. While several strategies for increased energy content are under exploration, they are often accompanied by negative attributes that lead to compromised cycle or calendar life, motivating investigation into degradation pathways. Thus, the understanding, control, and prediction of degradation and failure mechanisms becomes paramount. Several of the Accounts in this issue delve into understanding the failure mechanisms, developing the theory to explain them, modeling the outcomes, and exploring mitigation approaches. A broad theme that emerges is related to the investigation of interfaces. While interfaces are critical to electrochemical function, the full understanding of interfaces in electrochemical energy storage devices has been elusive. An example is the electrode surface electrolyte interphase called the SEI. While it has been known for many years that the SEI on the negative electrode is needed to protect the electrolyte from continuous degradation, the ability to understand and predict which electrolytes would be stable has been only recently clarified. Interface control via purposeful modification to deliver the desired characteristics is an emerging theme. Approaches include modification of a solid surface directly or tailoring the electrolyte such that the electrode−electrolyte interphase forms and reforms with desirable properties.

Many of the scientific advances that have been achieved are enabled through advancements in characterization method- ology and theory. The ability to probe active electrochemical systems in situ or operando has opened the door to a fuller understanding of the behavior of materials in their native environment, yielding more direct information about the governing mechanisms. Theory and modeling are undertaken in concert with

experiment over multiple length scales where density functional theory is considered in tandem with continuum modeling approaches.

This special issue of Accounts of Chemical Research reflecting recent research themes in energy storage research is particularly timely as it is an exciting and dynamic time. New chemistries are on the horizon to address emerging applications. Pursuit of fundamental understanding remains critically important to effectively design and control materials, interfaces, and their functional properties for energy storage systems in order to meet the needs of the future with high performance, longer lifetimes, and reliable, safe operation.

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